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Exploring the Role of Biodegradable Plastics

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Abstract

The aim of this Ph.D. thesis was to explore the role of biobased, biodegradable plastics in a new and sustainable plastics economy, where plastics serve their useful function without the associated negative externalities. This contributes to understanding and addressing the issue of plastic persistence and accumulation in the environment. A unique perspective is achieved through taking a multidisciplinary approach, with research relating to three different themes (A: material properties, B: environmental impact assessment and C: social attitudes) undertaken (**Figure 1**). These three themes, with their varying objectives, were selected deliberately. Material properties were the initial point of attention as, ultimately, if biodegradable plastics that offer comparable or enhanced material properties compared to conventional plastics cannot be developed, the role they play in the plastics system will be limited. However, throughout the course of the initial research it became clear that to satisfactorily explore the role of biodegradable plastics, the broader system they are used within would also need to be considered. This includes understanding the environmental impact of the materials being investigated as well as understanding how they will be viewed and interacted with in society. Hence, the scope of the research was expanded to include the environmental impact assessment and social attitudes themes.

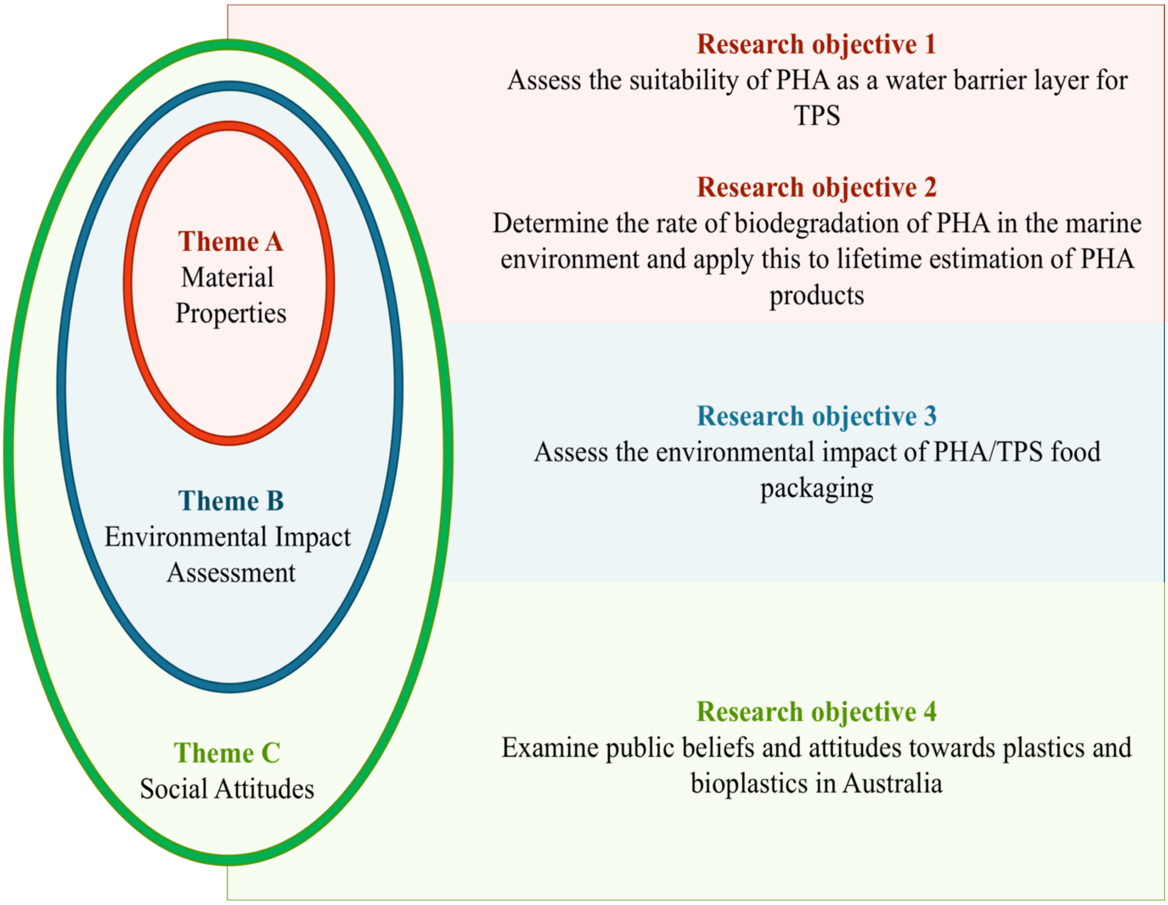

Figure 1: The three levels of investigation forming the research themes of this thesis

The focus on biobased, biodegradable plastics - as opposed to bioplastics in general - was also deliberate. The term ‘bioplastic’ refers to materials that are biobased and/or biodegradable, but not necessarily both, and is thus ambiguous. In regard to reducing the negative externalities associated with plastic persistence in the environment, it is the biobased, biodegradable plastics subset that is of interest. Within this subset, polyhydroxyalkanoates (PHAs) and thermoplastic starch (TPS) were focused on, as they show strong market growth and promising material properties (e.g. marine biodegradability, and good water (PHA) and oxygen (TPS) barrier properties).

Within the material properties theme, the first research activity focused on producing multi-layered PHA-coated TPS materials. The objective was to assess the suitability of PHA as a water barrier layer for TPS. Current multi-layered materials are non-recyclable and problematic from a waste management perspective but are widely used in food packaging due to their high gas-barrier properties. This research explored the possibility of producing biodegradable alternatives. The results were promising, with the PHA layer shown to protect the TPS from moisture absorption, helping the TPS to maintain its good oxygen barrier properties over time. However, poor adhesion between the layers was identified as an issue that would limit the use of the multi-layered material. Preliminary work showed that it may be possible to alleviate this issue by exploiting the differences in the melt viscosities of PHA and TPS to produce a multi-layered material during a single-pass extrusion.

Whilst this research demonstrated the material properties potential of biodegradable plastics, it became apparent that simply demonstrating the feasibility of producing a biodegradable plastic version of a product did not guarantee improvements from a sustainability perspective. Thus, an environmental impact assessment theme was included. The objective was to understand the environmental impact that would be associated with using a PHA/TPS multi-layered material for food packaging. Life-cycle assessment (LCA) methodology was used to compare the multi-layered biodegradable PHA/TPS material to a conventional plastic packaging material. Crucially, food production and wastage were included in the system boundary. The results showed that the impacts of the food contained within the packaging outweighed the impacts of the packaging itself, even if the packaging was biodegradable. This confirmed the importance of taking a systems level view when considering the role of biodegradable plastics - biodegradability isn’t in and of itself a sustainability achievement. In this case, the influence of the packaging material on food waste is the most important attribute.

Unexpectedly, the LCA was not only insightful in what it could tell us, but also what it could not tell us. The LCA was not able to take into consideration many of the important environmental impacts of plastics (e.g. ocean accumulation). It could also not give insight into the likelihood of any of the scenarios explored (e.g. how people will respond to biodegradable materials). As such, the LCA analysis of plastics is necessarily constrained in how it can inform debate and policy. This led to both a second research objective for the material properties theme as well as the inclusion of the social attitudes theme.

The second research objective for the material properties theme was to understand marine biodegradation of PHA, the idea being that if a truly marine biodegradable plastic was identified, it could be used in applications where a high volume of material leaks to the environment, providing considerable benefits compared to a conventional plastic. However, it is currently unclear how long proposed 'marine biodegradable' plastics will persist in the environment and this limits the ability to consider ocean impacts in an environmental impact assessment. A meta-study was performed to determine the rate of biodegradation of PHA in the marine environment, providing clarification as to what 'marine biodegradation of PHA' means in practice. It was estimated that the mean rate of biodegradation of PHA in the marine environment is 0.04 - 0.09 $mg.cm^{-2}.day^{-1}$ (equivalent to a lifetime of 1.5 – 3.5 years for a PHA water bottle). Whilst biodegradation was shown to be occurring, it is slower than would be desirable for preventing ecosystem impacts. Considerable uncertainty also still remains around how different factors influence this rate. This is a topic that requires more targeted attention so as to inform discussion about the benefits and trade-offs of using biodegradable plastics.

However, before time and effort are invested in addressing the detailed material properties questions, understanding of the market potential and social attitudes towards biodegradable plastics needs to be developed. The way plastics are used, and what will be used, is influenced by consumer, industry and government decisions. This was the focus of the social attitudes theme. Using survey methodology, it was found that the Australian public rate plastics in the ocean as the most serious environmental issue (from a list of nine issues), supporting government and industry focus on this topic. Attitudes towards bioplastics were also explored, with the research showing that whilst the public has limited knowledge about bioplastics, they view them positively and would like to see more items made from biodegradable plastics.

As a body of work, having considered three interlinking themes influencing the plastics system, this Ph.D. thesis shows that whilst biodegradable plastics have a role to play in a new and sustainable plastics economy, implementation will require careful consideration. Biodegradable plastics do have promising material properties and there is positive public sentiment towards them, but as LCA shows, biodegradability is not in and of itself a sustainability achievement. Also, the ability to predict lifetimes in the marine (and other) environments of biodegradable plastics such as PHA is still poor, currently limiting their utility in alleviating the impact of leakage to the environment.

Declaration by author

This thesis *is composed of my original work, and contains* no material previously published or written by another person except where due reference has been made in the text. I have clearly stated the contribution by others to jointly-authored works that I have included in my thesis.

I have clearly stated the contribution of others to my thesis as a whole, including statistical assistance, survey design, data analysis, significant technical procedures, professional editorial advice, financial support and any other original research work used or reported in my thesis. The content of my thesis is the result of work I have carried out since the commencement of my higher degree by research candidature and does not include a substantial part of work that has been submitted *to qualify for the award of any* other degree or diploma in any university or other tertiary institution. I have clearly stated which parts of my thesis, if any, have been submitted to qualify for another award.

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Publications included in this thesis

Peer-reviewed papers

Dilkes-Hoffman, L.S., Pratt, S., Lant, P.A., Levett, I., Laycock, B., 2018. Polyhydroxyalkanoate coatings restrict moisture uptake and associated loss of barrier properties of thermoplastic starch films. J. Appl. Polym. Sci. 135, 46379–46387. https://doi.org/10.1002/app.46379

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Contributor	**Statement of contribution**		
	Conception and design of the research	Analysis and interpretation of the data	Drafting and review of the publication
Leela Dilkes-Hoffman (candidate)	80%	80%	80%
Steven Pratt	5%	5%	5%
Paul Lant	5%	5%	5%
Ian Levett	–	5%	5%
Bronwyn Laycock	10%	5%	5%

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Book chapters

Dilkes-Hoffman, L.S., Pratt, S., Lant, P.A., Laycock, B., 2019. The role of biodegradable plastic in solving plastic solid waste accumulation, in: Al-Salem, S.M. (Ed.), Plastics to Energy. Elsevier Inc., London, pp. 469–505. https://doi.org/10.1016/B978-0-12-813140-4.00019-4

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Bronwyn Laycock	5%	5%	5%
Steven Pratt	5%	5%	10%
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Other publications during candidature

Conference abstracts

Dilkes-Hoffman, L.S., Biodegradable food packaging: positive GHG outcomes from reducing food spoilage or improving food waste handling, The University of Queensland EAIT Postgraduate Conference 2017, 6th June 2017, Brisbane, Australia.

Dilkes-Hoffman, L.S., Implications for packaging design as determined in the LCA of a multi-layer, biodegradable food packaging, 12th European Bioplastics Conference, 28th -29th Nov 2017, Berlin, Germany.

Dilkes-Hoffman, L.S., Glass half-full or glass half-empty? Materials innovation and consumers as collaborators to address the issue of plastic pollution, The University of Queensland EAIT Postgraduate Conference 2018, 6th June 2018, Brisbane, Australia. (Awarded best presentation)

Other

Dilkes-Hoffman, L.S., When the trail gets steep, Science (working life). 2019, 363, 662.

Contributions by others to the thesis

Contributions made by co-authors are as described on previous pages.

Statement of parts of the thesis submitted to qualify for the award of another degree

No works submitted towards another degree have been included in this thesis.

Research involving human or animal subjects

Consistent with the requirement for research involving human participants, ethical approval was granted by the University of Queensland's 'Engineering, Architecture and Information Technology, Low and Negligible Risk Ethics Sub-committee' on 27 April 2018, with an amendment approved on 15 May 2018. Copies of the approval letters are included in **Appendix F**. This process ensures that the research complies with the provisions contained in the *National Statement on Ethical Conduct in Human Research* and complies with the regulations governing experimentation on humans.

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plastic, bioplastic, biodegradable plastic, polyhydroxyalkanoate, thermoplastic starch, marine biodegradation, life-cycle assessment, public attitudes, sustainability, environment

Australian and New Zealand Standard Research Classifications (ANZSRC)

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ANZSRC code: 050204, Environmental Impact Assessment, 20%

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Fields of Research (FoR) Classification

FoR code: 0912, Materials Engineering, 80%

FoR code: 0502, Environmental Science and Management, 20%

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List of abbreviations

AD	Anaerobic digestion
ATR-FTIR	Attenuated total reflectance-Fourier transform infrared spectroscopy
BioPE	Biobased polyethylene
CO_2e	CO_2 equivalent
EOL	End-of-life
EU	European Union
GDP	Gross domestic product
GHG	Greenhouse gas
GWP	Global warming potential
HDPE	High-density polyethylene
HV	Hydroxyvalerate
KMO	Kaiser-Meyer-Olkin statistic
LCA	Life-cycle assessment
LDPE	Low-density polyethylene
MC	Moisture content
MP	Melt-press
OECD	The Organization for Economic Co-operation and Development
PCL	Polycaprolactone
PE	Polyethylene
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoate
PHB	Polyhydroxybutyrate
PHBV	Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate)
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinylchloride
RH	Relative humidity
RO	Research objective
SPSS	Statistical package for the social sciences
TPS	Thermoplastic starch
UK	United Kingdom
USA	United States of America
UV	Ultraviolet
WRAP	Waste Resource Action Program

CHAPTER 1 Introduction and thesis outline

1.1 Background

Since large-scale plastic production commenced in the 1950s, it is estimated that 8.3 billion tonnes of plastic has been produced (Geyer et al., 2017). This cumulative total is increasing at an exponential rate (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). On a global scale we do not have the ability to effectively re-process most of this plastic, so the majority is still present in one form or another (Geyer et al., 2017; Thompson et al., 2009). In fact, the ubiquity of plastic waste on every surface of the Earth has led to the suggestion that it could be considered as a geological indicator of the Anthropocene epoch (Zalasiewicz et al., 2016).

Waste management systems have improved in the past few decades and this has expanded the available end-of-life options for plastic and improved collection rates (Geyer et al., 2017). However, the final destination of many of the plastic products produced each year is still uncertain, especially in developing countries (Hoornweg and Bhada-Tata, 2012). This is due to a variety of factors, including a lack of world-wide data, few formal collection systems in many places, and unreported waste disposal, including illegal dumping and uncontrolled burning (Geyer et al., 2017; Hoornweg and Bhada-Tata, 2012; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). Our attempt to understand where plastics that are produced today will be found in 20 years shows that a majority will have been discarded (and this includes all packaging) (**Figure 1-1**). Whilst approximately one-third of plastics will be recycled (mainly down-cycled) or incinerated, the vast majority will end up in a landfill, and a smaller but significant volume will end up as unmanaged waste/litter which can then enter the oceans. In particular, it is packaging that is the largest source of litter and ocean plastic. Once plastic enters the oceans it is very hard to remove, and so consequently it accumulates (Jambeck et al., 2015; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

This is cause for concern. Plastic persistence and accumulation in the oceans (both micro and macro) harms marine life and has been linked to the transfer of pollutants and invasive organisms (Law, 2017; Thompson et al., 2009). The problems associated with marine plastic accumulation are also likely to get worse as solid waste generation is increasing rapidly without a corresponding improvement in the infrastructure to manage it (Hoornweg and Bhada-Tata, 2012; Hoornweg and

Thomas, 1999; van Beukering et al., 1999). If current plastic production and waste management trends continue, it is predicted that by 2050, 12 billion tonnes of plastic waste will be in landfills or the natural environment (Geyer et al., 2017) and there may be more plastic than fish (by mass) in the ocean (Jambeck et al., 2015; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

Developing a sustainable plastics economy, where the negative externalities associated with plastic persistence and accumulation are reduced, is of urgent importance.

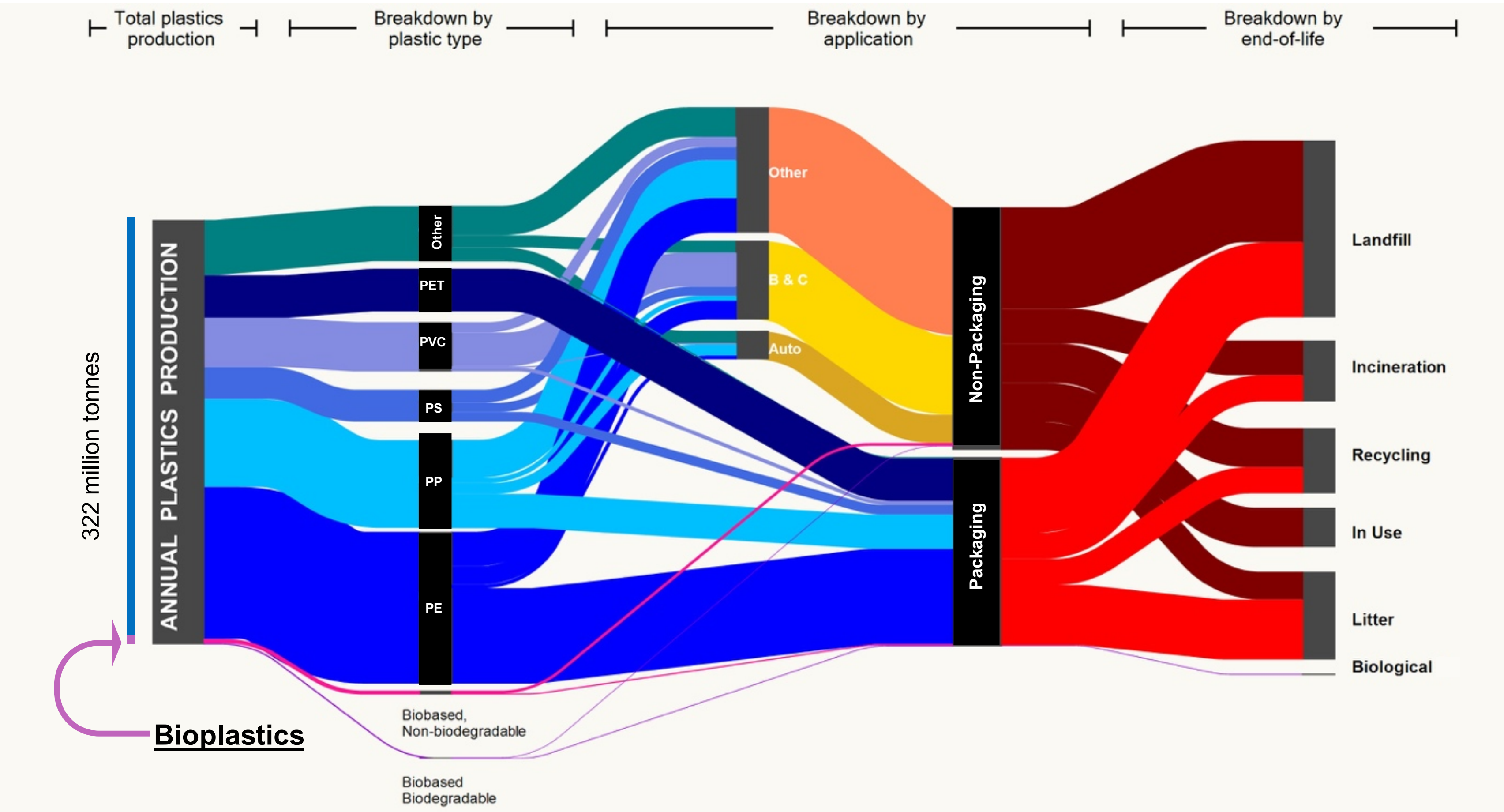


Figure 1-1: Predicted destination of all the plastics produced in 2015 in the year 2035 (plastic fibres not included)

Note to Figure 1-1: Polymer type breakdown based on Plastics Europe (2016). End-of-life destinations based on literature estimations (Geyer et al., 2017; Hoornweg and Bhada-Tata, 2012; Jambeck et al., 2015; PlasticsEurope, 2016; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). 'Long-term use' is taken to mean longer than a 20-year lifetime. Litter includes all directly littered items as well as mismanagement and illegal dumping of waste. It should be noted that it is hard to draw a definitive distinction between a poorly managed landfill and litter. Data collated by L. S. Dilkes-Hoffman. For details of mass flows see Appendix G.

1.2 Research evolution

Given the nature of the problem just outlined, when I decided to start my Ph.D. it seemed obvious to me that I wanted to work in the area of materials engineering, focusing on biodegradable plastics. I thought: if we could develop plastics that would biodegrade under the right conditions but provide the same material properties as our current plastics, wouldn't the issue of plastic pollution basically be solved? However, I soon realised that solving the waste and plastic crisis is far more complex than just developing good biodegradable plastics. In fact, their role in the plastics system remains to be understood, and materials development is only a small part of what needs to be investigated in order to ensure that biodegradable plastics can deliver sustainability outcomes. Even if from a materials property perspective a 'perfect' biodegradable plastic was developed tomorrow, its utility would be governed by economic factors, accessibility of waste collection and processing facilities for biodegradable materials, as well as public opinion and industry acceptance – and this is not an exhaustive list. I realised that the useful role and superior sustainability of biodegradable plastics compared to conventional plastics cannot be taken as a given and needs to be critically investigated. My evolution in thinking is reflected throughout this Ph.D. thesis and in the thesis outline a timeline is provided to set each piece of research in context (Section 1.4, **Figure 1-3**).

1.3 Aim and objectives

The aim of this Ph.D. thesis was to explore the role of biobased, biodegradable plastics in a new and sustainable plastics economy, where plastics serve their useful function without the associated negative externalities. A unique perspective is achieved through taking a multidisciplinary approach, with research relating to the three different themes (A: Material properties, B: Environmental impact assessment and C: Social attitudes) undertaken (**Figure 1-2**). These three themes, with their varying objectives, were selected deliberately in recognition of the fact that to satisfactorily explore the role of biodegradable plastics, the broader system they are used within needs to be considered.

The focus on biobased, biodegradable plastics as opposed to bioplastics in general was also deliberate. The term 'bioplastic' refers to materials that are biobased and/or biodegradable, but not necessarily both and is thus relatively ambiguous. In regard to creating a sustainable plastic system, specifically focusing on reducing the negative externalities associated with plastic accumulation and persistence in the environment, it is the biobased, biodegradable plastics subset that is of interest. As promising biodegradable plastics with good material properties (e.g. marine biodegradable (Deroiné et al., 2014b), good water and gas barrier properties (Halley and Averous, 2014; Shogren, 1997)) and strong market growth (European Bioplastics, 2018) polyhydroxyalkanoates (PHA) and thermoplastic starch (TPS) were focused on as the biodegradable plastics of interest.

1.3.1 *Theme A – Material Properties*

Research objective 1*: Assess the suitability of PHA as a water barrier layer for TPS.*

The material properties theme was included as, ultimately, if we cannot develop biodegradable plastics that offer comparable or enhanced material properties compared to conventional plastics, the role they will play in the plastics system will be limited.

This is particularly relevant for plastic food packaging. Packaging is the highest application sector for plastics in all countries. In Europe, packaging comprises 40% of plastics demand, whilst in the US packaging is 43% of demand (Germany Trade and Invest, 2016; PlasticsEurope, 2017). The worldwide conservative estimate is that 26% of all plastics are used in packaging applications (World Economic Forum and Ellen MacArthur Foundation, 2017). In certain applications, elimination or substitution of the packaging may be desirable and relatively easy. However, food packaging is not such a case. The benefit of food packaging in regard to the reduction of food waste, often justifies the use of the packaging (Williams and Wikström, 2011), meaning that better design, as opposed to elimination, needs to be considered. One of the ways in which packaging prevents food waste is through limiting water and gas transfer – in particular, reducing the amount of oxygen that enters a package (Dave and Ghaly, 2011). New materials, including biodegradable materials, will need to have good barrier properties in order to be considered as suitable alternatives to conventional packaging materials. The material properties research was undertaken in order to produce and test PHA-coated TPS materials as examples of biodegradable food packaging materials with high oxygen (and water) barrier properties.

Research objective 2*: Determine the rate of biodegradation of PHA in the marine environment and apply this to lifetime estimation of PHA products.*

Another desirable property of biodegradable plastics, that can position them well against conventional plastics, is marine biodegradability. If a truly marine biodegradable plastic was identified, it could be used in high leakage applications, providing considerable benefits compared to a conventional plastic. However, it is currently unclear what the rate of biodegradation of biodegradable polymers is *in situ* in the marine environment, and what lifetimes can be expected. Before possible applications of marine biodegradable plastics are seriously considered, their rate of biodegradation needs to be understood. A meta-study was undertaken as part of the material properties theme, aiming to understand the rate of biodegradation of PHA in the marine environment. Due to a lack of suitable references, a similar study could not be undertaken for TPS at this point, but it is hoped that similar methodology will be able to be applied for TPS in the future.

1.3.2 *Theme B – Environmental Impact Assessment*

Research objective 3*: Determine the environmental impact of PHA/TPS food packaging.*

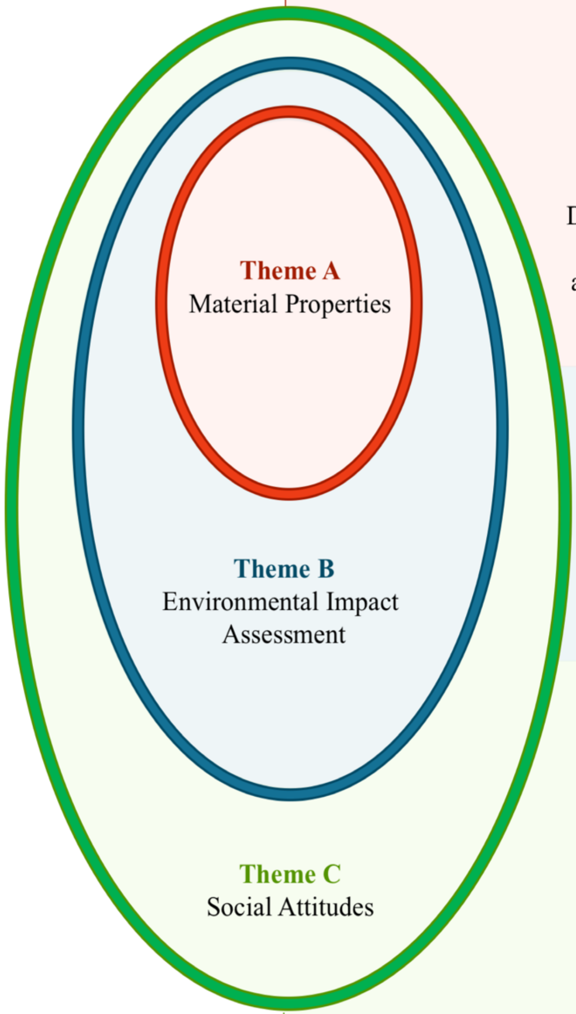
Environmental impact assessment can be used to determine the environmental trade-offs associated with switching from a conventional plastic to a biodegradable plastic, and to identify which design characteristics have the greatest influence over these. It is a bridge between the materials level and the social level, as whilst the properties of a material influence its environmental performance, the impact of socially controlled factors (such as whether a material will be disposed of in composting or landfill) can also start to be accounted for through scenario testing.

A life-cycle assessment for PHA/TPS food packaging (focused on in the material properties theme) versus conventional packaging was undertaken. Food waste was included in the system boundary given that literature has shown that food waste plays an important role in determining the overall footprint of a packaging material (Williams and Wikström, 2011).

1.3.3 *Theme C – Social Attitudes*

Research objective 4*: Examine public beliefs and attitudes towards plastics and bioplastics in Australia.*

A material's acceptance and utility are governed not only by material properties but also by social forces. A large portion of plastic is used in consumer applications (PlasticsEurope, 2017), meaning it is the public that will need to interact with any changes in the plastics system and are often responsible for disposing of plastic waste. It is thus important to understand the social context for the topic of plastics (Gelcich et al., 2014; Pahl et al., 2017) and connect materials engineering research to an understanding of how the materials are going to be perceived and used once on the market. Using survey methodology, this section of the research explores the knowledge and attitudes of the Australian public towards plastics and bioplastics. Questions relating to the subset of biodegradable plastics were asked after initial impressions of bioplastics had been collected, so that understanding and expectations of the term 'bioplastics' could be investigated.



Research objectives	Methodology	Output
Research objective 1 Assess the suitability of PHA as a water barrier layer for TPS	**Laboratory techniques** • Extrusion, melt-press and solvent casting used to produce multilayered PHA and starch materials with a specially designed die. • Testing of oxygen permeability and moisture diffusion.	Published research paper (As Chapter 3) + Chapter 4
Research objective 2 Determine the rate of biodegradation of PHA in the marine environment and apply this to lifetime estimation of PHA products	**Literature review and statistical analysis** • Data normalised to a rate of PHA biodegradation based on initial surface area for all studies. • Lifetime predictions based on 95% confidence interval.	Published research paper (As Chapter 5)
Research objective 3 Assess the environmental impact of PHA/TPS food packaging	**Life-cycle Assessment** • A comparison between a PHA coated TPS food packaging material and a polypropylene food packaging. • Global warming focused on as the impact factor. Food wastage included in the system boundary.	Published research paper (As Chapter 6)
Research objective 4 Examine public beliefs and attitudes towards plastics and bioplastics in Australia	**Public Survey** • Survey developed by the research team and then administered through a market research company with 2,518 responses collected from a nationally representative sample of the Australian public. • Inspiration for the survey questions taken from a variety of periodical environmental surveys.	Published research paper (As Chapter 7) Submitted research paper (As Chapter 8)

Figure 1-2: Thesis outline

1.4 Thesis in outline

This Ph.D. thesis is based on work that was published throughout the candidature. The works are included as published at the time, as such, some evolution of thought is evident within them. To place each of the presented chapters in context, a timeline of the research is presented in **Figure 1-3**. Within each chapter the relevant literature review and methodology is presented.

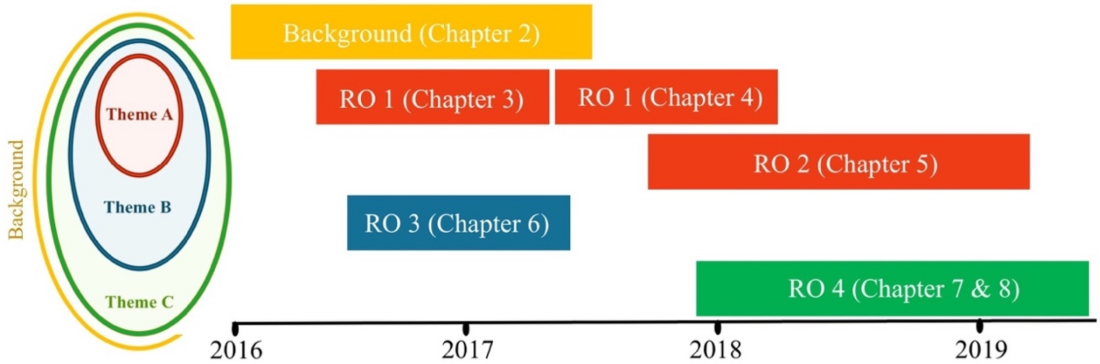


Figure 1-3: Timeline of the research presented

This thesis is divided into nine chapters, with six results chapters (**Chapters 3 - 8**) addressing the four research objectives. **Chapter 1** and **Chapter 2** present the background and motivation of the presented research, setting the scene for the subsequent research chapters. **Chapter 2** was published as a book chapter (Dilkes-Hoffman et al., 2019b).

As part of the material properties theme, **Chapters 3** and **4** present the results of experimental work considering the suitability of PHA as a water barrier layer for TPS, with **Chapter 3** published in Dilkes-Hoffman et al. (2018b). Also under the material properties theme, **Chapter 5** presents the results of a meta-study on the rate of biodegradation of PHA in the marine environment, the results of which are published in Dilkes-Hoffman et al. (2019a).

For the environmental impact assessment theme, **Chapter 6** presents the results of a life-cycle assessment which calculates the environmental impact of PHA/TPS food packaging. This is published in Dilkes-Hoffman et al. (2018a).

Chapter 7 and **8** address the social attitudes theme, using survey methodology to explore the public's attitudes towards plastics and bioplastics. **Chapter 7** presents the results relating to plastics in general, and has been published as Dilkes-Hoffman et al. (2019c). **Chapter 8** presents the results that relate to bioplastics and has been submitted to the journal Resources, Conservation and Recycling.

Finally, **Chapter 9** summarises the major results presented in this Ph.D. thesis and provides recommendations for future research.

CHAPTER 2 Setting the scene

Chapter summary

This chapter presents the current issues associated with conventional plastic use and disposal and introduces biodegradable plastics as alternative materials. It serves to set the research objectives pursued in this Ph.D. thesis in the context of current global trends. It is not intended as an extensive literature review, with each chapter containing its own targeted literature review.

In summary, the growth in the use of plastics has led to the issue of plastic waste accumulation. Close to 8 billion tonnes of plastic has been produced in just 60 years, and this plastic waste is accumulating both on land and in the oceans, with associated negative effects. In response, there is increasing discussion around the relative benefits and drawbacks of the variety of proposed "solutions". Biodegradation is often included in these discussions.

This chapter is based on a modified version of a book chapter published during the candidature.

Dilkes-Hoffman, L.S., Pratt, S., Lant, P.A., Laycock, B., 2019. The Role of Biodegradable Plastic in Solving Plastic Solid Waste Accumulation, in: Al-Salem, S.M. (Ed.), Plastics to Energy. Elsevier Inc., London, pp. 469–505. https://doi.org/10.1016/B978-0-12-813140-4.00019-4

2.1 Section 1: What is the issue?

Before we can consider the role of biodegradable plastics in a new and sustainable plastics economy, the current situation must be understood. Thus, this section sets out the current issues associated with conventional plastic use and disposal. In summary (with details of each of these statements discussed throughout sections 2.2 – 2.6), close to 8 billion tons of plastic have been produced in just 60 years, and this production rate is exponentially increasing. Almost all of this plastic is still present in one form or another, because we do not, on a global scale, have sufficient mechanisms for managing the waste effectively. This means that plastic waste is accumulating both on land and in the oceans and this has associated negative effects. In developed countries, plastic waste (that is collected) is still often disposed of in landfills, thereby requiring the use and contamination of valuable space. In developing countries, plastic waste is less well managed. It is typically collected in open dumps, burned, or littered. This leads to health concerns in the case of unregulated burning, and pollution as unsecured and littered material is washed into waterways and eventually oceans. Plastic accumulation in the oceans (both micro and macro) is not linked to the country of origin of the waste, traveling across the globe as dictated by ocean currents. It harms marine life and has been linked to the transfer of pollutants (which absorb into the plastic structure) and organisms (which adhere to the plastics surface).

2.2 Background: non-biodegradable plastics

The term plastic is used to describe (often synthetic) materials formed from organic polymeric substances of large molecular weight. Plastics are solid in their finished state, but during at least one stage of manufacture and/or processing are malleable and mouldable. They are commonly derived from petrochemicals and approximately 5 - 8% of world's oil is currently used for plastic production (and this is predicted to increase as the demand for plastic rises) (PlasticsEurope; Consultic and myCeppi, 2016; WEF; Ellen MacArthur Foundation and McKinsey & Co, 2016). Hundreds of different plastic materials are commercially available, but there are six dominant types that account for around 70-90% of total demand. These are low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS) and polyethylene terephthalate (PET) (Andrady and Neal, 2009; Storz and Vorlop, 2013).

The most common applications for each of the polymers are (Andrady and Neal, 2009; PlasticsEurope, 2016):

PE – Plastic packaging films, carrier bags, liquid containers (such as milk bottles), pipes, mulch films, insulators for electrical devices and toys.

PP – Flexible barrier film pouches, bottles, containers, cutlery, window and door frames, pipes and automotive parts.

PS – Expanded foam food and electrical packaging, hard food and electrical packaging and insulation.

PVC – Building components (window frames, floor coverings), upholstery, cling films, pipes and hoses.

PET – Bottles and some fibres.

Plastics are used in almost every aspect of modern life - including in the food system, building and construction, the medical and health product industry, automotive and aeronautics, clothing and electronics (PlasticsEurope, 2016). In the USA and Western Europe 100-140 kg of plastic per person is consumed per year. In Asia the use is still around 20 kg per person per year, but this is expected to rapidly grow (Germany Trade and Invest, 2016; Gourmelon, 2015).

Packaging is the highest application sector for plastics in all countries. In Europe packaging is 39.9% of demand, whilst in the USA packaging is 43% of demand (Germany Trade and Invest, 2016; Gourmelon, 2015; PlasticsEurope, 2016). The worldwide conservative estimate is that 26% of all plastics are used in packaging applications (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

2.3 Environmental impacts of plastic production, use, and disposal

The use of plastics has undeniable benefits for humanity and their utility is reflected in both the speed at which they were adopted and the vast range of applications they are now used in. Their positive characteristics include the fact that they are lightweight, easily mouldable, as tough or flexible as desired, easy to colour, transparent, water resistant, and cheap to produce. The variety of ways in which these characteristics provide benefits are broad and undeniable.

Thus, when it comes to considering the environmental impact of plastic use, the results are not black-and-white, with both positive and negative outcomes. Positive outcomes are often observed during

the production and use phase, whilst the majority of the negative outcomes are associated with disposal.
In terms of the production of plastic, as with every material, there is an associated greenhouse gas footprint, but the 'per functional unit' footprint may be less than that of alternatives. It is tricky to know exactly how to interpret this outcome though, as how much of a benefit this gives plastic versus other materials is highly dependent on how the impacts of the production are distributed across a product's life-cycle. For example, one of the most recent studies comparing re-usable ceramic cups and disposable plastic cups found that on a 'single-use basis' the plastic cup did have a lower environmental footprint, but once the ceramic cup is reused 60-130 times (and the impact of production is distributed over multiple uses), it then has the better environmental profile (Woods and Bakshi, 2014). Factors such as the efficiency of washing processes, and proportion of energy derived from renewable resources, will also influence this comparison and change the number of reuses required before a re-usable object has a better environmental profile than a single-use plastic object (despite an initially higher footprint of production). Thus, claims of sustainability based on production footprint are hard to interpret.

Perhaps counter-intuitively, where plastics can have clear positive environmental outcomes is during their use phase, due to both their properties and resource efficiency. A joint report by the American Chemical Council and Trucost calculates that there would be greater environmental consequences (up to 3.8 times increase for consumer goods) if plastic materials were replaced with alternative materials (e.g. wood, glass, metal) (ACC and Trucost, 2016). This would mainly be due to the large increase in the volume and weight of materials that would be associated with using the alternatives. A good example of this is the replacement of metal with plastic for many vehicle parts, which reduces the weight of the vehicle and thus reduces fuel usage for transport (ACC and Trucost, 2016; Andrady and Neal, 2009). Similarly, plastics have proven environmental benefits in the area of food packaging where their use contributes to the reduction of food wastage (Barlow and Morgan, 2013; Lindh et al., 2016; Wikström et al., 2016), and in building design where their use reduces energy consumption by presenting a cheap, effective option for building insulation (Andrady and Neal, 2009; PlasticsEurope, 2016). There is a link between plastic use and negative human health effects but these are not extensively considered in this Ph.D. thesis and for a discussion on this topic readers are advised to look at reviews by Andrady and Neal (2009), Thompson et al. (2009), Talsness et al. (2009) and Hauser and Calafat (2005).

The major environmental burden and concern associated with plastics is in their end-of-life phase. However, this has been neglected in many quantitative studies because the burden is difficult to quantify through a conventional life-cycle assessment (Tolinksi, 2011). One of the major issues is plastic accumulation, which occurs due to the durability of plastics in landfill and the environment (and the large amount of waste which is mismanaged) (ACC and Trucost, 2016). The impacts of accumulation can be separated into two distinct issues relating to plastics in landfills and plastics as litter in the ocean. These are explored further in the following section.

2.4 **Plastic accumulation**

The production rate of plastic materials has increased over 20-fold since they first started being used on a large scale in the 1960s (as illustrated in **Figure 2-1**) (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). It is estimated that a cumulative total of 8 billion tonnes of plastic has been produced over this time (Geyer et al., 2017).

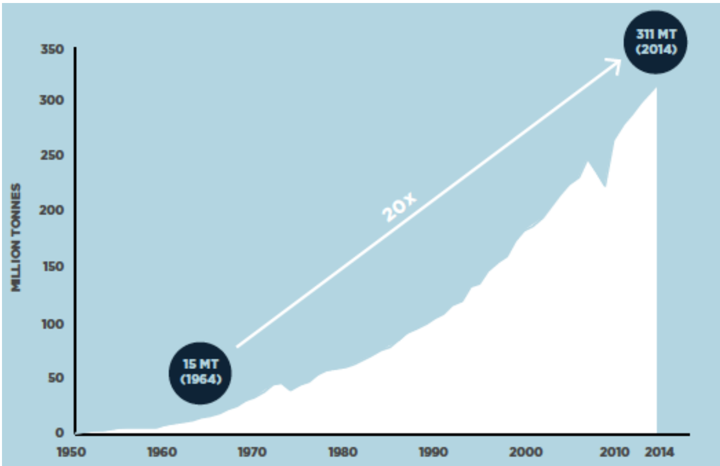


Figure 2-1: Growth in global plastics production per year

Note to Figure 2-1: Figure reproduced from World Economic Forum; Ellen MacArthur Foundation and McKinsey & Company (2016).

Due to their durability, unless incinerated or recycled, all of the plastics that have ever been produced are still present in the environment, whether in use, as litter, or as components of landfill (Geyer et al., 2017). In an ideal system, all plastics would be collected and then reused or separated and recycled to produce products of equivalent functionality (closed-loop) (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). This would drastically reduce requirements for virgin plastic production and eliminate the concept of waste. However, the reality is that on many local scales, let alone on a worldwide scale, the recovery and recycling of plastics is very low and isn't anywhere near keeping pace with plastic production.

In regard to the recovery of waste, the World Bank (Hoornweg and Bhada-Tata, 2012) estimates that in high-income countries close to 98% of waste is collected. However, in low-income countries this drops to only 41% being collected, with the remainder instead burned in the open or littered (which has pollution and health implications) (Gourmelon, 2015; Hoornweg and Bhada-Tata, 2012).

Obviously, low collection rates influence the opportunities for recycling, but even in countries with high collection rates, recycling rates are currently still low (ACC and Trucost, 2016; EPA, 2015; PlasticsEurope, 2016). For example, packaging (which has the highest recycling rate of all plastic products) is only recycled at a level of 24 - 42% in most high-income countries and worldwide plastic packaging recycling rates sit far below this at only 14% (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

Figure 2-2 shows our understanding of the current material flows of plastic, from production to final destination. This figure highlights that whilst there is uncertainty regarding the final destination of all plastics, in the current system the majority of plastics will be disposed of in landfill. A large proportion will also escape into the environment with a small but significant portion of this entering the oceans. In particular, it is packaging that is the largest source of litter and ocean plastic (Jambeck et al., 2015; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

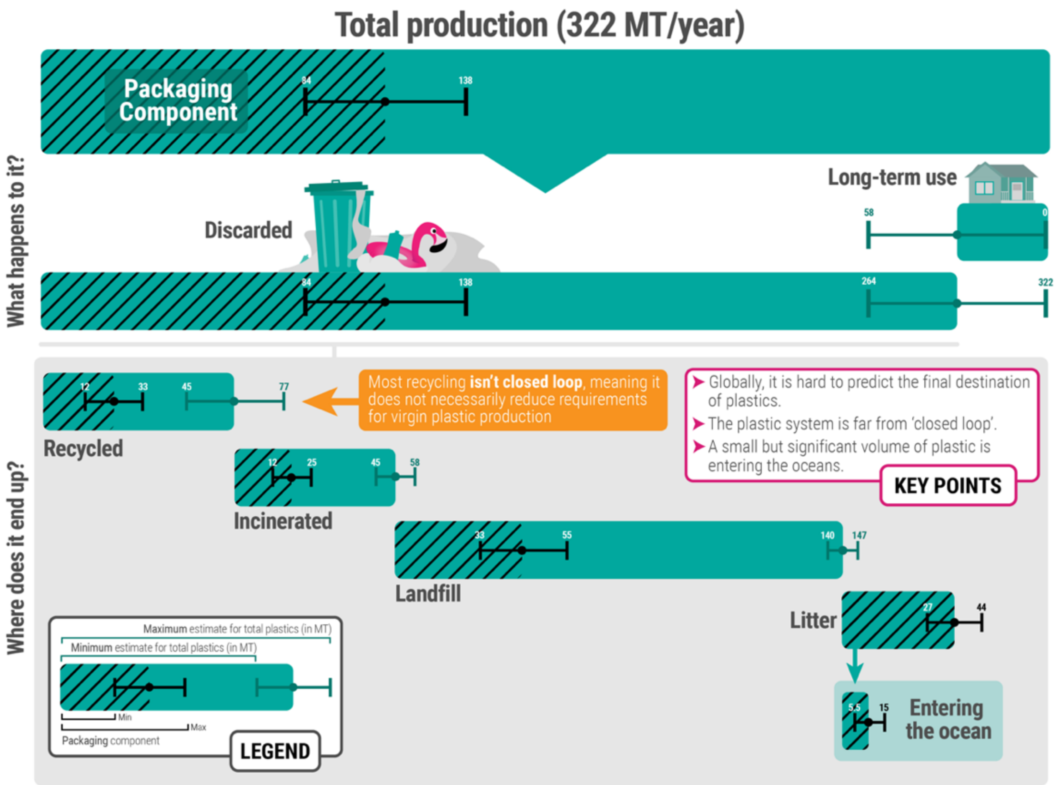


Figure 2-2: Predicted destination in the year 2035 for all plastics produced in 2015 (plastic fibres not included)

Note to Figure 2-2: End-of-of life destinations are based on literature estimations (Geyer et al., 2017; Hoornweg and Bhada-Tata, 2012; Jambeck et al., 2015; PlasticsEurope, 2016; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). 'Long-term use' is taken to mean longer than a 20-year lifetime. Litter includes all directly littered items as well as mismanagement and illegal dumping of waste. It is hard to draw a definitive distinction between a poorly managed landfill and litter.

2.4.1 *Accumulation in landfills*

There are major economic and social barriers to efficient recycling and reuse. These relate to the high price of recycled polymer versus virgin polymer, the cost of recycling versus alternative forms of disposal and the difficulty in ensuring efficient collection and sorting of waste materials (Hopewell et al., 2009). This means that by conservative estimates 22 - 43% of plastics end up disposed of in landfills (often the cheapest method of disposal) worldwide (See **Figure 2-2**) (Gourmelon, 2015; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

Even in developed countries, a large proportion of plastics are landfilled. For example, in the USA, most of the plastics that are not recycled (~90%) are sent to landfill (EPA, 2015), and in the EU landfilling rates are around 31% (PlasticsEurope, 2016). At least in these higher income countries, the landfills are well regulated and maintained, however, in lower and middle-income countries, open-dumps and open-landfills are the most common methods of waste disposal which can then result in leakage to the environment (Hoornweg and Bhada-Tata, 2012).

Unfortunately, it is predicted that the volume of plastic that is mismanaged or disposed of in landfills will actually rise in the coming years, as developing nations shift to a western consumption pattern of plastic use without the infrastructure to manage it appropriately at the end-of-life (Hoornweg et al., 2013; Hoornweg and Thomas, 1999; van Beukering et al., 1999). A city resident produces around four times as much waste as a rural resident, and this waste is more likely to contain a high proportion of plastics. As urbanisation is increasing, global solid waste generation is increasing (as illustrated in **Figure 2-3**) (Hoornweg et al., 2013; Hoornweg and Bhada-Tata, 2012) and a large volume of this will be directed to landfill.

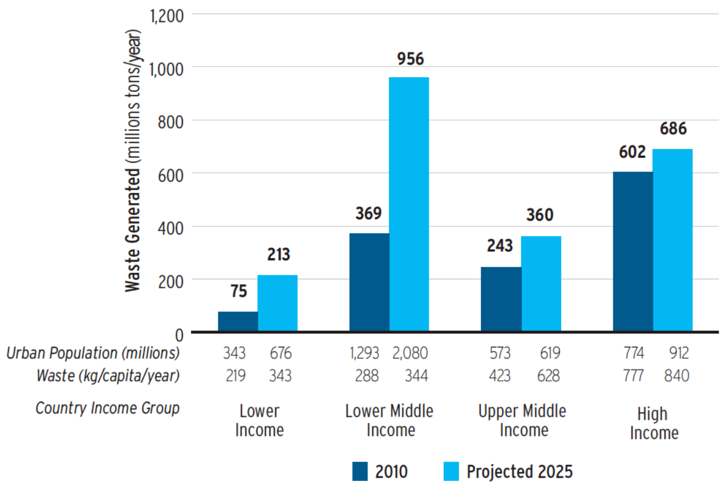


Figure 2-3: Waste generation by income group (2010 to 2025)

Note to Figure 2-3: Figure reproduced from a report for the World Bank (Hoornweg and Bhada-Tata, 2012).

One way to reduce the amount of waste going to landfill is through legislation and it is known that countries with a landfill ban achieve higher rates of recycling (PlasticsEurope, 2016). The World Packaging Organisation has also tried to alleviate the amount of plastic going to landfill through down-gauging and light-weighting of packaging materials (World Packaging Organisation, 2008). However, light-weighting is not a long-term solution, and even if perfect collection, recycling and reuse systems were to be implemented based on landfill bans, it is estimated that 30% of plastic packaging will never be eligible for recycling without fundamental redesign (for example, organically soiled packaging, multi-layer packaging and items such as small sachets that are physically or impractically recyclable) (Gross and Kalra, 2002; World Economic Forum and Ellen MacArthur Foundation, 2017). Thus, a certain level of dependence on incineration or landfill is currently inevitable (World Economic Forum and Ellen MacArthur Foundation, 2017). As such, preventing accumulation of plastics in landfills will require more than just regulation to reduce landfill disposal, with a clear need to focus on producing plastics that are themselves more sustainable by design (ACC and Trucost, 2016; Hopewell et al., 2009; World Economic Forum and Ellen MacArthur Foundation, 2017).

2.4.2 *Accumulation in the oceans*

The quantity of plastics in the marine environment is substantial but the exact amount and relative proportions of different plastic types in the ocean remain largely unknown. This is due to a lack of reliable information on the sources, originating sectors and users (Andrady, 2015; Thompson, 2015; UNEP, 2016).

However, recently Jambeck et al. (2015) attempted to model the inputs of plastic waste to the marine environments for 192 coastal countries, considering plastic waste mismanagement and direct litter. Their results for the distribution of plastic waste which is produced and mismanaged by country are presented in **Figure 2-4**. Population size and quality of waste management systems determine which countries contribute the greatest portion of plastics to marine litter with over 50% of the plastic waste entering the oceans predicted to originate in just five South-East Asian countries (Jambeck et al., 2015). Even their most conservative estimates predicted an input of 100 million tonnes of plastic waste to the oceans over the 15-year time period 2010-25 or around 1.5 - 4.1% of total plastic production entering the ocean per year (see **Figure 2-2**). This input is predicted to increase by an order of magnitude by 2025 (Jambeck et al., 2015) and by 2050 there are reports that the oceans could contain more plastics than fish by weight (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

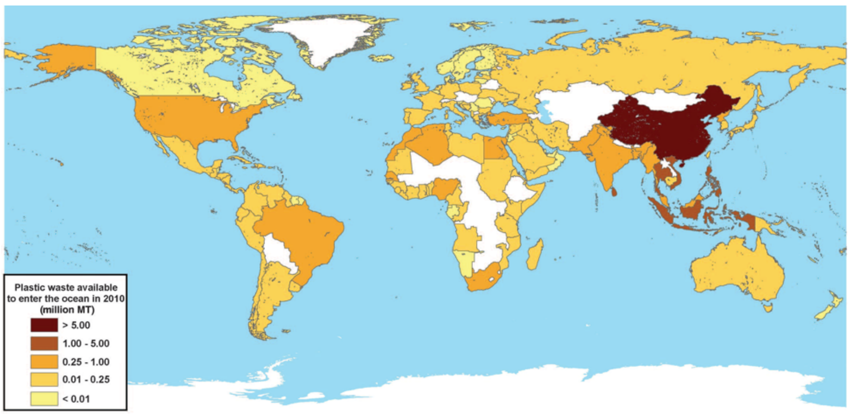


Figure 2-4: Estimated plastic waste produced and mismanaged by country for populations living within 50 km of the coast

Note to Figure 2-4: Figure reproduced from Jambeck et al. (2015).

When discussing plastic accumulation in the oceans there are two main groups of plastics referred to: macroplastics (which are solid articles such as bags, packaging, cigarette lighters etc.) and microplastics. Microplastics are small fragments of plastic debris and result from either direct release of small plastic particles (plastic pellets and powders), or the fragmentation of larger items (under conditions of UV, heat and physical action in the environment (Andrady, 2015)) or through everyday usage such as clothes washing (Browne et al., 2011; Hartline et al., 2016). Their presence has been documented from the poles to the equator (Thompson, 2015).

Whilst Jambeck et al. (2015) considered macroplastics, in regard to microplastics, it is estimated that in the USA alone 100 tonnes of microplastics directly enter the oceans annually, mainly through wastewater streams (Thompson, 2015). At least similar volumes would be expected from other developed nations. Although the measured volume may appear to be small compared to the scale of annual plastic flows (see **Figure 2-2**), microplastics have the potential to be a significant issue as they affect animals (such as fish) which are otherwise relatively unaffected by large plastic items and can accumulate up the food chain (Thompson, 2015).

In regard to the types of products most likely to enter the oceans, typically 40 - 80% of marine waste items are plastic, and these items are often light-weight, single use and can be linked to the food industry (Barnes et al., 2009). This is evident from records of coastal clean-ups as presented in **Table 2-1** (Andrady, 2015). The plastics most commonly associated with litter include the main polymer types as well as cellulose acetate, which although having low production levels is used in cigarette filters that probably enter the oceans due to their abundance and small size. This highlights the fact that light-weight, single use materials need to be the targets when attempting to address ocean litter. The main entry points for these plastics are wastewater discharge points, rivers and coastal areas. Other sources of marine plastics such as new agricultural products (e.g. polymer encapsulation for slow release fertilisers) have been identified, however their contribution to the problem is currently unknown (UNEP, 2016).

Table 2-1: Marine debris items removed from global coastlines and waterways during the 2009 international coastal clean-up

Note to Table 2-1: Table adapted from Andrady (2015)

Rank	Debris item	Count (millions)	Plastic used
1	Cigarette filter	2.19	Cellulose acetate
2	Plastic bags	1.13	PE
3	Food wrapper/container	0.94	PE, PP
4	Caps and lids	0.91	PP, HDPE
5	Beverage bottles	0.88	PET
6	Cups, plates and cutlery	0.51	PS
7	Glass bottles	0.46	-
8	Beverage cans	0.46	-
9	Straws and stirrers	0.41	PE
10	Paper bags	0.33	-

2.5 The impact of plastic accumulation

Due to the combination of poor waste management practices and direct litter, plastic is now so ubiquitous on every surface of the Earth (and has accumulated in both terrestrial and marine deposits) that there is the suggestion they should be considered as geological indicators for the Anthropocene epoch (Zalasiewicz et al., 2016). The environmental impacts of this plastic accumulation vary depending on whether the issue of choice is land based plastic (accumulation in landfills) or plastic that has entered the ocean.

2.5.1 *In landfills*

The greatest concerns arising from plastic accumulation in landfill relate to two aspects: toxicity of leachates from the stored materials and occupation of land space. Plastic's durability and resistance to microbial attack is useful during its lifetime, but it also means it will occupy increasing amounts of landfill space in a world that has decreasing suitable landfill sites. In the 1980s the issue of solid waste disposal almost reached crisis level in the USA due to growing volumes of municipal solid waste but shrinking landfill capacity and public opposition to the sites proposed for new solid waste facilities (Philp et al., 2013). Japan also struggles due to high pressures on land use and negative public opinion of waste disposal facilities making it hard to find suitable and accepted disposal sites (Philp et al., 2013). Even in Australia, with low populations and large land area, a report commissioned to assess the projections for landfill capacities in Australia, offered the conclusion that landfill, as a final waste solution, should be used conservatively (Philp et al., 2013; Pickin, 2009). These issues are magnified in countries with high populations but small land areas. Furthermore, whilst landfills may be safe for temporary storage of materials, over geological timescales, plastics stored in landfills may end up being released through the action of erosion etc. and then enter the environment (Zalasiewicz et al., 2016).

2.5.2 *In the oceans*

A United Nations Environmental Program marine plastics report states that the accumulation of plastic litter in the oceans is a concern for all mankind (UNEP, 2016). It is a truly global challenge because low density plastics such as PE and PP float in seawater and are thus distributed across the globe by wind and currents, meaning litter does not necessarily remain close to the point of entry to the ocean and impacts are felt globally (Zalasiewicz et al., 2016).

Moore (2008) defined eight complex problems caused by plastic in the marine environment:

1. Impact on the tourism industry due to beach litter
2. Entanglement and death of marine life
3. Ingestion and death of marine life
4. Dispersal of invasive species
5. Source and sink of persistent organic pollutants
6. Inhibition of gas exchange and impacts on sediment inhabitants on the sea floor
7. Threat to coastal species due to destroyed habitat
8. Interference with boats leading to accidents and damage and in some cases death

These complex problems can basically be grouped under two broad categories of economic and environmental impacts.

- Economic impact:

The exact economic impact of marine litter is hard to determine although it is agreed that there are significant economic drawbacks for both the cost of non-action (e.g. plastic littered beaches resulting in reduced tourism and associated income for the affected areas, or flooding and sewer clogging) and action (e.g. high costs associated with clean-ups) (Bugnicourt et al., 2014; Moore, 2008; Thevenon, 2014; UNEP, 2016). Relatively few groups have tried to quantify the economic damages (Beaumont et al., 2019; McIlgorm et al., 2011; Mouat et al., 2010; World Economic Forum and Ellen MacArthur Foundation, 2017). In two most recent reports that do, one places the global impact of plastic litter at US $40 billion per year (World Economic Forum and Ellen MacArthur Foundation, 2017) whilst the other estimate that the economic costs of marine plastic are between $3,300 and $33,000 per tonne of marine plastic per year (Beaumont et al., 2019). Whilst a useful exercise, it must be emphasised that these estimates are far from accurate, more the purpose is to draw attention to the fact that there is an often unconsidered cost associated with plastic pollution that needs to be considered.

- Environmental impact:

The expected impacts of different marine debris on birds, turtles and mammals is recorded in **Table 2-2**. This shows that the top items in regard to impact are associated with fishing activities, whilst after that are again plastic bags and single-use, food related items that wash in from the land. The most well-known and visual consequence of these items of plastic litter is entanglement and ingestion leading to the choking and starving of animals such as sea birds, turtles, whales, dolphins, seals and fish (Moore, 2008). The top items causing these impacts are associated with fishing activities, whilst after that are plastic bags and single-use, food related items that wash in from the land (UNEP, 2016). As mentioned above, this indicates that plastic bags and food service items are the focus for reducing the impact of plastic waste. There is also new research suggesting that the presence of plastics in the ocean can have less obvious but still negative impacts on coral reef and sedimentary habitats (Green et al., 2017, 2015; Lamb et al., 2018). The effect of microplastic on marine life is less clear. It is known that microplastics can be attachment points for persistent organic pollutants (hydrophobic compounds) in the water and can transport these to organisms in the marine sediments and then up the food chain through ingestion (Teuten et al., 2007). But it is unknown whether ingestion of plastics is sufficient to expose sea-life to detrimental levels of chemical additives (Thompson, 2015). Floating plastic debris is also known to be a vector for transfer of invasive species or non-native organisms which can threaten the populations of (until now) isolated environments (Barnes, 2002; Barnes et al., 2009; Gregory, 2009).

Table 2-2: Ranking of marine debris items by expected impacts on marine animals

Note to Table 2-2: Table adapted from UNEP (2016).

Item	Expected impacts on large marine animals (birds, turtles, mammals)
Ghost fishing gear (traps, monofilament line, fishing nets).	High
Plastic bags	Med-high
Plastic food items (utensils, caps, packaging, containers)	Med-high
Balloons	Medium
Cigarette butts	Medium
Cans	Med-low
Unidentified plastic fragment	Med-low
Larger plastic items (plates, bottles)	Med-low
Glass bottles	Low
Paper bags	Low

2.6 Existing technologies for managing plastic waste

The widely used 'waste hierarchy' (**Figure 2-5**) ranks the available waste disposal options by most preferred to least preferred based on the sustainability of each practice. Prevention and reuse obviously negate the need for waste management and are ranked at the top of the hierarchy and are a key way of reducing plastic accumulation. However, in the case that this is not practical, recycling is the most desired outcome for conventional waste management with landfilling the least desired outcome.

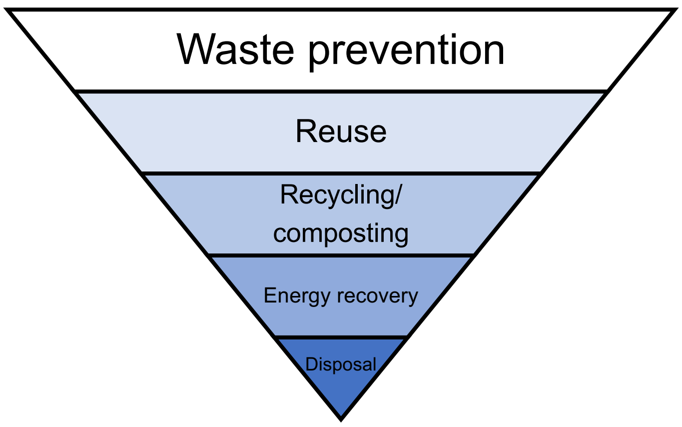


Figure 2-5: Waste hierarchy

More recently, the waste hierarchy has been re-envisioned to highlight both the technical and biological aspects of waste management (**Figure 2-6**). The inner loops of the systems diagram are considered more desirable, and reflect the higher rungs on the waste hierarchy, prioritising reduction and reuse. If reduction and reuse cannot be achieved, then the outer loops are considered. Recycling of conventional plastics sits in the technical materials cycle, whilst options such as composting sit in the biological cycle.

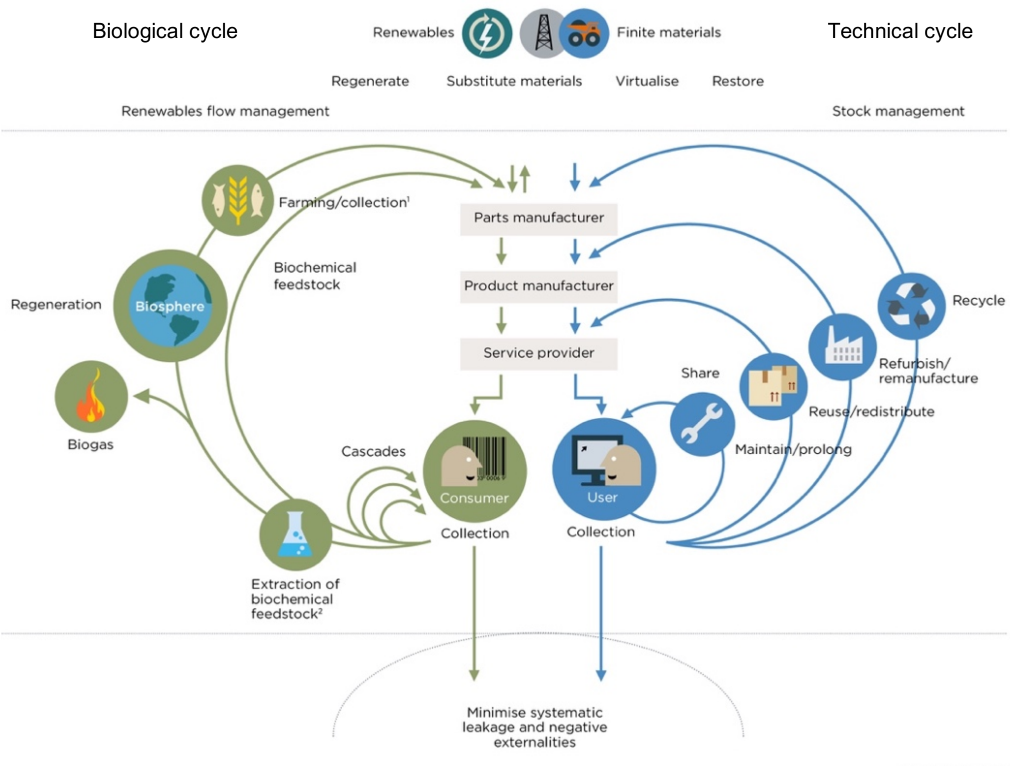


Figure 2-6: Circular economy systems diagram

Note to Table 2-6: Figure adapted from Ellen MacArthur Foundation website.

Recycling itself encompasses a variety of different methods, as presented in **Figure 2-7**. The specifics of each of these technologies with regards to management of plastic wastes are discussed in section 2.6.1 and a comparison of the technologies is presented in **Table 2-4**.

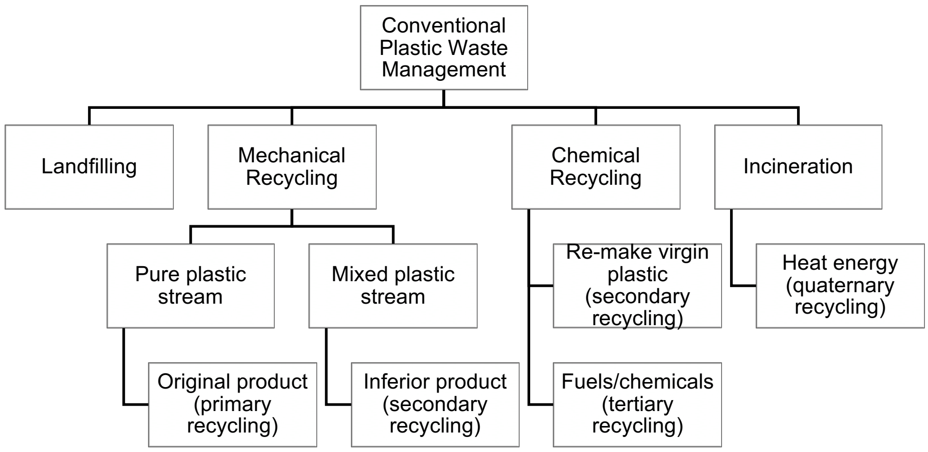


Figure 2-7: Conventional plastic waste management options

2.6.1 *Specifics of waste management technologies*

- *Mechanical Recycling*

Mechanical recycling involves the separation of different types of plastics, then physical shredding of plastics into smaller flakes, followed by cleaning and then reprocessing of the flakes to give a new plastic product (Merrington, 2011). This is called primary recycling (or closed-loop recycling) and means that the performance characteristics and functionality are considered equivalent to the original virgin plastic material (the only truly profitable example of this type of recycling is that of PET and HDPE bottles which have a steady supply) (Niaounakis, 2013). However, it is known that in reality, after each cycle the mechanical properties are lower compared to the starting material (Soroudi and Jakubowicz, 2013). Secondary recycling (or down cycling) occurs when the recycled materials are used to make a product of lesser value or material demand than the original products (an example is mixed virgin plastics being used to make flooring tiles or a park bench) (Merrington, 2011). Sorting the collected materials is an important aspect of mechanical recycling and there are various techniques used to identify the plastics including manual sorting, density separation, electrostatic processes, and various optical systems (near infrared, ultraviolet, X-ray analysis, and fluorescent light or laser radiation).

- *Chemical recycling*

Chemical recycling involves the conversion of polymers into low molecular weight materials or smaller hydrocarbon molecules (Ariffin et al., 2010). It is aimed at retaining and reusing material resources and ideally aims to convert waste plastics into their original monomers for re-polymerisation thus reducing the need for virgin feedstock whilst regaining the properties of a virgin polymer. In this way, chemical recycling overcomes the mechanical degradation associated with mechanical recycling. In some cases, chemical recycling refers to cascading utilisation meaning that secondary products, such as industrial chemicals, are produced from the depolymerisation products as opposed to virgin plastics (Ariffin et al., 2010).

There are three main approaches to chemical recycling (Panda et al., 2010):

1. Depolymerisation: This is mainly performed on polymers that were produced via condensation reactions (such as polyesters and PET but not polyolefins) and aims to reverse the synthetic reactions to give the original monomers. These monomers can then be used to form a virgin polymer.
2. Partial oxidation: This involves partial combustion of the polymers under controlled conditions to yield a mixture of hydrocarbons, CO and hydrogen.
3. Cracking/pyrolysis: This involves breaking down the polymer chains into lower molecular weight compounds with the vision that the products can be used as fuels or chemicals. The procedure can be achieved through reaction of the waste with hydrogen over a catalyst, degrading the polymer by heating it at high temperatures in the absence of oxygen, or using a catalyst and heating the polymer at moderate temperatures in the absence of oxygen.

- *Incineration*

Incineration of plastic waste results in energy generation if conducted in properly controlled facilities. This has become a popular waste management technique as the calorific value of plastics is similar to that of fuel oil (as reported in **Table 2-3**) and thus the incineration of a plastic product produces thermal energy of the same order of magnitude as the oil used in its manufacture (Panda et al., 2010). However, incineration also results in the production of greenhouse gases (i.e. release of the fossil carbon contained in the plastic) and in some cases highly toxic pollutants such as dioxins and furans (Yee and Foster, 2014). Public support can be weak and strict regulation and air emission controls are required, especially in developing countries (Panda et al., 2010).

Table 2-3: Calorific values of plastics compared with conventional fuels

Note to Table 2-3: Table adapted from Panda et al. (2010).

Fuel	Calorific value (MJ/kg)
Methane	53
Gasoline	46
Fuel Oil	43
Coal	30
Polyethylene	43
Mixed plastics	30-40
Municipal solid waste	10

- *Landfill*

Landfilling is one of the oldest approaches to waste management and involves disposal of waste materials by burying. In a conventional (dry, sanitary) landfill, the waste is deposited at site, spread out, compacted and then covered with a layer of soil. The covering is needed to minimise waste blown off the landfill to become litter (Barnes et al., 2009). The bottom of the landfill is lined to reduce leachate interaction with the environment. When the landfill reaches capacity, it is covered with a layer of clay to reduce leakage of waste and gas and to prevent water from entering the landfill (European Bioplastics, 2015a). Conventional plastic materials show very little degradation in landfill and will remain entombed for hundreds of years (Webb et al., 2012).

2.6.2 *How do the technologies compare to each other?*

A comparison between the different technologies is presented in **Table 2-4**. It can be seen that each technology has its strengths and weaknesses, however, in the case of recycling many of these weaknesses can be addressed through the improvement of sorting or materials design. On the other hand, the main issue associated with landfill and incineration is that they do not reduce the requirements for virgin polymer production, and this is a fact that cannot be changed. Recycling (mechanical or chemical) of fossil derived polymers has the lowest non-renewable energy demand of the presented waste management methods due to the re-use of non-renewable resources reducing requirement for virgin polymer (Gironi and Piemonte, 2011; Heyde, 1998). Chemical recycling is less energetically favourable than mechanical recycling but still acts to conserve resources (Hopewell et al., 2009).

Table 2-4: Comparison of the strengths and weaknesses associated with different waste handling methods

Green= strength, Pink = weakness, Light grey = neutral.

	Mechanical Recycling	Chemical recycling	Incineration	Landfill
Reduces requirement for virgin feedstock?	Yes - if it is a closed-loop process.	Yes - if it is a closed-loop process.	No	No
Separation of waste required?	Yes.	Yes, but less sensitive to contamination than mechanical recycling.	No.	No
Suitable for all plastic types?	No, thermoplastics can be recycled as they can be re-melted whereas thermosets cannot.	No.	Yes	Yes
Energy Recovery?	No, but materials are conserved.	No, but materials are conserved.	Yes	Not for plastics.
Space requirement for infrastructure?	Low	Low	Low	Ever increasing amount of space is required (weakness when available space is restricted).
Process complexity	Relatively simple process.	Complicated process.	Simple process	Simple process
Pollution risk	Low	Low	Can present a health and pollution risk due to production of hazardous ash if poorly operated.	Long-term risk of contamination of soil and groundwater.
Cost?	Capital intensive at start-up.	Capital intensive at start-up and during process.	Capital intensive at start-up.	Least capital intensive.

2.7 Section 2: How could biodegradable plastics address the issue?

As set out in the first half of this chapter, the use of plastics has created the issue of plastic solid waste accumulation and associated environmental impacts. In response, there is increasing discussion around the relative benefits and drawbacks of the variety of proposed "solutions". Biodegradation is often included in these discussions, with a range of views as to whether it can be considered as a positive attribute or not (e.g., Tabone et al. (2010) list it as a green design metric when considering the engineering of materials but Robertson (2014) argues that we should be focusing on the development of biobased but not biodegradable materials). Here, this debate surrounding the role of biodegradable plastics in solving plastic solid waste accumulation is reviewed and commented upon. In short, we conclude that marine biodegradable plastics could be an important part of the solution. On land they could expand the available waste management options to include composting and anaerobic digestion (AD), while in the ocean they could reduce marine life impacts through reduced persistence compared to conventional plastics.

2.8 Background: biodegradable plastics

The term 'bioplastic' encompasses a number of subsets including:

a) biobased but not biodegradable plastics,
b) biodegradable but not biobased plastics or
c) biodegradable AND biobased plastics as illustrated in **Figure 2-8**.

This thesis focuses on the biobased and biodegradable subsection of plastics. The term 'biodegradable plastic' is used to refer to a plastic that is both bioderived AND biodegradable, whilst 'bioplastic' refers to any plastic produced from biomass.

	Fossil-based	Biobased
Fully biodegradable	• Polybutylene succinate • Polycaprolactone • Polybutyrate • Some polylactic acids and polyhydroxyalkanoates	• Thermoplastic starch (TPS) • Starch blends • Polylactic acid (PLA) • Polyhydroxyalkanoates (PHA)
Non-biodegradable	• Polyethylene (PE) • Polypropylene (PP) • Polyethylene terephthalate (PET) • Polystyrene (PS) • Polyvinylchloride (PVC)	• Biobased polyethylene • Polyethylene furanoate

Figure 2-8: Subsets of bioplastics

Note to Figure 2-8: The biobased and biodegradable subsection is coloured green, whilst the widely used, conventional plastic subset is coloured blue. Figure adapted from Shen et al. (2009) and Philp et al. (2013).

Biodegradability is governed by the ASTM D6400 (USA), EN13432 (Europe) and ISO17088 (International) standards. These state that truly biodegradable plastics are those that biodegrade as a result of the natural action of microorganisms such as bacteria, fungi and algae. The resulting products should be CO_2, methane, water and biomass with an absence of ecotoxic effects or residual by-products. Thus, oxo- and photo-degradable plastics (which contain additives to induce degradation under particular conditions) are not truly biodegradable according to the standard, as residual microfragments remain after the breakdown period (European Bioplastics, 2015b).

The most common biodegradable and bioderived plastics include thermoplastic starch (TPS), polylactic acid (PLA) and polyhydroxyalkanoates (PHA) (European Bioplastics, 2017) (chemical structures are shown in **Figure 2-9**).

Briefly, starch is a polysaccharide that is produced by plants for energy storage. The biodegradable plastic TPS is a material that is produced by heating and mixing starch with a plasticiser such as water or glycerol (Storz and Vorlop, 2013; Tomka, 1991). PLA is a polyester that can be produced from lactic acid (which is produced by fermentation of biomass such as sugar or starch) (Storz and Vorlop, 2013). PHAs are a family of polyesters predominantly produced by bacterial fermentation of biomass leading to intracellular accumulation of the polymer (Braunegg et al., 1999; Jiang and Zhang, 2013).

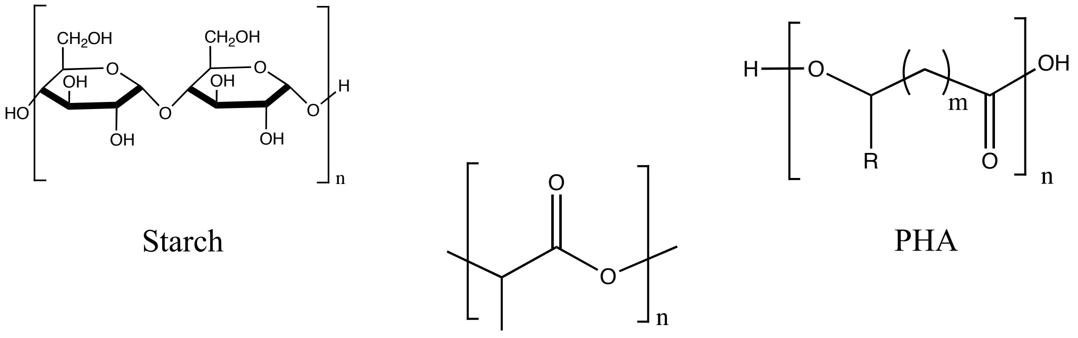


Figure 2-9: Chemical structures of common biodegradable polymers

2.9 Current market share for biodegradable plastics

The sales of compostable and biodegradable plastic products only represent a small proportion of the total biobased plastics market, as is illustrated in

Figure 2-10. This translates to an even smaller portion of the total plastics market, as the biobased plastics sector is itself small, currently sitting around 7 million tonnes per year* (around 2% of the staggering 300 million tonnes of plastics produced per year (Aeschelmann and Carus, 2015)). That said, a strong growth rate in the production of bioplastics is expected to continue from 2015 to 2020, specifically with a strong growth rate of the PHA market (European Bioplastics, 2018). In terms of absolute volumes of production, PLA and starch blends continue to dominate the biodegradable sector (Aeschelmann and Carus, 2015; Kaeb et al., 2016).

* According to the Nova-Institute which includes epoxies, polyurethanes, some rubbers and cellulose acetate in their biopolymer market share analysis.

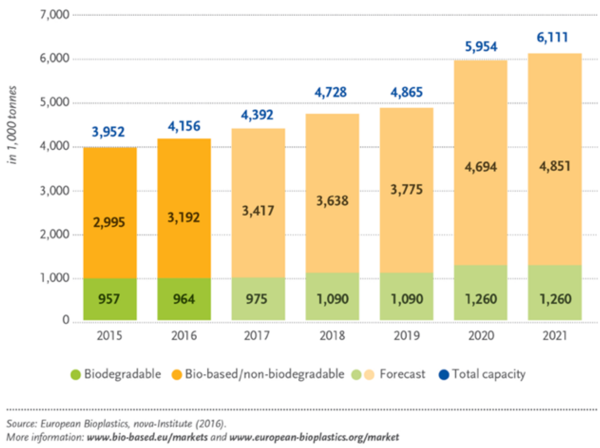


Figure 2-10: Market breakdown of global production capacities for biobased verses biodegradable plastics

Note to Figure 2-10: Figure adapted from European Bioplastics (2017).

In regard to applications, the packaging field contributes to over 50% of global biodegradable plastics consumption (European Bioplastics, 2017), and the best-selling items are waste/carrier bags (2/3 of total market), flexible and rigid packaging and disposable crockery as illustrated in **Figure 2-11** (Kaeb et al., 2016). Overall, rigid and flexible packaging materials are predicted to remain the strongest growth areas (Aeschelmann and Carus, 2015).

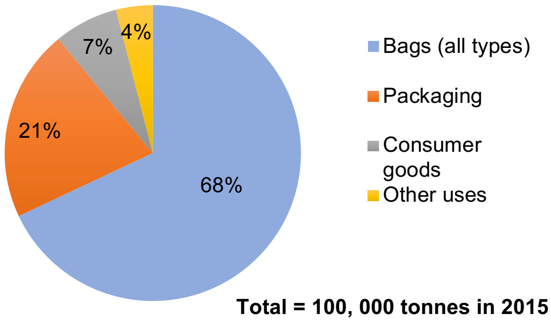


Figure 2-11: Consumption sectors for biodegradable plastics by application in the European Union

Note to Figure 2-11: Figure adapted from European Bioplastics (2017).

2.10 Substitution potential of biodegradable plastics

Although current production levels and substitution levels remain low, with the focus being on packaging, theoretically, biodegradable polymers are able to substitute for most of the conventional polymers currently used. Shen et al. (2009) have studied the technical substitution potential of biobased plastics to replace their petrochemical counterparts in considerable depth. As of 2007, they estimated the total maximum technical substitution of conventional plastics by biobased plastics as 94% - comprised of a 31% substitution by biodegradable plastics and a 63% substitution by biobased but not biodegradable plastics. Using similar methodology for considering the biodegradable replacement potential of containers and packaging in Japan, Yano et al. (2014) concluded that 87% would be replaceable by PLA or PLA blends (of note is that PET containers were excluded as there is a well-functioning collection and recycling system for these in Japan).

However, as mentioned, actual production and substitution levels remain far from this theoretical maximum. It will probably not be possible to exploit this potential in either the short to medium term due to factors such as economic considerations, difficulty in fast scale-up, difficulty in assuring availability of bio-based feedstocks and the slow adoption of new plastics by the plastics sector (Shen et al., 2009). Nor is it clear that the aim should be to substitute biodegradable plastics for all plastics.

2.10.1 *Consideration of land availability*

Of course, when discussing substitution potential, the availability of the raw materials to produce the polymers must be included. Biopolymers are produced from biomass, the production of which often requires agricultural land. Thus, producing biopolymers presents a food-versus-liquid fuel-versus-polymer feedstock issue. Colwill et al. (2012) have considered this issue through analysing a hypothetical 2050 scenario where all plastics and liquid fuels are produced from renewable resources. Different consumption and productivity scenarios were considered. The conclusion is that it would be possible to manage the production of all products simultaneously on currently available agricultural land. However, it is highlighted that when developing bioderived products, the focus should be on the efficient use of materials and alternative feedstocks to ensure that competition with food production is minimized. The ideal case would be better land management and less wastage, which would lead to an increase in the productivity of our agricultural land and reduce competition for land space. The analysis of Colwill et al. (2012) suggest that plastics production would require 5% - 7% of the total agricultural land. This assumption was based on the production of bio-PE to generate an upper limit scenario, as bio-PE is considered the most land-intensive plastic to produce. Currently, the Institute for Bioplastics and Biocomposites suggests that bioplastic production accounts for less than 0.01% of global land area and based on the percentage of plastic production that is currently biobased, this predicts lower than 5% of agricultural land area would be required, even if all plastics were biobased (IfBB, 2016).

2.11 **How do biodegradable plastics expand waste management practices?**

The discussion around the utility of biodegradable plastics as a way to combat solid waste has been considered in various forms for many years (e.g. Huang et al. (1990)). An important part of this discussion is understanding how biodegradable plastics actually fit into and expand current waste management practices. Most biodegradable polymers can be processed via conventional waste management techniques (technical cycle) as well as techniques specific to the property of biodegradability such as anaerobic digestion and composting (biological cycle). Colwill et al. (2010) challenge the tendency of the literature to assume that the end-of-life (EOL) management of biopolymers SHOULD only be biodegradation and that this by default provides ecological benefits. This idea will be expanded on below. It will also become evident that for most of the conventional waste management options there is little difference between the management of biodegradable versus conventional polymers, except for the case of landfill, where methane emissions can prove problematic.

- *Mechanical recycling*

Some authors (for example Soroudi and Jakubowicz (2013)) argue that sustainable use of carbon sources, regardless of whether they are fossil or bio-based, must include the production of recyclable materials. This extends service life prior to chemical or biological recycling and thus has a positive environmental impact. However, Colwill et al. (2010) note that the 'opportunities for conserving resources through the recycling of biopolymers are rarely addressed' and this comment is expanded upon in their 2012 paper (Colwill et al., 2012). In principle starch, PHA, and PLA can be recycled through conventional mechanical means.

Despite technical feasibility, one caveat to the ability of biodegradable plastic to be processed via recycling is that they would need to be available in the necessary critical mass to justify separate recycling streams, and ensure commercial success (Cornell, 2007). This mass has been placed around 200 million kg of a single polymer produced annually (Niaounakis, 2013). The inclusion of minor resins into the current collection and sorting systems results in an extra cost for recyclers due to increased complications in sorting. This cost will often need to be borne by the minor resin if it is desired as a clean stream for recycling (Cornell, 2007). Thus, in order for biodegradable polymer recycling to be viable Cornell (2007) suggests that the following criteria would need to be met:

1. Availability of collection infrastructure,
2. Investors for the market,
3. Profitable applications for biodegradable plastics, and
4. Access to enough materials in the recycling stream.

The author concludes that identification of profitable applications and ensuring availability of a high enough volume of materials in the recycling stream are the two areas that require the most attention. It is also noted that research into the recycling (chemical or mechanical) of biopolymers is still new and lacks a deep understanding of the factors affecting performance, economy and sustainability (Soroudi and Jakubowicz, 2013). This means, that like most polymers, recycling of biodegradable polymers is still often a 'down cycling' process as desirable properties are compromised when successive recycling leads to shortening of the polymer chains (Myung et al., 2014).

Starch: Work detailing the mechanical recycling of pure starch is scarce with most work focusing on blends with non-biodegradable materials. However, some fully biodegradable blends have been considered. La Mantia et al. (2002) looked at the mechanical recycling of a starch-polycaprolactone system. They found that this resulted in degradation, predominantly of the polycaprolactone, whilst some cross-linking of the starch was observed. They concluded, that under the selected conditions, changes in rheological and mechanical properties only occurred after 5 recycles (La Mantia et al., 2002). Conversely, Lopez et al. (2011) found the recyclability of Mater-Bi starch blend to be very poor and suggested it is directly composted. There is currently no further clarity to these contradicting messages.

PHA: Shah et al. (2012) have considered the impact of mechanical recycling on the properties of PHB and also considered the impact of virgin and recycled material blends. They found that on the 10^{th} regrind a 10% reduction in tensile strength occurred, and an associated large change in viscosity was observed. This is not unexpected though, and still means the material can be recycled multiple times. In regard to blending the material, there was only a small drop in tensile strength and viscosity for a 50:50 virgin to regrind ratio. Zaverl et al. (2012) showed that PHBV is recyclable with little change in properties and chain length up to 4 cycles but begins to demonstrate decreased properties after 5 cycles. It has also recently been shown that inclusion of a small amount of non-PHA material (obtained by extraction from biomass along with PHA material), can improve the mechanical properties of compounded PHA plastics (Werker et al., 2016). This could prove a useful discovery for improving the properties of recycled PHA plastics.

PLA: A range of results for the impacts of mechanical recycling on PLA properties are recorded. A few studies have reported minimal/acceptable changes in polymer properties for up to 10 regrind cycles, similar to PHA (Lopez et al., 2011; Soroudi and Jakubowicz, 2013; Żenkiewicz et al., 2009). Another found that the mechanical properties of the PLA had become too poor for industrial use after 7 regrinds (Pillin et al., 2008). Again, this still represents an acceptable number of reuse cycles to make mechanical recycling an attractive target.

- *Chemical recycling*

Like conventional polymers, some of the biodegradable polymers have the correct chemical structure to be chemically recycled.

Starch: To date, the author of this thesis had found no literature regarding the chemical recycling of starch.

PHA: There have been multiple investigations into the controlled chemical degradation of PHB via enzymatic hydrolysis, thermal degradation and ring-closing depolymerization (Ariffin et al., 2010; Myung et al., 2014; Soroudi and Jakubowicz, 2013; Yang et al., 2014). For example, Ariffin et al. (2010) investigated the conversion of PHAs into vinyl monomers through heating in the presence of specific catalysts, demonstrating they could produce useful chemicals at reduced temperatures. Yang et al. (2014) used microwaves to thermally degrade PHB into functional chemicals. Myung et al. (2014) investigated an efficient 'closed-loop' chemical recycling process for PHAs with hydrolysis/ pyrolysis of the PHA polymer giving monomers that can then be re-polymerised to give PHA in a bioreactor. They propose that this is a more efficient recycling strategy than recycling via the natural carbon cycle (i.e. biodegradation). However, whilst promising, all of these experiments have only been proved viable at the laboratory scale.

PLA: There are two main processes for the chemical recycling of PLA. The first is high temperature hydrolysis to obtain lactic acid, the second is thermal degradation to obtain L,L-lactide. Both of these products can be used in polymerization to produce virgin PLA (Soroudi and Jakubowicz, 2013).

- *Incineration*

Biodegradable plastics can undergo incineration in the same manner as non-biodegradable plastics. Furthermore, the energy capacity of biodegradable polymers is similar to that of conventional plastics. The difference is that the incineration of bioderived materials is a closed-loop process because CO_2 fixed during plant growth is released to the atmosphere, as opposed to the release of fossil CO_2 which occurs upon the incineration of petroleum derived polymers. Certified compostable plastics may also have lower environmental impacts upon incineration as the certification requires it to be proved that the plastic contains low levels of heavy metals (European Bioplastics, 2015c).

- *Landfill*

The disposal of biodegradable plastics in landfill is where a difference in regard to end-of-life impacts can be noticed when compared to non-degradable plastics. Whilst conventional plastics are assumed to experience no degradation in landfills the assumed levels of degradation in landfill for biodegradable plastics can differ from 0% up to 85% (Yates and Barlow, 2013). Upon degradation the assumption is that methane is produced (Yates and Barlow, 2013) which has a higher global warming potential than CO_2. It is hard to draw definitive conclusions in regard to the impact of biodegradable plastics in landfill due to conflicting evidence in the literature. For example one research group investigating the possibility that biodegradable plastics could help to decrease the amount of space occupied in landfills by plastics found that there was very little volume and weight change under anaerobic conditions, and that only PHBV demonstrated appreciable degradation rates when the landfill was aerated (Ishigaki et al., 2004). Conversely, Levis and Barlaz (2011) modelled the greenhouse gas (GHG) emissions of a variety of degradable substrates in landfill, and found a degradable polymer to contribute significantly to the GHG emissions of the landfill, and suggested that only inert materials should be landfilled. Even if there is no definitive answer, it is likely that there will be SOME methane emissions associated with depositing biodegradable plastics in landfill and that this practice should thus be avoided if there is no landfill capture in place and GHG emissions are to be minimised.

2.12 **Waste management practices specific to the property of biodegradability**

- *Industrial composting*

Composting is the biological transformation under aerobic conditions of organic matter to CO_2, water, heat and plant available biomass by micro-organisms, and is considered a biological recycling mechanism for biodegradable plastics (De Wilde et al., 2014a). There are a variety of methods for composting (windrow composting, table composting, row composting, tunnel composting, drum reactors) but all have the same flow of materials and final products (De Wilde et al., 2014a; European Bioplastics, 2015b).

There are a variety of standards that govern aerobic compostability under industrial conditions including ASTM D6400 (USA), ISO 18606, ISO 17088:2012 (international) and EN 13432 (Europe). These standards require that at least 90% of the organic matter is converted to CO_2 within 6 months due to biological action and that no more than 10% of the residue is retained by a 2mm sieve after 3 months composting. The quality of the compost should also not deteriorate as a result of the added plastic material. Compost then slowly degrades when applied to soil and releases nutrients essential for plant life and improves soil structure through binding the soil particles together and increasing water retention (De Wilde et al., 2014a).

- *Anaerobic digestion*

Although a more complex and expensive system than composting, anaerobic digestion (AD) has the benefit of energy production (De Wilde et al., 2014a). AD is a technology for the biodegradation of biomass and organic waste under anaerobic conditions to generate biogas (CO_2 and methane) that can be used directly as an energy carrier or used in a combined heat power plant for energy production (De Wilde et al., 2014b). The residual materials (>40% of the organic matter) remain in the digestate. In most cases this is then composted so that it aerobically degrades further and the compost can be used in agriculture.

In general, little pre-treatment of materials is required before they enter the digester (inert materials and plastics may be fed to the reactors, although in practice recyclables are normally first removed)

(De Wilde et al., 2014b). The average retention time in a digester is 20-30 days and the gas yield depends on the effectiveness of the digester, as well as factors such as the overall digestibility of the substrate, the digestion kinetics, temperature, pH, retention time, and mixing behaviour. Because it has a higher capital cost than other waste management option, outside of Europe where the costs for waste treatment and disposal in general are lower, AD has not been implemented to a large extent (De Wilde et al., 2014b).

No current standards focus solely on the recovery yield of compostable bioplastics as biogas and there is no clear definition available of what biodegradable products are suitable for AD. The European Bioplastics Association notes that further research is needed in this area (European Bioplastics, 2015d). Anaerobic biodegradability can be estimated from standards such as ISO 11734, ISO 14853, ISO 15985, ASTM D5510, ASTM D5511, ASTM D5526, but this does not necessarily give information about volumes of biogas production.

2.12.1 *The reality of composting and AD for the waste management of biodegradable plastics*

One of the key potential benefits of using biodegradable plastics is that there would be no need to separate biodegradable contaminants, such as food waste, from the plastic waste streams if they were treated via composting or anaerobic digestion (De Wilde et al., 2014a). However, the current reality is that composting and AD only seem to be feasible for clean (source separated) fractions of biodegradable plastics. This is due to poor organic collection systems, the small number of available facilities, and the fact that existing facilities are often not equipped to take biodegradable plastic materials (Álvarez-Chávez et al., 2012; Doyle, 2015; Hottle et al., 2013). For example, a study performed in a wet AD facility equipped with a pre-treatment demonstrated that all compostable plastics were skimmed off by a rake and ended up in the light fraction, which was then discharged into a rubbish container (De Wilde et al., 2014b). Although this issue has been known about for some time (Korner et al., 2005; WRAP, 2009), it has not yet been resolved.

2.13 **Sustainability of biodegradable plastics**

As introduced in section 2.1, when it comes to assessing the environmental impacts of plastic, the production, use and end-of-life phases all need to be considered.

With regard to a comparison between the production of biodegradable plastics and conventional plastics, Piemonte (2011) considered a cradle-to-gate life-cycle assessment (LCA) study to determine the primary energy requirements for producing PLA and starch packaging compared to PET or PP packaging. The result was an energy requirement reduction of 27 - 40% for the biobased packaging. This is not as large a reduction as it seems though. Biobased packaging will normally have a lower non-renewable energy demand in production due to the biogenic nature as opposed to fossil nature of the carbon used to produce the polymer, however, the CO_2 footprint of production is normally fairly similar (Gironi and Piemonte, 2011; Pietrini et al., 2007; Yates and Barlow, 2013).

From a waste management perspective, there have been many studies considering the environmental burden of end-of-life options for both biodegradable and non-biodegradable plastics, although, in almost all cases, 'environmental burden' has been assessed in terms of carbon footprint and occasionally non-renewable energy use. For all plastics, mechanical recycling is often found to be the superior option (Cosate de Andrade et al., 2016; Hottle et al., 2017; Madival et al., 2009; Piemonte, 2011; Piemonte et al., 2013; Rossi et al., 2015; Yano et al., 2014; Yates and Barlow, 2013) assuming suitable collection and recycling technology are in place. This is an interesting result for biodegradable polymers, as mechanical recycling does not require inherent biodegradability. Of note though is that none of the LCA studies take into account that the properties of mechanically recycled materials may have been reduced upon mechanical processing. And thus, whilst mechanical recycling may appear superior via LCA in the short-term compared to other waste management techniques, the ability to recycle the polymer via chemical means or organic means (AD, composting) may be useful in the long-term (after repeated mechanical recycling loops).

LCA also provides a few other unexpected results. Firstly, regarding composting, Rossi et al. (2015) found landfill and industrial composting to provide the worst environmental impacts for biodegradable polymers, challenging the idea that composting is, by default, an environmentally friendly option. Secondly, regarding AD, Hermann et al. (2011) compared AD, composting (home and industrial) and incineration via LCA and found that AD presented as the most environmentally friendly option only if the efficiency of incineration plants do not increase. Yates and Barlow (2013) support this outcome, as after reviewing the literature they also conclude that with sufficient energy recovery, incineration could be a more environmentally friendly option than those end-of-life (EOL) options that require biodegradability. Thirdly, regarding landfill, biodegradable polymers are normally found to contribute significantly higher CO_2e than conventional polymers meaning that the impacts of using biodegradable polymers appear to be large if landfill remains the dominant waste management option (Heyde, 1998). These results challenge traditional assumptions about biodegradable plastics; however, it should be noted that the basis of comparison was limited to carbon footprint and that the studies did not consider food contamination, in which case, the conclusions may change.

It appears that at this stage, LCA cannot provide us with clear answers in regard to what mix of EOL scenarios would provide the optimal reduction of biodegradable polymers' environmental impacts (Hottle et al., 2013). Indeed, in a review of 10 LCAs for disposable beverage cups, van der Harst and Potting (2013) concluded that no single material emerged as environmentally superior and commented that it was actually difficult to deduce any trends at all. This stems from the fact that little life-cycle data is available for the impacts of different manners of disposal for biodegradable polymers (Hottle et al., 2013) and there is discrepancy concerning the level of degradation of biodegradable polymers in landfill, AD and compost, the methane emissions resulting from landfill or composting, the carbon credits which should be applied for composting, and the rates of electricity production from landfill and AD (Hottle et al., 2013; Yates and Barlow, 2013). LCA data across broad impact categories is also unavailable meaning analysis is normally limited to GHG emissions (Yates and Barlow, 2013).

A significant outcome of reviewing LCAs, is that there is actually a disconnect between LCA and the accurate assessment of mismanaged plastics either on land or in the marine environment. LCAs are limited in their ability to consider, let alone quantify, many of the environmental impacts associated with plastic accumulation, particularly in the oceans. This means that, whilst the information provided by LCA can give some environmental insight and point to the fact that the use of biodegradable plastics needs to be properly considered, it should not be the sole basis upon which to determine which plastics are most useful for reducing plastic accumulation.

2.14 **Marine biodegradability of common biodegradable plastics**

Marine biodegradability is an important attribute for reducing the impact of plastic litter but varies widely between different biodegradable (and compostable) polymers. There are only a few standards that govern the measurement of marine biodegradability of plastics. ASTM D6691-09 considers aerobic biodegradation in seawater, ASTM D7991-15 considers aerobic biodegradation in marine sediment, and ISO 18830:2016 and ISO 19679:2016 consider the aerobic biodegradation of plastic products in the seawater/marine sediment interface. Whilst a useful start, all of the standards are for controlled aerobic laboratory conditions and involve cultured organism populations. The Open-Bio Consortium recommends that more of a focus needs to be placed on understanding the marine degradation of different polymer types as well as different shapes of products and in a broader range of environments. To achieve this, the standards should incorporate a wider range of possible marine habitats which will lead to a better understanding of the effects of light, temperature, pressure, nutrient contents and the presence of different microbial populations on biodegradation. Anaerobic conditions also need to be considered (Weber et al., 2015).

In regard to what is known so far, results from most studies show that PHA has the best biodegradation profile in marine environments followed by starch materials (which exhibit a much slower rate). PLA shows almost no biodegradation (Accinelli et al., 2012; Greene, 2011; Volova et al., 2010). Even for starch and PHA films, full or partial biodegradation only occurred after 13-25 weeks, meaning there is still the potential for it to harm marine life, although the risk is reduced over the long-term (Greene, 2011; O'Brine and Thompson, 2010; Thellen et al., 2008). It is noted that films degrade faster than solid objects (Volova et al., 2010) so the degradation time of a solid object can be expected to be longer than 25 weeks.

In regard to trying to predict harm to marine life due to plastics in the marine environment, an interesting study has been performed to investigate the break-down of different polymer shopping bags in the gastrointestinal fluids of sea turtles. HDPE bags and oxo-degradable bags showed no degradation, whilst starch based bags showed some level of degradation. However, in 49 days the maximum degradation of the starch was 9% and so it was concluded that the timeframe for complete degradation was still too long to prevent death from ingestion (Muller et al., 2012). Research into marine biodegradable plastics could focus on improving this outcome.

2.15 **Factors driving the uptake of biodegradable plastics**

Factors driving the rate of uptake and role of biodegradable plastics will not only include their sustainability credentials but also financial, regulatory, consumer and technology/material aspects (Shen et al., 2009).

Brief examples of these aspects are as follows:

- Financial:

Fiscal policy measures will be required to support the transition to biodegradable polymers. These include support for low GHG processes and high prices on landfilling (which will improve the cost competitiveness of biological waste management) and price level monitoring of agricultural feedstocks (to ensure they are competitive with fossil fuels in order to drive the movement towards biobased materials) (Shen et al., 2009). The driving of technology by economics is demonstrated by the example of The Netherlands (where landfill is costly due to being close to sea level) and Japan (where excavating landfills requires digging into hard volcanic rock) being two of the countries with the best recycling and incineration systems (Hopewell et al., 2009) and it is envisioned this could extend to biological waste systems.

- Regulatory:

The introduction of policies and measures could substantially change the rate of uptake of biobased polymers (Shen et al., 2009). It is thought that bioplastics suffer from a lack of favourable policy regime when compared to those that were implemented to support biofuel production (Philp et al., 2013).

There is a strong correlation between landfill bans (zero waste to landfill or reducing recyclable waste to landfill) and lower landfill rates of plastics (PlasticsEurope, 2016). However, a word of caution is that such policies should be coupled with specific recycling targets and then monitored to ensure that they don't just lead to large increases in plastic waste incinerated (PlasticsEurope, 2014). Under the European Union Circular Economy Package, a target for reuse and recycling of plastic packaging materials of 55% has been suggested, with a maximum of 10% of all municipal waste to be disposed in landfill by 2030 (ACC and Trucost, 2016; European Comission, 2015a, 2015b). Bans on single-use plastic items can also be considered.

- Technology/material properties:

There are two aspects of technology that will play a role in the uptake and sustainability of biodegradable plastics – the materials development and the waste management.

Firstly, on the waste management side. Improved composting infrastructure including sorting after composting will allow biodegradable plastics to be processed in composting facilities (Gross and Kalra, 2002; Korner et al., 2005). Improved sorting technology that is financially viable would also ease concerns regarding recycling. A promising technology in this area includes fluorescent markers (Nextek, 2017; WRAP, 2009). Fluorescent markers entail marking the resin with a dye which when irradiated produces a signal that can be detected and used to sort the materials. Other companies (e.g. Tomra) are working on improving efficiency and reliability of plastic sorting equipment.

In regard to material properties, a key aspect is being able to produce biodegradable plastics with equivalent properties to conventional plastics, in order to provide industry competitiveness.

- Consumer:

Consumers will need to be supported in identifying and disposing of biodegradable plastics (WRAP, 2009). The Open-Bio project (a European initiative), is aiming to address this issue by 'increasing the uptake of standards, labels and harmonised product information lists for bio-based products in Europe' and developing a database on biobased products and their properties (Open-BIO, 2017).

To facilitate appropriate disposal, it has been recommended that there is the development of an identification code for compostable bags and containers to help separate them from recyclable materials.

2.16 **Summary and scope for the Ph.D. thesis**

Plastic accumulation both on land and in the oceans is an important issue of our time, and current trends predict that the magnitude of the issue will exponentially increase. It is thus imperative that we implement solutions to decrease the rate of plastic accumulation, as well as work towards reducing the impacts that inevitable accumulation will have. There will be no single solution to addressing these issues, so it is determining the effective combination of solutions that is important.

There is the potential that biodegradable plastics, with a theoretically large substitution potential and growing market share, could offer a solution through shifting waste management from the technical cycle to the biological cycle. However, it still remains unclear what the uptake of biodegradable plastics will/should be. It is also apparent that biodegradability is not in and of itself a 'sustainability achievement'. A multidisciplinary approach is required to explore the role of biodegradable plastics. This Ph.D. thesis takes such an approach (as explained in detail in the introduction (**Chapter 1**)), exploring three important drivers (material properties, environmental impact assessment, and social attitudes) relating to the use of biodegradable plastics in a sustainable plastics economy.

CHAPTER 3 PHA as a suitable coating for TPS

Chapter Summary

Within the material properties theme, the first objective was to '*Assess the suitability of PHA as a water barrier layer for TPS*'. This chapter presents the first of two research activities that relate to this research objective. The results presented in this chapter show that PHA is a suitable coating material for TPS, reducing moisture uptake and helping it to maintain its oxygen barrier properties.

This chapter was published in full as a journal article during the candidature.

Dilkes-Hoffman, L.S., Pratt, S., Lant, P.A., Levett, I., Laycock, B., 2018. Polyhydroxyalkanoate coatings restrict moisture uptake and associated loss of barrier properties of thermoplastic starch films. J. Appl. Polym. Sci. 135, 46379–46387. https://doi.org/10.1002/app.46379

3.1 Introduction

The redesign of plastic packaging materials is necessary in order to address the issue of plastic pollution. In the food packaging space, a multi-layered combination of thermoplastic starch (TPS) and polyhydroxyalkanoate (PHA) could be an interesting new material to consider as it has the potential to exhibit the high-barrier properties desired in food packaging, whilst also overcoming the current waste management issues of non-recyclable multi-layer materials as it is biodegradable.

It was recently estimated that a cumulative total of 8.3 billion tonnes of plastic has been produced to date (Geyer et al., 2017). So far, 6.3 billion tonnes of this plastic has been discarded as waste, and a majority of this waste is still present, having accumulated in both landfills and the environment (Geyer et al., 2017; Thompson et al., 2009). To reduce continued plastic accumulation and pollution there is a need to improve materials design and waste management practices (Barnes et al., 2009).

Plastic packaging accounts for more than a quarter of total plastic production, with a large portion directed to the food sector (PlasticsEurope, 2016; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). In addition to recycling difficulties, plastic food packaging represents a disproportionate and significant source of plastic pollution due to its single use nature (Andrady, 2015; Barnes et al., 2009). Food packaging can be hard to recycle due to the prevalence of multi-layered materials (with a different polymer type in each layer) and organic contamination. Despite this, the food industry is increasing its use of multi-layered materials, as they can provide higher resistance to water and gas transfer than single layered materials and thus reduce food spoilage (Barlow and Morgan, 2013; Dave and Ghaly, 2011). Whilst, there are many causes of food spoilage, with breaks in the cold-chain, over-production, consumer behaviour within the household environment being just a few, high-barrier packaging is known to play a significant role in managing such spoilage.

Multi-layer food packaging is thus a key target for redesign in the effort to reduce the impacts of plastic use (Peelman et al., 2013; Siracusa et al., 2008). High-barrier, biodegradable food packaging could present a useful alternative to non-recyclable, multi-layered packaging as it expands the range of end-of-life options to include biological processing.

One of the most promising candidate materials for this application is thermoplastic starch. TPS exhibits excellent biodegradability, is cheap, renewable, non-toxic and shows good oxygen and carbon dioxide barrier properties (Cooper, 2013). However, TPS is hydrophilic, absorbing water at high relative humidity (RH) which leads to a loss of its barrier and mechanical properties (Krochta and De Mulder-Johnston, 1997). This currently limits its use.

A common way to address this is via lamination of the TPS film with a hydrophobic polymer layer. Examples include lamination with a non-biodegradable polymer such as polyethylene (Cooper, 2013; Dole et al., 2005) or biodegradable alternatives such as poly(lactic acid) (PLA) (Martin et al., 2001; Sanyang et al., 2016), and polycaprolactone (PCL) (Martin et al., 2001; Ortega-Toro et al., 2015; Wang et al., 2000). However, a non-biodegradable coating excludes the package from biological processing, whilst many of the degradable polymer alternatives have relatively poor water barrier properties (Greene, 2011; Guzman-Sielicka et al., 2011; Shogren, 1997).

In this work, polyhydroxyalkanoates (PHAs) are considered as alternatives for coating of TPS. PHAs exhibit the highest water barrier properties of biodegradable polymers and have an excellent biodegradation profile, including in the marine environment (Averous, 2009; Shogren, 1997; Thellen et al., 2008). These polymers are intracellularly produced by bacteria, with the most common short chain length PHAs being poly(3-hydroxybutyrate) (PHB) and the co-polymer poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) (Laycock et al., 2014).

A layered material comprised of TPS coated in PHA is thus of interest, as it should exhibit reduced water sensitivity, good oxygen barrier properties and marine biodegradability. However, there is limited work confirming the ability of PHA to sufficiently protect a TPS film from moisture uptake while maintaining good barrier properties.

Previous work using both TPS and PHA has mainly focused on blends (Don et al., 2010; Imam et al., 1998; Parulekar and Mohanty, 2007; Reis et al., 2008; Thiré et al., 2006). However, these blended materials do not satisfactorily address the issues of TPS moisture sensitivity. Only a few papers have considered lamination (Averous, 2009; Fabra et al., 2016a; Martin et al., 2001). In Averous (2009) and Martin et al. (2001), PHBV-TPS laminated materials were produced as part of a broader study into the preparation of biodegradable, multi-layer TPS films, however, the PHBV coating was not looked at in detail. Only Fabra et al. (2016a) and Fabra et al. (2016b) have focused specifically on layered PHB-TPS materials, indicating that PHB may be a suitable coating for TPS with respect to an improvement in the barrier properties of TPS over time.

Whilst the results of the work so far are promising, they are still preliminary, with a need for a more focused investigation of the ability of PHA to reduce the water uptake of TPS in order to maintain its barrier properties. This provides the platform for the current work, which, for the first time:

- Assesses the suitability of Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) as a water barrier layer for TPS by quantifying and modelling water transfer for a range of relative humidities over time.
- Establishes a relationship between the reduction in water transfer due to the PHBV coating and improved barrier properties of the TPS.

This work contributes to an understanding of the suitability of PHAs as a coating for TPS in food packaging applications and the insights will inform further developments of PHA/TPS multi-layered materials.

3.2 **Experimental**

3.2.1 *Materials*

Polymers and plasticiser

High-amylose, hydroxypropylated corn starch (13% moisture content) was provided in powder form with no additives by Plantic Technologies Ltd (Australia) under the trade name Ecofilm. Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) with 5 mol% HV content was supplied in powder form with no additives by TianAn Biopolymer (China) under the trade name of ENMAT Y1000. Glycerol (98%) was obtained from Chem-supply, and chloroform (99%) was obtained from Merck. All products were used as received without further purification.

Thermoplastic starch (TPS) films

Extrusion: Corn starch, water and glycerol were pre-mixed using a polymer:water:glycerol weight ratio of 60:16:24. The pre-mixed formulation was then flood-fed to a co-rotating, twin-screw extruder (EuroLab 16, ThermoScientific) with a diameter of 16 mm, a length-to-diameter ratio of 40:1 and a melt-pump and flat-film die attachment (cross sectional dimension of 100 mm by 5 mm). The temperature profile and screw profile were as follows: Temperature (°C): from feed - 40, 75, 120, 140, 160, 140, 120, 110, 100, 95, 90, 90 – to die. Screw: from feed – 14 x forward screw, 9 x 30º forward, 3 x 60º reverse, 2 x forward screw, 9 x 60º forward, 3 x 60º reverse, 2 x forward screw, 9 x 30º forward, 3 x 60º reverse, 3 x 60º reverse, 2 x forward screw, 5 x 30º forward, 6 x 90º, 6 x forward screw – to die. These parameters were found to provide optimum conditions for ease of processing to produce TPS films with a thickness of 617 ± 32 μm.

Melt-press: To produce TPS films suitable for barrier property testing, the extruded sheets were melt-pressed to produce TPS films of thickness 80 µm to 100 µm. In this process, the as extruded sheets were first cut into 4 cm by 4 cm samples and stored at 5% relative humidity (RH) (See **Table 3-1**) for one week to minimise their water content, before they were placed between two Teflon coated plates into a pre-heated melt-press (160 °C). The samples were then held at 160 °C for 1 minute with no pressure. The pressure was then increased to 20 metric tonnes over 1 minute, and then retained at 20 metric tonnes for 1 minute. The sample was rapidly cooled to 100 °C whilst under pressure and then further cooled on the bench. This produced TPS films of thickness 80 µm to 100 µm.

Table 3-1: Saturated salt solution and corresponding equilibrium relative humidity of the storage chamber

Salt	Drierite	LiCl	CH_3COOK	$MgCl_2$	$Mg(NO_2)_2.6H2O$	NaCl	KNO_3
RH (%)	5	17	28	36	56	75	94

RH = relative humidity

PHBV films

Extrusion followed by melt-press (Extrusion-MP): PHBV was dried at 105 °C in a vacuum oven at a gauge pressure of -80 kPa for 24 h. The dried PHBV was then flood-fed to a co-rotating, twin-screw extruder (EuroLab 16, ThermoScientific) with a diameter of 16 mm, a length-to-diameter ratio of 40:1 and a slit die attachment (cross sectional dimension of 13 mm by 2 mm). The screw profile consisted of forward conveying elements with no mixing zones and the temperature profile was as follows: Temperature (°C): from feed - 150, 175, 180, 180, 165, 164, 163, 162, 161, 160 – to die. The resulting product was cut into 6 cm length sections and placed between two Teflon coated plates into a pre-heated melt-press (180 °C). The samples were held at 180 °C for 1 minute with no pressure. The pressure was then increased to 20 metric tonnes over 1 minute, and then retained at 20 metric tonnes for 1 minute. The sample was then rapidly cooled to 100 °C whilst under pressure and then further cooled on the bench. This produced PHA films with a thickness of approximately 115 ± 9 µm. Just melt-pressing of the PHBV powder was trialled, however, the required density of material could not be achieved, which led to the films having holes in them, thus the above method was adopted.
Casting: Casting was used to obtain thinner films than the ones obtained by extrusion followed by melt-press. An 8% w/v solution of PHBV in chloroform was heated at 60 °C for 3 hours. The solution was then cast on a polished glass plate with a doctor blade system and the solvent slowly evaporated (~30 mins). Sheets of either 31 ± 7 µm or 91 ± 9 µm thickness were produced depending on the volume used.

Blended films

Production of 10% PHBV/TPS w/w blends and 20% PHBV/TPS w/w blends was achieved in a similar manner to the production of extruded TPS films. The starch premix was first combined with PHBV powder using a PHBV:premix weight ratio of 10:90 (10% PHBV sample) or 20:80 (20% PHBV sample). The same screw profile was used as for the production of TPS films, whilst the temperature (°C) profile was adjusted as follows: from feed - 40, 75, 120, 140, 160, 180, 140, 110, 100, 95, 90, 90 – to die. This produced blended films with a thickness of 500 ± 32 μm.

Multi-layer films

TPS films as produced via extrusion, and PHBV films as produced via both extrusion-MP and casting, were used to produce the multi-layer films. All films were stored at 5% RH (See **Table 3-1**) for 1 week prior to the production of multi-layer films. The TPS was cut into 2cm by 2cm (~0.5 g) squares and placed between two PHBV sheets (equivalent thicknesses) in a pre-heated melt-press (180 °C). The pressure was immediately increased to 0.25 metric tonnes (i.e. the minimum pressure of the melt-press) and the samples were maintained at this pressure and 180 °C for 1 minute. The samples were then rapidly cooled to 100 °C whilst under pressure and then further cooled on the bench.

Samples were equilibrated with no further modification at 5% RH (see **Table 3-1**) for three days, before the exposed starch edges were sealed with high-vacuum, water resistant grease. The exact thicknesses of the PHBV and TPS films were only determined after a sample had been used for its intended experiment. At the end of an experiment, the samples were delaminated (i.e. the PHBV films were manually peeled off the starch layers) and the thickness of the films was recorded.

Details of materials for each experiment

For each experiment, the details of the materials produced, the RH of equilibration and the RH of storage during the experiment are recorded in **Table 3-2**. **Figure 3-1** provides a simple illustration of the experimental set-up. All experiments were performed at 23 °C.

Table 3-2: Details of the materials produced for the experimental work and relative humidity values

Experiment	Films types assessed	RH* during equilibration	RH during storage
Oxygen permeability of TPS films	a) Melt-press TPS film	2 films at 5%, 2 films at 35%, 2 films at 55%, 2 films at 75%	N/A
Moisture content of films after 2 weeks storage at different RHs	a) 115 µm PHA film, b) Extruded TPS film, c) Extruded TPS film with 115 µm PHA coating	For each film type: 3 samples at 5%	For each film type: 3 samples at 17%, 3 samples at 28%, 3 samples at 36%, 3 samples at 56%, 3 samples at 75%, 3 samples at 94%
Change in moisture content of films over time	a) Extruded TPS film, b) Extruded TPS film with 31 µm PHA coating c) Extruded TPS film with 91 µm PHA coating d) Extruded TPS film with 115 µm PHA coating	For each film type: 3 samples at 5%	For each film type: 3 samples at 75%
Effect of PHA blend as core layer on adhesion at high humidity	a) Extruded TPS film with 115 µm PHA coating, b) Extruded 10%PHA/TPS blend film with 115 µm PHA coating c) Extruded 20%PHA/TPS blend film with 115 µm PHA coating	For each film type: 3 samples at 5%	For each film type: 3 samples at 94%

*RH = relative humidity

3.2.2 *Methods*

Film thickness determination

To determine the average thickness of the films produced via a specific method, the thickness of three separate films were recorded in five places using a digital micrometer, and the average calculated. To determine the thickness of specific films used for barrier testing, the thickness of a film was recorded in five places using a digital micrometer, and the average calculated.

Equilibration and relative humidity of storage

Prior to all experiments, films were equilibrated at 5% RH for 1 week at 23 °C. Controlled humidity environments were established using saturated salt solutions as presented in **Table 3-1**. Relative humidities were monitored with a Hobo data logger which has an inbuilt relative humidity sensor.

Oxygen permeability

The oxygen permeability in mol.m/(m^2.s.Pa) was derived from the oxygen transmission rate in cm^3/(m^2.day) recorded using an OX-TRAN® Model 2/21 oxygen transmission rate analyser (MOCON, USA), following ASTM standard D3985-05. Experiments were carried out at 24 °C and the humidity set as desired. Samples were conditioned in an RH environment corresponding to the desired RH of permeability testing for 1 week prior to testing. Once mounted in the OX-TRAN® the samples were then conditioned in nitrogen for 1 hour before exposure to an oxygen flow of 20 mL/min. The exposure area during the test was 5 cm^2 for each sample. The measurements were performed in duplicate. In order to obtain the oxygen permeability, the oxygen transmission rate was multiplied by the film thickness and divided by the pressure difference of oxygen (101 kPa).

Moisture content

In most cases, moisture content (%) was directly determined using a Mettler-Toledo moisture analyser. Only the moisture content of the TPS layer was measured and all measurements were repeated in triplicate. In the case of multi-layered films, the grease was first wiped off and then the PHBV layer manually peeled off the TPS layer prior to testing.

When investigating the change in moisture content of the TPS over time, the moisture content was indirectly calculated using equation 3-1;

$$MC\ (\%) = \frac{wet\ starch\ (g) - dry\ starch\ (g)}{wet\ starch\ (g)} \times\ 100\ \% \qquad \text{(Eq 3-1)}$$

Multi-layer samples of TPS coated in a variety of PHA thicknesses were prepared and the initial mass (i.e. mass of TPS, PHBV and grease) of each sample was recorded. The initial MC of three TPS samples was determined using the Mettler-Toledo moisture analyser after being cleaned of grease and the PHBV manually peeled off. The remaining samples were then placed in a 75% RH environment and their mass recorded regularly for 4 weeks. All samples were run in triplicate to reduce the possibility of error arising from film defects. At the end of the experiment, each sample was weighed, before it was wiped clean of grease and delaminated and the mass and moisture content of the TPS determined. This allowed the mass of dry starch to be calculated using equation 3-1, as well as the mass of grease and PHA included in the sample. Moisture content was then calculated for all samples by subtracting the mass of PHA and grease from the recorded weights and substituting the experimentally determined mass of wet TPS at each time point and the calculated mass of dry TPS into the above equation.

Model development

A simple model was developed based on Fick's first law of diffusion. The purpose of this model was to gain a quantitative understanding of the rates of water transport across PHBV films of different thicknesses and the resulting accumulation of water in the inner TPS film. The model was fit to the experimental data for the moisture content of the TPS film over time when stored at 75% relative humidity.

Key simplifying assumptions for the model were as follows:

1. Diffusion through the inner TPS film is rapid, so the resistance to mass transfer due to diffusion through the TPS film can be neglected. Consequently, it is assumed that the concentration of water is homogenous within the TPS film, and diffusion across the PHBV film is rate determining.
2. The apparent diffusivity of the PHBV is constant. Although not strictly true, with diffusivity varying with water concentration (Follain et al., 2014), the aim of the model is just to give a quick way to quantitatively compare the apparent rate of water permeation between the different materials and the assumption was deemed to be appropriate for this purpose.
3. The concentration profile across the PHBV film is linear.
4. The concentration profile across the PHBV film is established rapidly, i.e. there is no lag time.
5. Conservation of mass, i.e. water transported across the PHBV film equals the accumulation of water within the inner TPS film.

By applying these assumptions, Fick's 1st law of diffusion can be applied to model the water transport across the PHBV film and simplifies to equation 3-2;

$$N = -D_A\left(\frac{C_1 - C_2}{L}\right) \qquad \text{(Eq 3-2)}$$

where C_1 and C_2 are the concentrations of water on either side of the PHBV film, with C_1 being the outside of the film and C_2 being the inside of the film adjacent to the TPS layer (see **Figure 3-1**). N is the flux across the film, L is thickness of the PHBV film and D_A is the apparent diffusion coefficient. Dirichlet boundary conditions were defined based on the equilibrium expressions given in equation 3-3 and equation 3-4;

$$C_1(0,t) = S \times p \qquad \text{(Eq 3-3)}$$

$$C_2(L,t) = K \times C_{st}(L,t) \qquad \text{(Eq 3-4)}$$

where S is the solubility of water in PHBV, p is the partial pressure of water in the air adjacent to the PHBV film, K is the partition coefficient of water between starch and PHA and C_{st} is the concentration of water in the starch film (kg water/kg dry starch), assumed to be constant throughout the starch film (assumption 1).

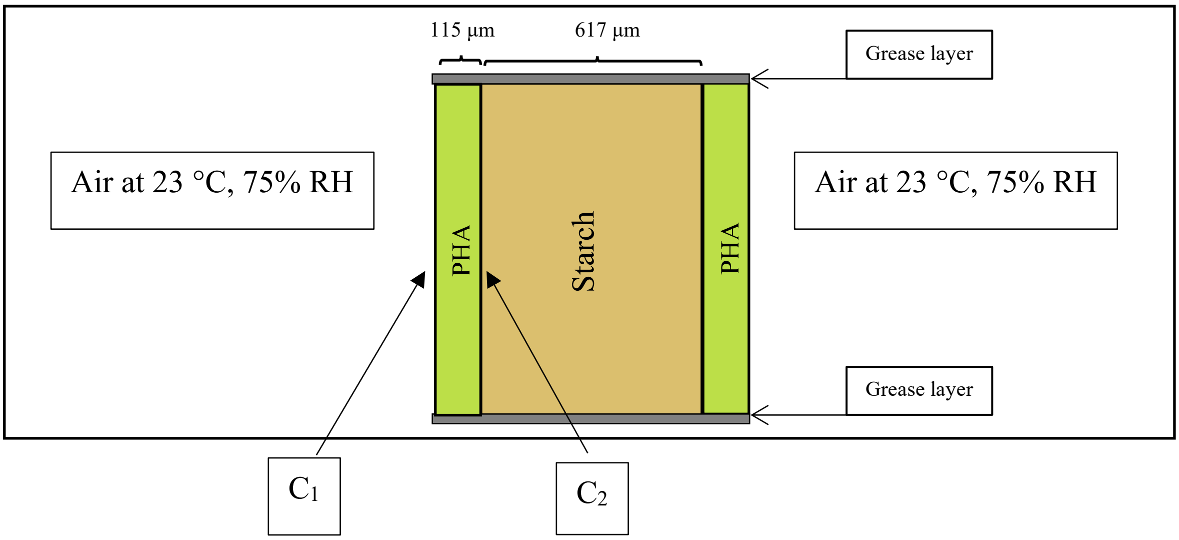


Figure 3-1: Cross-sectional illustration of the experimental set-up and modelling parameters

Note to Figure 3-1: Example shown for a 115 µm PHA layered sample.

The initial conditions were defined based on the experimentally determined initial TPS moisture content, shown by equation 3-5;

$$C_2(L,0) = K \times C_{st}(L,0) \tag{Eq 3-5}$$

Since permeability is commonly reported for water transport across polymers, D×S is replaced with the permeability coefficient, P, as presented in equation 3-6 and assumed to be constant (assumption 2);

$$N = -\left(\frac{Pp - DKC_{st}}{L}\right) \tag{Eq 3-6}$$

At equilibrium, there is no net flux and the concentration within the TPS reaches its equilibrium concentration, $C_{st,eq}$. The lumped parameter, DK, was solved based on the initial guess of the permeability and the experimentally determined equilibrium concentration within the TPS as presented in equation 3-7;

$$DK = \frac{Pp}{C_{st,eq}} \tag{Eq 3-7}$$

The initial flux was estimated based the experimentally determined initial moisture content of the starch film. The concentration of water within the TPS film at the subsequent time point was then estimated assuming all the water transferred accumulates within the TPS layer, which is at constant moisture content as summarized in equation 3-8;

$$C_{st,i} = C_{st,i-1} + \frac{N_{i-1} \times \Delta t \times A}{M_{st}} \tag{Eq 3-8}$$

where Δt is the time step set for the discrete model, A is the total area for mass transfer (both sides of the film) and M_{st} is the experimentally determined mass of dry starch. The concentration of water can then be converted into the moisture content of the TPS layer as presented equation 3-9;

$$MC_{st,i} = \frac{C_{st,i}}{C_{st,i}+1} \times 100\ \% \qquad \text{(Eq 3-9)}$$

The model was solved numerically in Microsoft Excel using an explicit finite difference approach with a time step of 2 hours and fit to the experimental data by optimising the permeability coefficient (P) for water through the PHBV film using a least squares approach.

3.3 Results and discussion

3.3.1 *Oxygen barrier properties of the TPS film*

The results presented in **Figure 3-2** demonstrate that the oxygen barrier properties of the TPS films produced in this work decrease with increasing moisture content (MC) (where the corresponding moisture sorption profile is presented in **Table 3-3** for reference). Two measurements were recorded for each film and these are presented as 'TPS sample 1' and 'TPS sample 2'. The average permeability of an extruded and then melt-pressed PHBV film is included to indicate the moisture content at which TPS becomes a worse oxygen barrier than the PHBV film. Values for the permeability of polyethylene (PE) are also presented for comparison and were taken from literature (Kurek et al., 2012). The most conservative value for PE permeability is presented although there are also examples where the permeability of PE is found to be an order of magnitude larger (Follain et al., 2014; Mousavi et al., 2010). It was found that at ~23% moisture content, the oxygen barrier properties of the TPS film are poorer than the PHBV films produced in this work. The sensitivity of starch films to moisture content, as well as other plasticisers such as glycerol, has been previously observed (Chang et al., 2000; Dole et al., 2004; Forssell et al., 2002; Gaudin et al., 2000; Krochta and De Mulder-Johnston, 1997). The change in properties of the TPS film is thought to be due to increasing moisture content leading to relaxation of the polymer structure and an increase in free volume making it easier for a permeant molecule to travel through the structure (i.e. diffusivity increases) (Benczedi, 1999; Miller and Krotchta, 1997). This is directly related to the glass transition temperature of the starch sample (Chang et al., 2000; Dole et al., 2004; Forssell et al., 2002). Shogren (1997) suggests that gelatinised starch remains strong up to 22% MC at which point room temperature corresponds to the glass transition temperature and the polymer exists in a rubbery state leading to a reduction in mechanical and barrier properties. The results presented in the current work support this explanation. The moisture content relating to a change in glass transition temperature will be different for different starch systems (depending on the amount of plasticiser etc. present in the starch). It can be inferred that for the TPS used in this study, the moisture content corresponding to a room temperature glass transition is between 14% and 23% (as presented in **Figure 3-2**). Thus, the films as produced in this work require a protective layer to reduce their moisture uptake and subsequent loss of barrier properties. Of note is that although it would have been useful to be able to compare the measured oxygen permeability results to industry best practice, the industry does not appear to have defined such a target at this point. In the absence of this, it is assumed that barrier properties just need to be as good as possible.

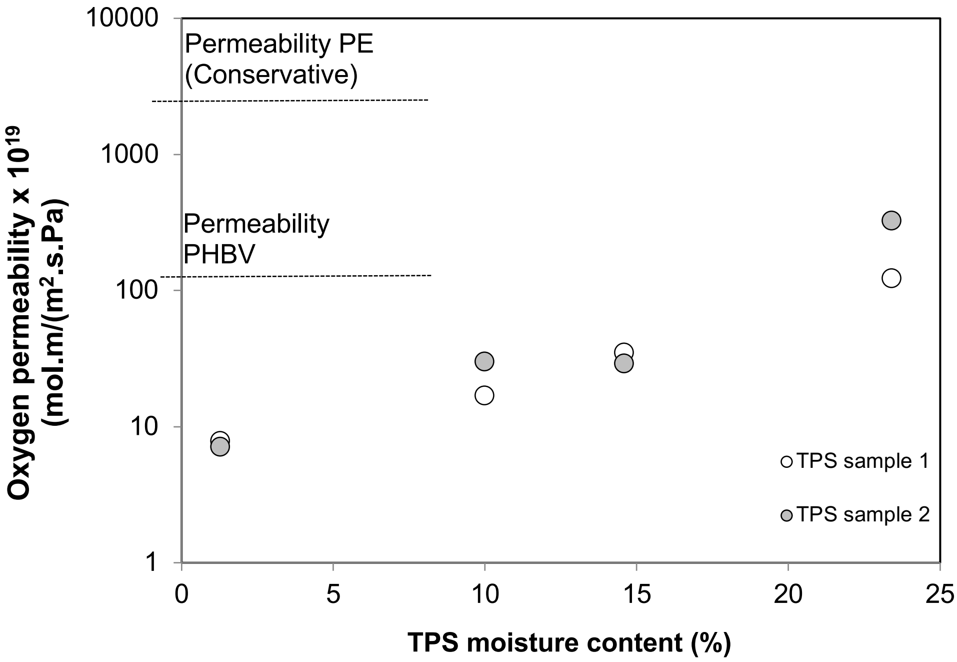


Figure 3-2: Oxygen permeability of TPS films at different moisture contents

Table 3-3: The relationship between relative humidity of storage and equilibrium moisture content of the TPS films

Note to Table 3-3: Moisture content values correspond to those presented in **Figure 3-2**. Error values are standard deviations.

Relative humidity of storage	**5%**	**35%**	**55%**	**75%**
Equilibrium moisture content (%)	1.3 ± 0.1	10 ± 0.3	15 ± 0.3	23 ± 0.2

3.3.2 *Influence of a PHA coating*

As illustrated in **Figure 3-3**, a PHA coating can reduce the rate of water transfer into a TPS film. The samples tested were either uncoated TPS or TPS coated with 115 µm PHBV. Within the 'zone of confidence' a TPS sample >500 µm thick, coated in a layer of PHBV > 110 µm thick, and stored under the specified conditions should exhibit good oxygen barrier properties. Outside of these conditions (i.e. longer time period, thinner TPS or PHBV, or higher relative humidity of storage) it is expected that the oxygen barrier properties will be negatively affected. The results suggest that a PHA coating can extend the period of time over which a TPS film will exhibit good barrier properties. This expands on the outcomes presented by both Fabra et al. (2016a) and Fabra et al. (2016b) by considering a range of relative humidities of storage. In the current results, the water vapour sorption isotherm of the uncoated TPS is characteristic of a high-glycerol content film and matches with the isotherms presented in literature (Godbillot et al., 2006). It is also shown that over a period of two weeks a PHBV-coated TPS film had a lower moisture content under storage at relative humidities of up to 75%. Importantly, the coating retains the TPS at a moisture content below that at which barrier properties are compromised (as determined from the results presented in **Figure 3-2**). Under storage at a relative humidity of 93%, the PHBV had delaminated from the TPS, which explains why under these conditions the moisture content of both the uncoated and coated TPS is very high. The results presented in **Figure 3-3** correspond closely to the results of a study performed by Dole et al. (2005) using TPS coated in LDPE, indicating that PHBV could be a suitable alternative coating. Dole et al. (2005) also noted that at storage RH of above 90% delamination of the multi-layered materials occurred and moisture uptake of all materials was high, supporting the observations of the current experiment.

3.3.3 *Change in moisture content over time*

Storage of the multi-layer films at 75% RH was selected for further investigation as it is the scenario in which TPS by itself is above the critical MC leading to loss of barrier properties, but the layered material is below (**Figure 3-3**). Also, issues of delamination are not yet apparent at 75% RH.

The moisture content of the TPS film over time was monitored and correlated to changes in O_2 barrier properties. Compared to directly measuring oxygen barrier properties of the layered materials, this provides a more sensitive way to track the impact of the PHA coating (and different PHA thicknesses). Specifically, it allows one to monitor and understand the changes that are occurring in the TPS, even if over the given time period a significant shift in the TPS structure (and therefore barrier properties) does not occur.

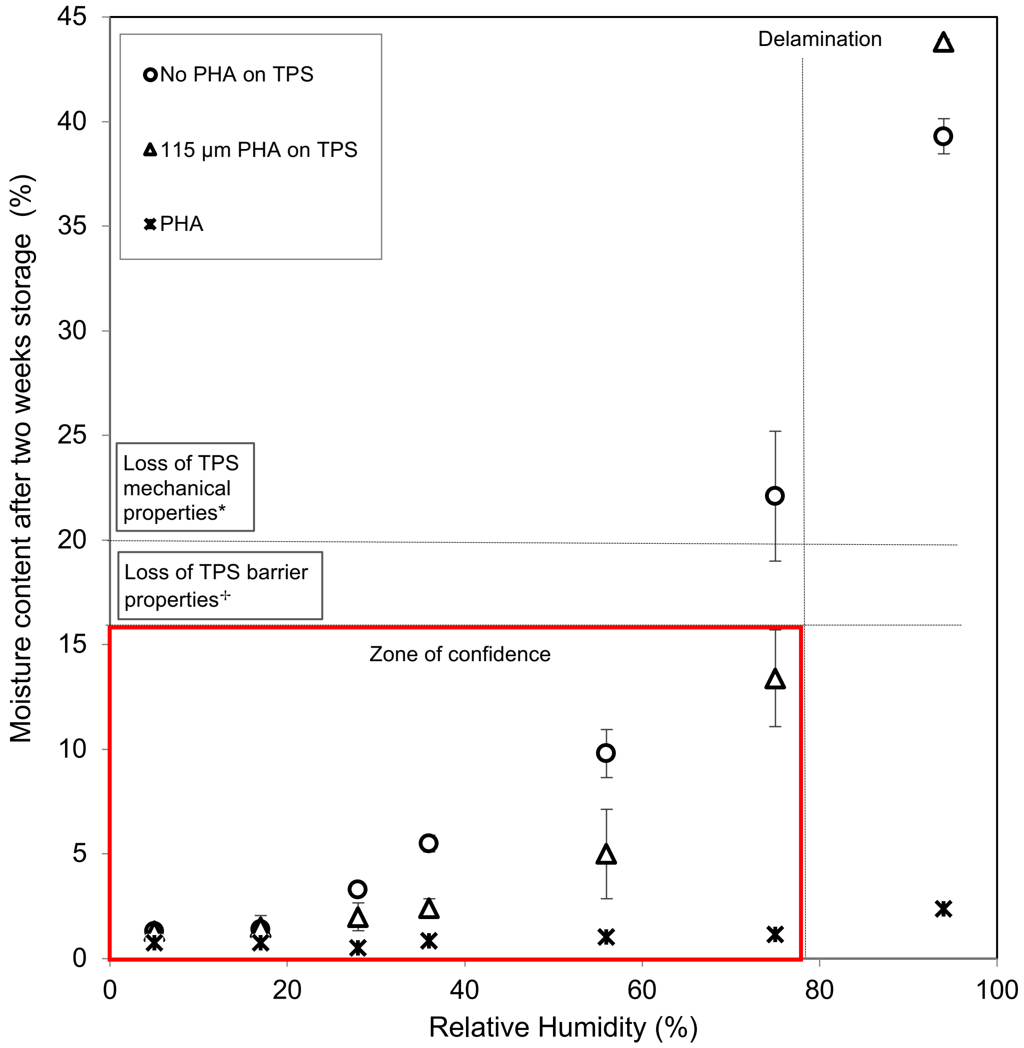


Figure 3-3: The moisture content of a TPS film after two weeks storage at a variety of relative humidities

Note to Figure 3-3: * Shogren (1997) +Experimentally determined (see **Figure 3-2**). Error bars are standard deviation.

Measured moisture content over time

Figure 3-4 illustrates how the moisture content of a variety of layered materials changes over time when stored at 75% RH after equilibration at 5% RH. The samples tested included uncoated TPS, TPS coated in 31 µm PHBV, TPS coated in 91µm PHBV and TPS coated in 115 µm PHBV. This expands on the results presented in **Figure 3-3** and is the first quantification of performance over time for a PHA-coated TPS film. Upon exposure to a 75% RH environment, the moisture content of TPS with no coating layer rapidly increases to reach equilibrium within two days (**Figure 3-4**). With a PHBV coating, the increase in moisture content is slowed and it is evident that the thickness of the coating influences the rate of change of moisture content. For the sample coated with a 31 µm PHBV film, although the rate of moisture uptake is slowed, after 13 days the MC of the TPS is no longer different to that of the uncoated sample. However, for a coating thickness of 91 to 115 µm the moisture content of the TPS remains significantly lower than both the uncoated TPS and the 31 μm PHBV-coated TPS for the duration of the experiment (>25 days). The TPS film in the 91 μm multi-layer system had a slightly higher dry mass than the TPS in the 115 μm multi-layer system which is why it sits lower on the plot of moisture content versus time.

From these results, it can be concluded that the thicker the PHBV coating, the longer the TPS will retain its barrier properties. Although it should be noted that, if given enough time, it is expected that the moisture content of all the layered TPS samples will tend towards that of the equilibrated uncoated TPS. This is because thermodynamics dictates that moisture migration will continue until all elements of the system attain the same water activity (with the final water activity a function of the humidity of the environment but independent of the layers within a material) (Ergun et al., 2010).

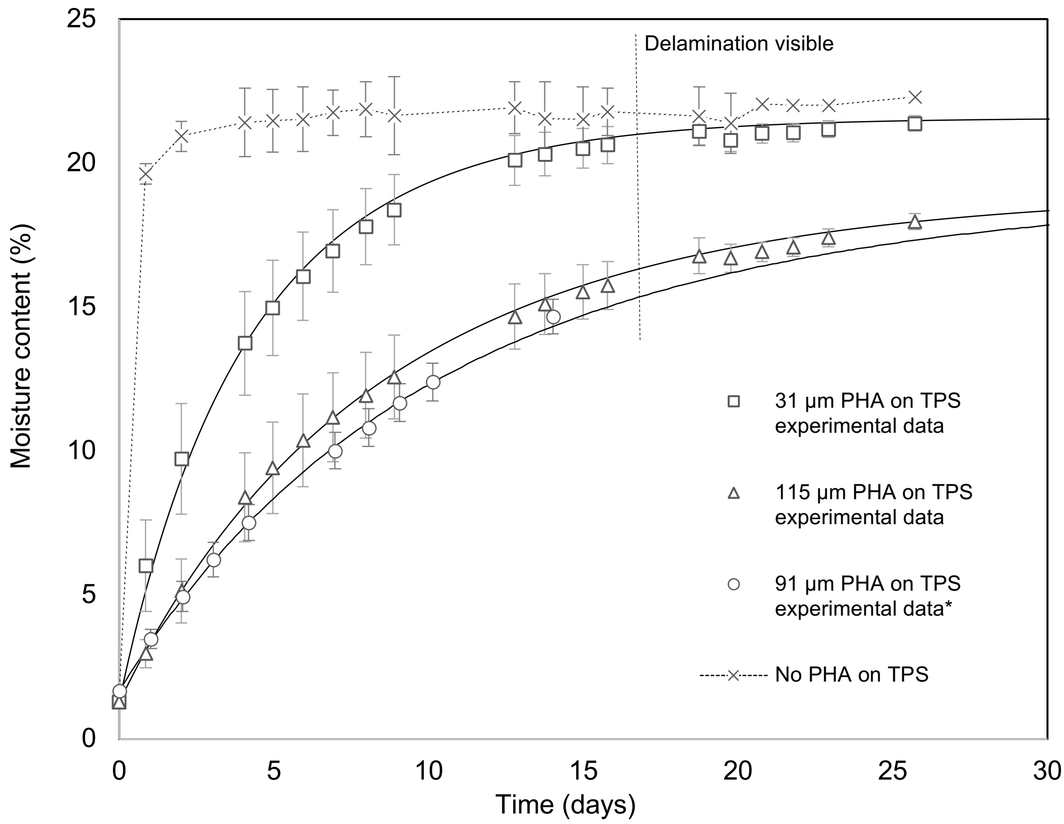


Figure 3-4: Change in the moisture content over time for a PHBV-coated TPS film

Note to Figure 3-4: Error bars are standard deviation. The results of fitting Fick's law are presented as solid lines. *The results for the 91 μm PHBV-coated samples are lower than that for the 115 μm PHBV samples due to the 91 μm PHBV-coated samples containing a higher starting mass of TPS.

Modelling results

The curves as produced by the model closely match those of the experimental data as illustrated in **Figure 3-4**. However, in order for the model to fit closely to the experimental results both the equilibrium moisture content of the TPS layer and the permeability of the PHBV film need to be optimized (See **Table 3-4**).

The TPS coated in 91 μm or 115 μm PHBV film appears to tend towards an equilibrium moisture content lower than that of uncoated or 31 μm PHBV-coated TPS, despite the fact that all the films should theoretically reach the same equilibrium moisture content (as discussed in 'measured moisture content over time' section). However, here we suggest that the presence of a thick and therefore rigid PHBV layer may initially limit the swelling of the TPS which would limit moisture uptake until delamination occurs, after which point the data would be expected to tend towards the uncoated equilibrium moisture content.

In the literature it has been shown that TPS can swell up to 90% in the thickness dimension and up to 70% in the area of the face of the film when immersed in water (Russo et al., 2007). This indicates that swelling of the TPS plays an important role in allowing it to approach maximum water content. In the work by Russo et al. (2007) it is also noted that water absorption is faster once the solvent front within the TPS has penetrated the film and the swelling is no longer restricted by a stiff inner core. It then appears logical that a stiff outer coating could provide the same restriction to the swelling of the film and thus reduce water uptake.

Table 3-4: Key parameters for the model

Parameter	31 μm PHBV	91 μm PHBV	115 μm PHBV	Method
Thickness of the PHBV (mm)	0.031	0.091	0.115	Experimentally determined
Equilibrium moisture content of the TPS (g water/ g wet TPS, %)	21.6	19.0	19.0	Optimised for curve fit
Permeability of the PHBV (g.mm/(m^2.h.Pa))	1.45 x 10^{-5}	1.88 x 10^{-5}	2.15 x 10^{-5}	Optimised for curve fit
Average mass of dry TPS (g)	0.786	0.97	0.718	Experimentally determined
Average area of the film (m^2)	0.0015	0.0015	0.0015	Experimentally determined

In regard to the permeability of the PHBV films, the current modelling parameters show the permeability of the film increasing as the thickness increases (**Table 3-4**). Whilst in theory the permeability of a film should be independent of thickness, in practice the permeability can show signs of thickness dependence (Hwang and Kammermeyer, 1974). In the current case, one of the assumptions for the model is that the TPS provides negligible resistance to water transfer. However, as shown in **Table 3-5**, this assumption becomes more appropriate as the PHBV film becomes thicker.

For the thinner films, the contribution of the TPS resistance is more significant and therefore, when the full resistance of the film is ascribed to only to the PHBV layer (as is done in the model) its apparent permeability is lower.

Table 3-5: Contribution of the TPS film's resistance to water transfer

Film	Thickness (T) (mm)	Permeability coefficient* (P) (g.mm/m2.h.Pa)	Permeance (g/m2.h.Pa) [=P/T]	Resistance (R) [=1/permeance]	% overall resistance from TPS film compared to PHBV film [=R(TPS)/(R(TPS)+R(PHBV))]
TPS	0.313	5.6 x 10^{-3}	1.8 x 10^{-2}	56	–
PHBV	0.031	2.5 x 10^{-5}	8.1 x 10^{-4}	1.2 x 10^{3}	4.3
PHBV	0.091	2.5 x 10^{-5}	2.7 x 10^{-4}	3.6 x 10^{3}	1.5
PHBV	0.115	2.5 x 10^{-5}	2.2 x 10^{-4}	4.6 x 103	1.2

* as measured by Fabra et al. (2013) and Fabra et al. (2016a)

3.3.4 *Adhesion and storage at high RH*

Adhesion is known to be weak between PHBV and TPS polymer films, particularly for multi-layer films formed by melt-pressing (Martin et al., 2001). This work did not set out to form intimately adhered materials, so the materials were not optimised in this regard. For most of the experiments, adhesion was found to be adequate for the experimental purposes. It was only storage under very high humidities, (**Figure 3-3**), or over long-time periods (**Figure 3-4**) that adhesion became an issue, with the PHA completely delaminating from the TPS. Dole et al. (2005) also observed this for PE layered TPS stored at high humidity (even with the assistance of a tie layer) and Martin et al. (2001) observed this for PHBV layered TPS materials stored for long periods of time. With a view to tracking the delamination of the PHBV from a film stored at 94% RH over a period of weeks, an experiment similar to that performed for the 75% RH environment (**Figure 3-4**) was set-up. However, within 24 hours delamination had occurred and the TPS moisture content had rapidly increased. This indicates that for storage at high humidities, adhesion will need to be a primary focus and that the initial production techniques explored in this study were insufficient. Further, preliminary experiments were therefore performed to investigate this issue, following an innovative solution presented by Martin et al. (2001) for improving the adhesion between TPS and other biopolymers. They showed that by blending a small amount of the coating biopolymer into the inner TPS layer (in their case PCL, PLA or PEA), subsequent adhesion between the TPS layer and the coating layer was improved, although, the idea had not yet been explored for PHBV. Through using 10% PHBV/TPS and 20% PHBV/TPS blends as the inner layers for forming the PHBV-coated materials, the current work determined that this concept is also valid for PHBV. The onset of delamination was delayed by three to four days (as noted by visual observation). Further investigation will be required to improve this result and extend the adhesion time to weeks.

3.4 Conclusion

This is the first work to have shown that a PHBV coating can reduce the water uptake of a TPS film, extending the time period over which the multi-layer film will exhibit good oxygen barrier properties. For PHBV coating thicknesses of 91 to 115 μm the moisture content of the TPS remains significantly lower than uncoated TPS for the duration of the experiment (>25 days), and for the first two weeks remains below the moisture content at which oxygen barrier properties are compromised. This work has also demonstrated that the process can be modelled by Fickian diffusion and has shown that the thickness of the PHBV film will affect the rate of water uptake. This builds on the currently limited knowledge regarding the suitability of a multi-layered PHBV/TPS film for food packaging applications. Building on the work presented here, further work is being pursued in the area of layered PHBV/TPS blends with a focus on adhesion.

CHAPTER 4 Flow-induced migration to produce multi-layer films

Chapter Summary

This chapter presents the second of two research activities that relate to the first objective of the material properties theme: '[To] *Assess the suitability of PHA as a water barrier layer for TPS*'.
This work evolved from the work presented in **Chapter 3**, where it was identified that poor adhesion between the PHA and TPS layers was an issue that would limit the use of such a material. The results presented in this chapter show that the differences in the melt viscosities of PHA and TPS leads to gradients in phase composition through an extruded blend profile, that could potentially be used to produce a multi-layered material during a single pass extrusion. This could help to alleviate adhesion issues.

4.1 **Introduction**

To reduce the impact of plastic use, improve recycling rates, and move towards a more sustainable plastics system, it is increasingly important to redesign plastic packaging (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). In particular, multi-layer food packaging is a target product for redesign, as it has desirable material properties (such as high gas and water barrier properties (Butler and Morris, 2013)) but is a single use material which is difficult to recycle (Barnes et al., 2009). One option for the redesign of multi-layered materials is to produce biodegradable multi-layer plastics. This means the high-barrier functionality of the product can be retained, but issues of recycling can be overcome by allowing biological processing of the packaging at the end-of-life. This is particularly relevant if the packaging is contaminated by organics and hence unsuitable for direct thermal recycling. Recently, it has been shown that polyhydroxyalkanoate (PHA) coated thermoplastic starch (TPS) materials could be interesting target materials, with the PHA reducing the water uptake of the TPS, meaning it retains its desirable high-oxygen barrier properties (Dilkes-Hoffman et al., 2018b) (**Chapter 3**).

However, one of the problems restricting the further development of such materials is the lack of affinity between the hydrophilic TPS and hydrophobic PHA interfaces. Only weak adhesion exists between the layers, which leads to delamination and a subsequent loss of barrier properties of the TPS over time (Dilkes-Hoffman et al., 2018b; Martin et al., 2001). In this work, flow-induced migration during co-extrusion of PHA and TPS is presented as a means to address this problem. In flow-induced migration, blended polymer mixtures phase separate due to the shear forces within an extruder die, driving polymer migration into different zones of the extrudate. This can lead to the development of position-dependent gradients (Agarwal et al., 1994). In other words, from an initially blended polymer mixture, one polymer migrates to dominate the mid-plane, whilst another concentrates along the surface. In theory, this means that this method could be used to produce a material with a multi-layered structure but without the adhesion issues, given that the surface polymer will be anchored as that polymer is likely also distributed in part through the bulk (Martin et al., 2001).

Flow-induced migration has been studied for many years. In the mid-1970s, Lee and White (1974, 1975) placed LDPE and HDPE side-by-side in capillary rheometers and noted that the lower viscosity melts migrated outwards to begin encapsulating the higher viscosity melts. They also noted that the rate of encapsulation increased with increasing viscosity differences and longer residence times. Later, Karagiannis et al. (1990) presented experimental results and early theory using a polyolefin polymer identified as Dow Styron. They again noted that the degree of encapsulation is dependent on the viscosity ratio of the two melts, and that a longer flow path leads to a greater degree of encapsulation.

Bélard et al. (2009) were the first to try and exploit this phenomenon for the intentional creation of multi-layered plastic materials from a blended mixture. They performed combined extrusion experiments with TPS and polycaprolactone (PCL), and demonstrated the feasibility of producing multi-layered structures by flow-induced migration. In this case, the PCL migrated towards the walls whilst TPS remained in the mid-plane, and it was identified that the phase separation was driven by the molecular weight (chain length) of PCL which is linked to its molten state viscosity. This was a promising result. However, a PCL surface film of only 1 μm was achieved, and this may not be sufficient for protecting the TPS from moisture uptake, which would limit the material's use in food packaging.

Dorgan and Rorrer (2015) then provided the analytical theory to explain this phenomenon, based on principles of non-equilibrium thermodynamics. Using their model, they predicted that for viscous materials, the influence of a gradient in shear force will drive the migration of shorter chain lengths towards the wall and longer chain lengths towards the mid-plane. Components with lower viscosity are also expected to reside closer to the wall where the shear rate is highest, in order to reduce free energy (shear stress being a function of viscosity) (Dorgan and Rorrer, 2015).

Although there is no previous research that directly considers the application of flow-induced migration to forming PHA and TPS layered materials, the theory and experimental results appear to support the concept. In fact, Willett and Shogren (2002) observed unintentional surface enrichment of PHA when producing a PHA and starch foam, which gives confidence that Bélard's methodology could be adapted to a PHA and TPS system. Building on the experimental and theoretical work to date, the current research explores the feasibility of producing multi-layered TPS and PHA structures through flow-induced migration.

4.2 Methodology

The following material formulations and extrusion parameters were selected based on extensive testing to provide optimum ease of processing (e.g. testing of extruder screw profiles, temperature profiles, screw speed and barrel length, PHA addition point, and percentage PHA in the blend).

4.2.1 *Die development*

Based on the paper by Bélard et al. (2009), a custom-designed, 14.5 cm long, adjustable channel-thickness, extruder die was built. The length of the die was a compromise between the need for a long die (given that multiple publications noted a higher length to diameter ratio gives greater separation (Karagiannis et al., 1990; Lee and White, 1975, 1974; Schreiber et al., 1966; Schreiber and Storey, 1965; Tirrell and Malone, 1977)) but the knowledge that the polymer would need to move through the die based only on the force applied from the extruder screws at the entrance to the die (which would prove hard if the die was too long). The custom-designed die can be seen in **Figure 4-1A** and **Figure 4-1B**.

4.2.2 *Materials*

High-amylose (80%) corn starch (14% moisture content) with no additives was obtained in powder form (supplied by Ingredion). The 80% amylose starch was specifically selected given that a higher amylose content is associated with higher viscosity of the TPS melt, which should promote phase separation under shear conditions (Xie et al., 2009). Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) with 5 mol % HV content and a weight-average molecular weight ($\overline{M_w}$) of 590 kDa was supplied in powder form with no additives by TianAn Biopolymer (Ningbo, China) under the trade name of ENMAT Y1000. Glycerol (99% purity) was obtained from Chem-supply (Gillman, Australia). All products were used as received without further purification.

Starch mixture preparation: Corn starch (14% moisture content) and glycerol were pre-mixed using a wet-polymer:glycerol weight/weight ratio of 2:1. The mixture was placed in a sealed container for 12 hours before use, after which the moisture content of the total mixture was 17%.

PHA/starch mixture preparation: To the starch mixture, PHA powder was added and thoroughly mixed to give the desired PHA percentage (e.g. 30 g PHA added to 70 g starch mixture for the 30% PHA sample).

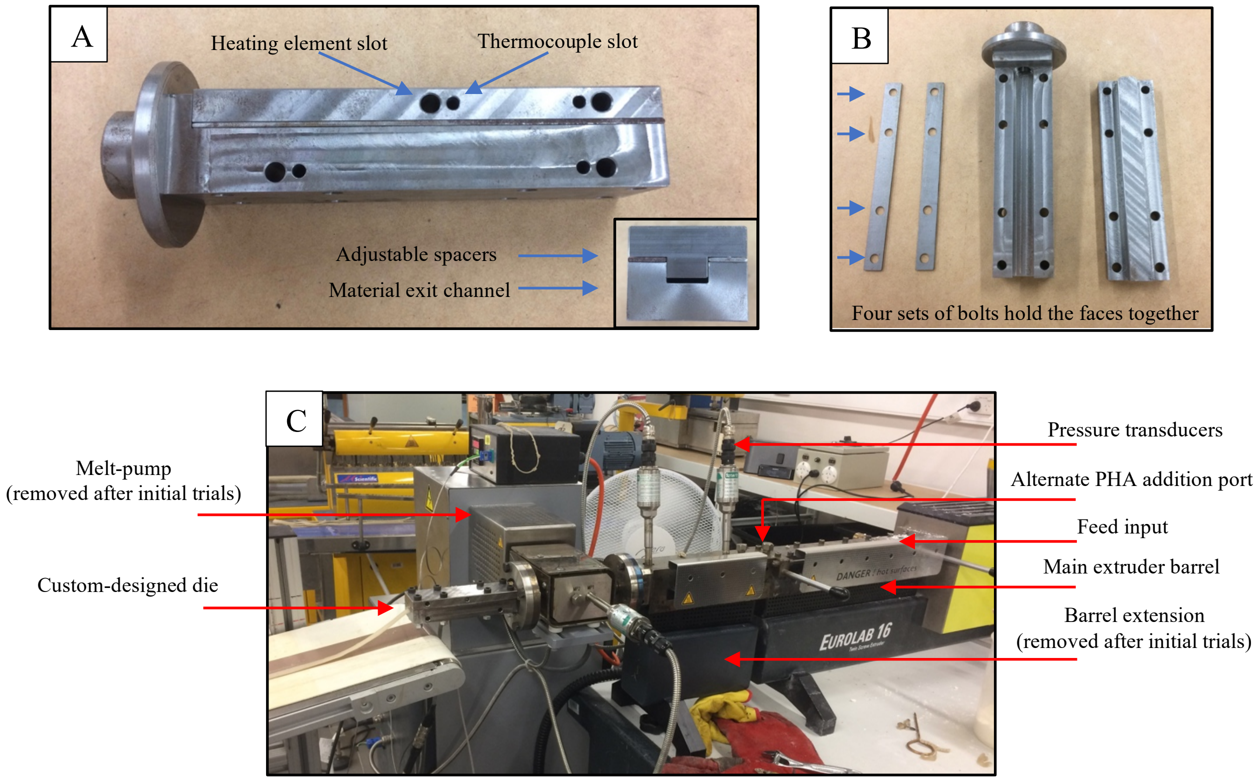


Figure 4-1: Custom-designed die

4.2.3 *Extrusion method*

The extrusion processing of the PHA/TPS blends was conducted using a co-rotating twin screw extruder (EuroLab 16, ThermoScientific) with a diameter of 16 mm, a length-to-diameter ratio 22:1 and the custom-made die attached with the exit channel set at 3.7 mm by 15 mm. After determining the optimal extrusion set-up to produce materials (e.g. only using the main extruder barrel, without the barrel extension or melt-pump, **Figure 4-1C**), three different procedures for PHA addition were trialed:

i) PHA/starch mixture added at the start of the barrel (PHA content ranging from 23% w/w to 50% w/w);

ii) Starch mixture added at the start of the barrel with PHA addition through a port at the end of the barrel;

iii) Starch mixture added at the start of the barrel with PHA addition at the end of the barrel and an extra mixing zone added in the extruder screws just before the die.

In each procedure, either the starch mixture, or the PHA/starch mixture (depending on the addition point for the PHA being trialed), was flood fed into the extruder, with the screw and temperature profiles being selected after extensive testing.

The final profiles are reported below, with the evolution of the profiles explained in results section 4.3.1.

For procedure i) and ii)

Screw: from feed—4 x forward screw, 9 x 30° forward, 3 x 60° forward, 3 x 60° reverse, 2 x forward screw, 5 x 60° forward, 4 x 30° forward, 3 x 60° forward, 3 x 60° reverse, 10 x forward screw – to die. Temperature profile (°C): from feed − 100, 160, 160, 170, 170, 155 —to die start − 145, 150, 125—to die end. Screw speed 120 rpm.

For procedure iii)

Screw: from feed—4 x forward screw, 9 x 30° forward, 3 x 60° forward, 3 x 60° reverse, 2 x forward screw, 5 x 60° forward, 4 x 30° forward, 3 x 60° forward, 3 x 60° reverse, 4 x 60° forward, 1 x forward screw – to die. Temperature profile (°C): from feed − 100, 160, 160, 170, 170, 155 —to die start − 145, 150, 125—to die end. Screw speed 120 rpm.

4.2.4 *Imaging*

Imaging using a microscope and iodine staining was used to investigate the location of the TPS and PHA phases in the material after extrusion. Cross-sections of the sample (~0.5 mm thick) were cut and placed in a vial with iodine crystals for five minutes before the face of the cross-section was observed via transmitted-light microscopy (**Figure 4-2**).

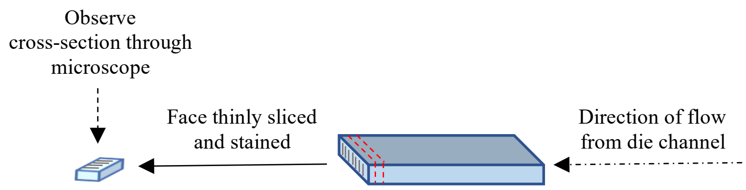


Figure 4-2: Imaging process

4.3 **Results and discussion**

4.3.1 *Processing parameters*

To produce multi-layered TPS and PHA films through flow-induced migration, a method and set of parameters that would allow blending of the PHA and starch (whilst simultaneously plasticising the starch) within the extruder barrel, but then facilitate the formation of a PHA surface layer as the material travelled along the length of the die, needed to be developed. There were two key challenges associated with this: firstly, to enable the material to pass along the length of the 14.5 cm die based only on the force applied from the extruder screws at the entrance to the die, and secondly, to ensure both PHA and starch were melted so that polymer migration could occur, without degrading the starch in the process.

With regard to addressing the first challenge: building on the methods developed in Dilkes-Hoffman et al. (2018b) (presented in **Chapter 3**), initially a long extruder barrel was used as well as a melt-pump. However, the melt-pump was removed after it was found that it reduced the pressure that could be applied to the material at the entrance to the die, meaning the polymer plug did not move through. The starch was also found to be plasticising too early in the extruder barrel, drying out before it reached the die, which again hindered its movement through it. This was resolved by removing the barrel extension. The result was that the shortest possible extruder barrel (35 cm) and screw were used. The use of the custom-designed die caused enough pressure to build-up at the entrance to the die channel to allow plasticisation of the starch to occur at the very end of the barrel and in the start of the die.

With regard to addressing the second challenge: this type of starch-blend processing strategy is relatively unreported to date, meaning there are no existing guidelines. Bélard et al. (2009) used polymers where differences in the melting points did not present a challenge; the potential degradation of starch was avoided given that PCL has a low melting point. Willett and Shogren (2002) were also using starch and PHA, but used a very cool temperature profile – with apparent success based on their reported results. However, in the current work, copying this profile resulted in unmelted PHA powder passing through the barrel. So, the starting point for the temperature profile used in this work was found by extruding PHA whilst slowly reducing the temperature of each heating block to identify the lower limit of the temperature profile that would still produce melted PHA. This temperature profile was then optimised for starch plasticisation. Glycerol was used as the plasticiser (instead of water) so that the temperature profile required for PHA melting could be used without producing steam, which caused pressure build-up in the extruder barrel.

Once these processing challenges had been solved, three different procedures for PHA addition were investigated (as defined in section 4.2.3). The two methods involving addition of PHA at the end of the barrel were not found to be effective. The first issue was that the two polymers did not appear to blend, with a TPS section and then an unmelted PHA section exiting the die. Adding an extra mixing zone in the screws, just after the point of PHA addition, did not solve the issue, appearing to cause too much mixing too late in the process, with a homogenous mixture exiting the die. Whilst these initial experiments did not produce the desired results, the concept of adding PHA at different points along the barrel should be further explored. In the present set-up there was only one possible addition point for the PHA and this was just before the entrance to the die, which appears to be too late in the process. An entrance point around half-way down the extruder barrel could be interesting to consider.

The most promising results came from the PHA/starch pre-blended mixtures (procedure I, as detailed in section 4.2.3, **Figure 4-3**). **Figure 4-3A** presents an image of pure TPS, stained with iodine and then observed under the microscope, as a comparison for the other images. **Figure 4-3B** then presents the material that was obtained when TPS was added to the extruder after it had been processing PHA. Here a distinct PHA skin can be seen forming, which demonstrates the preference of the PHA to travel along the wall of the die and for the TPS to travel through the centre. In **Figure 4-3 C** to **E**, similar sections of skin formation are observed for PHA/starch blends added at the start of the barrel. The first thing to note is the presence of PHA globules throughout the TPS matrix. This suggests that the PHA is successfully being melted and is then aggregating. The second thing of note is that some of the PHA globules are remaining on the outer edges of the material, forming thin skins. Whilst not necessarily a clear example of flow-induced migration (given that a gradient in PHA enrichment from the centre to the surface is not observed), these experiments demonstrate the potential to produce PHA coatings through blended extrusion if parameters are optimised. This remains to be further explored through detailed experiments testing the effect of parameters such as PHA loading, HV content of the PHA, screw-speed and die temperature profile.

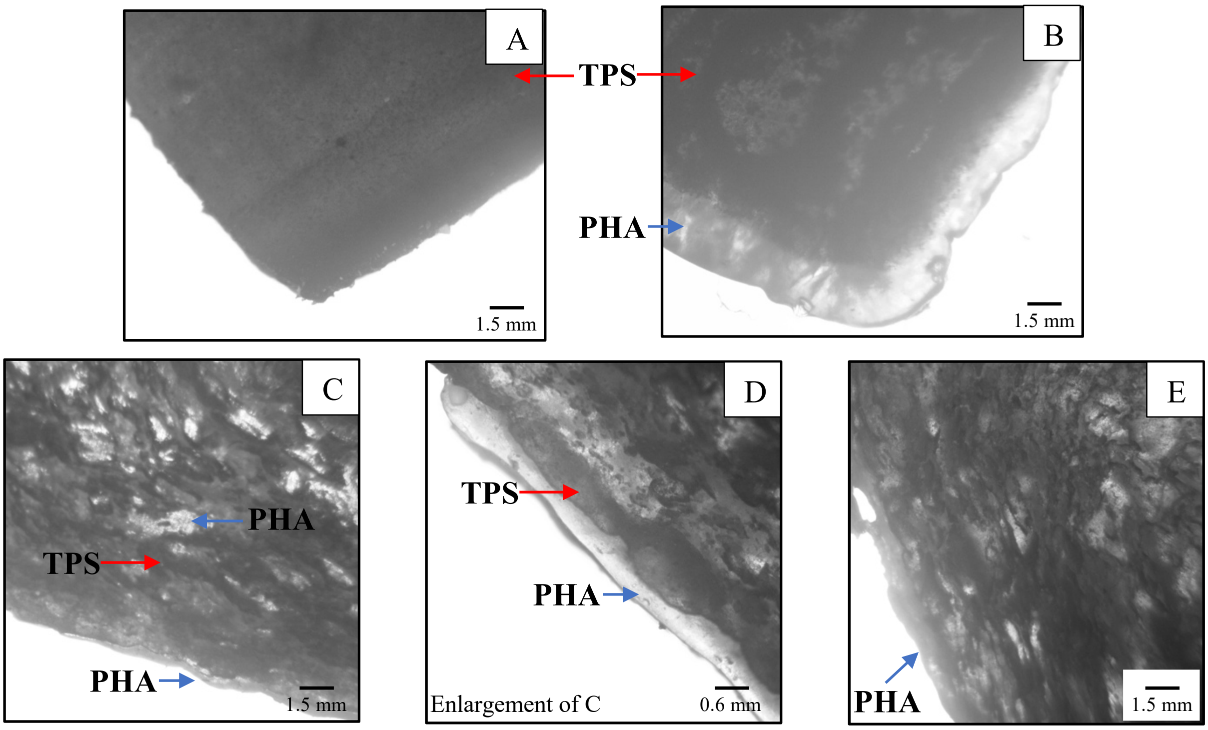


Figure 4-3: Iodine-stained light microscope images of extruded samples.

A) Just TPS, 4X magnification; B) TPS pushed through when the barrel is full of PHA, 4X magnification; C) 28% PHA/TPS, 4X magnification; D) 28% PHA/TPS, 10X magnification; E) 23% PHA/TPS, 4X magnification.

4.3.2 *Imaging*

A key challenge remains in identifying the location of each of the polymers within the blend. This challenge needs to be solved before further research into the extruder parameters can be undertaken. Iodine staining worked to an extent, but it cannot provide insight into the intricacies of the migration of each of the polymers, such as whether enrichment, but not complete separation, is occurring. Ideally, a visualisation method is required that can both map the composition as well as the thickness of each section of the film with detail. To date, this problem has not been satisfactorily solved either by this work, or within the literature. Bélard et al. (2009) used Attenuated Total Reflectance Fourier-Transform Infrared Spectroscopy (ATR-FTIR) and Electron Spectroscopy for Chemical Analysis for imaging their materials. However, this only revealed the composition of the top 1 μm for ATR-FTIR and 10 nm for electron spectroscopy. It is the same for Willett and Shogren (2002) who used X-ray photoelectron spectroscopy to observe surface enrichment, which only gives information about the top 1 nm. For the purposes of trying to produce packaging materials, which will require better knowledge of the material composition throughout its thickness, these are not suitable imaging techniques unless they can be combined with scanning capabilities. With this in mind, ATR-FTIR and Raman spectroscopy are identified as promising imaging methods that should be explored. The spectra they generate provide a unique molecular fingerprint, allowing them to determine blend composition, and they can both be coupled with scanning capability (Guillory et al., 2009; Larkin, 2011; Mieth et al., 2016; Widjaja and Garland, 2011).

4.4 Conclusion

The feasibility of producing PHA/TPS multi-layered structures through flow-induced migration was demonstrated and a methodology was developed to produce materials using a custom-designed, 14.5 cm long, extruder die. Initial experiments produced promising results, with samples starting to show the formation of PHA ‘skins’. There is further work in testing different processing conditions to improve the understanding of flow-induced migration for PHA/TPS systems. However, further research first needs to focus on development of an appropriate imaging method, which can provide a detailed mapping of the location of each of the polymers within the blend. Without this, the effect of changing processing parameters cannot be thoroughly examined (for example, in cases where enrichment, but not complete separation, is occurring).

CHAPTER 5 Marine biodegradation of PHA

Chapter Summary

Within the material properties theme, the second objective was to '*Determine the rate of biodegradation of PHA in the marine environment and apply this to lifetime estimation of PHA products'*. This chapter presents the research that addressed this objective. Through a meta-analysis, it was determined that the mean rate of biodegradation of PHA in the marine environment is 0.04–0.09 $mg.day^{-1}.cm^{-2}$ ($p = 0.05$) and that, for example, a PHA water bottle could be expected to take between 1.5 and 3.5 years to completely biodegrade.

This chapter was published in full as a journal article during the candidature.

Dilkes-Hoffman, L.S., Lant, P.A., Laycock, B., Pratt, S., 2019. The rate of biodegradation of PHA bioplastics in the marine environment: A meta-study. Mar. Pollut. Bull. 142, 15–24. https://doi.org/10.1016/j.marpolbul.2019.03.020

5.1 **Introduction**

Polyhydroxyalkanoates (PHAs) are normally presented as marine biodegradable plastics (Deroiné et al., 2014a; Volova et al., 2010). Supporting this claim is a reasonably extensive body of literature recording mass loss over time of PHA in the natural marine environment. However, to date, this research has been disparate, making it hard to draw overarching conclusions regarding PHA biodegradation rate or to estimate lifetimes. With production levels of PHA set to quadruple in the next five years (European Bioplastics, 2018), PHA is an important polymer to understand from a marine lifetime estimation point of view, to avoid implementing what may be a solution to recalcitrant plastics in theory, but a problem in practice. More broadly, an understanding of estimated lifetimes of marine biodegradable plastics is required in order to facilitate an informed discussion as to whether biodegradable plastics should be included in bans and taxes on plastic. This research determines an average rate of biodegradation of PHA in the marine environment based on the relevant available literature so that lifetime estimation of PHA products can be undertaken, allowing the risks and benefits to be more transparently discussed.

One hundred million tonnes of plastic waste are predicted to enter the oceans between the years 2010 to 2025 (Dilkes-Hoffman et al., 2019b; Jambeck et al., 2015) (**Chapter 2**). This has led to growing concern over the impacts of plastics in the marine environment (UNEP, 2016). Plastics that enter the oceans have a wide range of environmental and economic impacts including threat to marine organisms (through ingestion, entanglement, or habitat destruction), dispersal of invasive organisms and pollutants, and disruption of the tourism and fishing industries (Codina-García et al., 2013; Kedzierski et al., 2018; Moore, 2008). One of the key issues is that conventional plastics break down very slowly and only in the presence of UV radiation, heat, and/or oxygen (Andrady, 2015). Thus, these plastics can persist in the environment for hundreds to thousands of years, with degradation in the marine environment being particularly slow due to low temperatures in the ocean and minimal UV exposure once submerged (Andrady, 2015). One of the proposed solutions is to produce marine biodegradable plastics, such as PHAs, that have shorter lifetimes in the marine environment. However, it remains unclear what the timeframe for the biodegradation of such marine biodegradable plastics actually is.

Understanding the lifetime of biodegradable polymers starts with understanding the mechanisms through which biodegradation can occur. In this chapter, biodegradation is taken to mean the complete breakdown of materials through biological activity, such as through the action of microorganisms such as bacteria, archaea, fungi and algae. PHAs are biodegraded under both aerobic and anaerobic conditions by PHA degraders present in most natural environments, including the marine environment (Jendrossek and Handrick, 2002; Shah et al., 2008). Under aerobic conditions the resulting products should ultimately be biomass, CO_2 and water, whilst under anaerobic conditions the resulting products should be biomass, CO_2, methane and water (Gu, 2003).

The biodegradation of PHA is known to primarily occur through surface erosion via enzyme catalysed hydrolysis (Guerin et al., 2010; Laycock et al., 2017), meaning that when considered in simple terms, the rate of mass loss of a PHA object is related to the surface area accessible to enzymatic attack, and even whilst mass loss occurs, bulk material properties are normally preserved (Doi et al., 1992; Mergaert et al., 1995; Rutkowska et al., 2008; Sashiwa et al., 2018; Tsuji and Suzuyoshi, 2002a, 2002b). Attempting to understand the interplay of factors that influences the rate of biodegradation at any specific time point is when complexity is introduced. Properties of the polymer such as crystallinity, side-chain length, shape, and surface morphology as well as properties of the biodegradation environment such as temperature, UV exposure, nutrient levels, strength of mechanical forces, types of bacteria present, pH and oxygen levels can all influence the rate of biodegradation (Deroiné et al., 2014b; Laycock et al., 2017; Woolnough et al., 2013). Furthermore, as biodegradation proceeds, the surface of the polymer changes, pores can form and a shift of the mechanism towards bulk degradation and autocatalytic hydrolysis rather than purely surface degradation can occur (Ho et al., 2002; Laycock et al., 2017; Tsuji and Suzuyoshi, 2002a, 2002b). All of these factors act in synergy leading to a complex interplay which influences the rate of biodegradation.

Unfortunately, the data required to tease out the influence of the individual factors often doesn't exist, making the development of complex models hard. However, the lack of the information required to inform a complex model does not mean useful understanding cannot be developed from combining the literature. It is important to consider the timescales that one is interested in. Over longer timescales and when focused on macro properties such as time to complete biodegradation, the importance of each individual parameter diminishes, and bulk parameters can be considered appropriate.

Thus, in order to determine average rates of biodegradation of PHA for the purposes of this research, a simple approach to biodegradation has been adopted. A simplified biodegradation process has been conceptualised with three key steps and a rate for each defined (**Figure 5-1)** (Haider et al., 2018; Lucas et al., 2008). It is acknowledged that precise differentiation between each step in the process, as presented here, is not entirely accurate (all steps occur in a concurrent and iterative manner), but simplification is required for the purpose of communication. One of the three steps that has been defined is biofilm formation, the rate of which has been designated R_B. The biofilm is a unique and complex association of microbes formed from surface-associated microbial cells that are embedded in a self-produced extracellular polymeric substance (EPS) matrix consisting of polysaccharides, proteins and entrapped organic and inorganic particles (Donlan, 2002; Flemming, 1998). A lag time (ranging from a few days up to a few weeks) is often observed before a steady rate of biodegradation is reached, as it takes time for a biofilm to form and the microbial population to adapt (Imam et al., 1999; Woolnough et al., 2008). Another step that has been defined is enzyme catalysed, hydrolytic depolymerisation, the rate of which has been designated R_D. This is when extracellular depolymerases catalyse the hydrolytic bond cleavage of the polymer, eventually leading to the formation of oligomers, dimers and monomers. Finally, the uptake of small molecules by the cell during bioassimilation for either growth and reproduction or mineralisation has been combined into a single parameter. The resulting products of this process are increased cell biomass and simple end products like CO_2 in aerobic environments and methane in anaerobic environments. The rate of the bioassimilation and mineralisation has been designated R_M. R_B and R_D are considered to be rate limiting, meaning that the effects of any factors on R_M can be considered to be less significant (Chinaglia et al., 2018; Hong and Yu, 2003; Spyros et al., 1997).

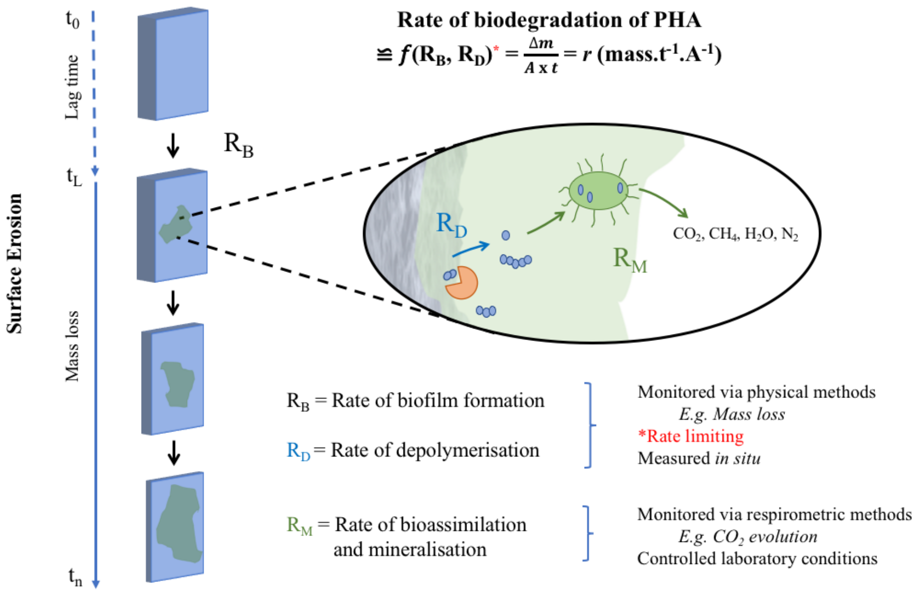


Figure 5-1: Rate of biodegradation of PHA

Note to Figure 5-1: R_B = Rate of biofilm formation; R_D = Rate of depolymerisation, R_M = Rate of bioassimilation and mineralisation; t_0, t_L, t_n = initial time point, lag time and final time point respectively; A = surface area, m = mass. It should be noted that whilst the steps of biodegradation are shown as occurring sequentially for means of communication, all processes are concurrently taking place, one process does not occur in totality before the next commences.

Two key methods by which biodegradation is normally monitored are physical methods (such as mass loss) or respirometric methods (such as CO_2 evolution and biological oxygen demand). Respirometric methods are used in laboratory studies to provide a complete picture of the polymer biodegradation, as they are the only way to prove that the final step of mineralisation has occurred. This is normally the focus of international biodegradation standards - such as the five active standards that exist for monitoring the biodegradation of a polymer in the marine environment (Harrison et al., 2018). These standards present a framework for proving that biodegradation occurs in the marine environment, and for comparing the rate at which biodegradation occurs between different materials. A recent and comprehensive review of these standards has been completed by Harrison et al. (2018). However, whilst a material that passes these standards can claim to be marine biodegradable, this does not provide the information required to allow for prediction of *in situ* rates of biodegradation, giving no indication of how long an item made out of that material would actually take to break down in the open environment. This is why physical methods, such as monitoring mass loss, are employed, as they are the only practical way to monitor biodegradation in the target environment.

Results from both respirometric methods and mass loss experiments can be used to calculate rates of biodegradation. Using data from respirometric methods allows for the rate of biodegradation to be calculated as a function of R_B (biofilm formation), R_D (depolymerisation) and R_M (bioassimilation and mineralisation) and for the effect of specific factors to be tested. Using data from mass loss experiments only allows for the rate of biodegradation to be calculated as a function of R_B and R_D and is less sensitive to the effect of different factors (Mohan Krishna and Srivastava, 2010; Shah et al., 2008). There are a few points to be noted for calculating rates based on the different methods. Firstly, results from CO_2 evolution experiments should only be used for estimations of rates up until the point at which 80% of the polymer carbon has been evolved as CO_2. At this point, mass loss is likely to have been completed even if not all of polymer has been evolved as CO_2 due to some of it being converted into biomass (Kasuya et al., 1998). Using data from beyond the 80% conversion timepoint to calculate rates or undertake lifetime estimations can lead to the estimation of longer times than are actually required for disappearance of the material. Secondly, for PHA the rate of mass loss (a function of R_B and R_D) is a suitable proxy for the rate of biodegradation even if it does not account for mineralisation. It has been suggested that the rate limiting step in the biodegradation of PHA is the biofilm formation phase and attachment of enzymes to the polymer surface leading to catalytic depolymerisation (Hong and Yu, 2003; Spyros et al., 1997). For the majority of the biodegradation process when direct mineralisation of the polymer is the focus, not mineralisation of the formed biomass, the assimilation and mineralisation of PHA is assumed to be rapid ($R_M \ll R_B$ & R_D; Chinaglia et al., 2018). Combining this assumption with the understanding that PHA is degraded via a surface erosion mechanism and only macro changes in polymer integrity over long timescales are of interest leads to the conclusion that the rate of biodegradation of PHA can be suitably estimated as the linear rate of mass loss over time.

The main aim of this work was to determine the rate of biodegradation of PHA in the natural marine environment and apply this to the lifetime estimation of various PHA products. This can be achieved by drawing together the existing literature on PHA weight loss in the natural environment in order to understand what the upper and lower boundaries of the biodegradation rate are. The aim of this research is not to develop a theoretical model for determining the rate of biodegradation of PHA under a specific set of conditions. Two secondary aims were to: a) compare the biodegradation rate of PHA in a marine environment to the biodegradation rate in soil, compost and anaerobic digestion; and b) collate information on the currently known factors influencing the biodegradation rate to determine what initial conclusions can be drawn at this point and identify what targeted studies need to be performed in order to better understand the significant parameters. This analysis was used to identify some of the gaps in understanding that still exist and to suggest initial improvements for further studies. It should be noted that this review does not cover work that only analyses the biodegradation of PHA by specific bacteria/enzymes, or that only assesses the microbial communities that are present during biodegradation but not the associated rates of biodegradation.

5.2 Methodology

Scopus and Google scholar were searched (final search December 2018) using a combination of search terms (**Table 5-1**) relating to polymer type and degradation environment. Only papers that focused on the biodegradation of PHA in a natural setting, or in a laboratory setting using a natural inoculum, were included. Any papers that focussed only on inoculation with specific bacteria were excluded. A final selection of 20 papers relating to biodegradation in the marine or aquatic environment was identified, and a list of these papers is detailed in **Table 5-2**. A list of the papers identified for soil, compost and anaerobic digestion environments is included in Appendix A (**Table A1**).

For the selected marine/aquatic papers, information relating to the following points was recorded if mentioned (presented in **Table 5-2**):

- The material and method (polymer composition, location of the study, method of monitoring biodegradation (mass loss, CO_2 evolution, loss of mechanical properties), shape of sample, length of study);
- The controlling variables (temperature, dissolved oxygen, salinity, UV exposure if near the surface, nutrients, pH, bacterial concentration and identification);

- The outcomes (final extent of biodegradation (based on mass loss, CO_2 evolution or other property changes as recorded in the study considered), molecular mass changes (M_w or M_n), crystallinity changes).

Table 5-1: Search terms

Note to Table 5-1: One term from each category was included and all combinations were tested.

Polymer type	Environment	Biodegradability
PHA	Marine	Biodegrad*
PHB	Seawater	
PHBV	Aquatic	
Biopoly*	Soil	
	Compost	
	Anaerobic	

Of the literature identified in **Table 5-2**, that focusing on the biodegradation of PHB or PHBV in the natural marine environment was reviewed. The studies that included sufficient information on the biodegradation rate and material characteristics (starting mass, sample shape and surface area) were selected. A brief summary of these papers is presented in Appendix A (**Table A2**). Data was normalised to a rate of polymer biodegradation based on initial surface area ($mg.day^{-1}.cm^{-2}$) (equation 5-1). This enabled a comparison between studies in the marine environment as well as between environments (i.e. marine, soil, compost, anaerobic digestion).

$$r = \frac{\Delta m}{A \times t} \quad \text{(Eq. 5-1)}$$

where r represents the specific rate of mass loss ($mg.day^{-1}.cm^{-2}$) and is a function of R_B and R_D as presented in section 5.1; Δm is the change in mass (mg); A is the initial surface area (cm^2) and t represents time (days). The area of the face of a film was taken as the length multiplied by the width, and does not account for surface topography, pores and voids. Change in mass was calculated from the start of the experiment to the final time point presented and the rate was assumed to be linear, with no adjustments made for lag time or biodegradation plateaus. Furthermore, no adjustments were made for the acceleration of biodegradation that can occur as a result of autocatalysed hydrolysis or increase in surface area (from increased surface roughness or fragmentation). Incorporating the lag time into the total time for biodegradation, rather than focusing only on the portion of the mass loss curve where mass loss is seen to occur, gives an average rate of biodegradation, rather than a maximum rate, leading to more accurate lifetime predictions. Where a starting mass was not reported, the density (assumed to be 1.24 $g.cm^{-3}$ (TianAn PHBV (4%))) and dimensions of PHA were used to estimate the initial mass.

The 95% confidence interval of the mean for the specific rate of mass loss was calculated and then converted into a rate of surface erosion per day (mm.day^{-1}) as shown in equation 5-2;

$$\lambda = \frac{r}{\rho} \qquad \text{(Eq. 5-2)}$$

where λ represents the rate of surface erosion (mm) and density (ρ) was taken as 1.24 g.cm^{-3}. An estimation of the likely polymer lifetime can then be made using equation 5-3;

$$t_d = \frac{h_0}{2\lambda} \qquad \text{(Eq. 5-3)}$$

where h_0 is the starting thickness of the film (mm) and t_d is time to complete biodegradation (days). Dividing by two accounts for surface erosion on both faces of the film.

5.3 Results and discussion

The rate of biodegradation of PHA in a marine environment was calculated through collating the results from eight identified papers that contained sufficient information to allow for normalisation of biodegradation rate on a mass per surface area per time basis (**Figure 5-2A**). Given that biodegradation of PHA occurs via a surface erosion mechanism (Guerin et al., 2010; Laycock et al., 2017) and it has been shown that surface area is an important factor influencing biodegradation rate (Chinaglia et al., 2018), normalising to surface area is important in order to allow comparison between the different studies. The rate of biodegradation is also influenced by a variety of factors which differ across studies and cannot be controlled for. This is a limitation that will exist for any collation of rates measured in a natural and continuously fluctuating environment and is why the 95% confidence interval of the mean should be focused on rather than the sample mean. The calculated 95% confidence interval of the mean is 0.04 – 0.09 mg. day^{-1}.cm^{-2} (See Appendix A, **Table A2** for individual data points) – i.e. there is 95% confidence that this interval contains the true mean of the rate of biodegradation of PHA in the marine environment. The factors that may then influence the biodegradation rate of a PHA product in a specific environment (within this range) are discussed in **Table 5-3** and **Table 5-4.** It is highlighted that this result is based on the best information available to date and should be updated once the controlling factors are more thoroughly understood.

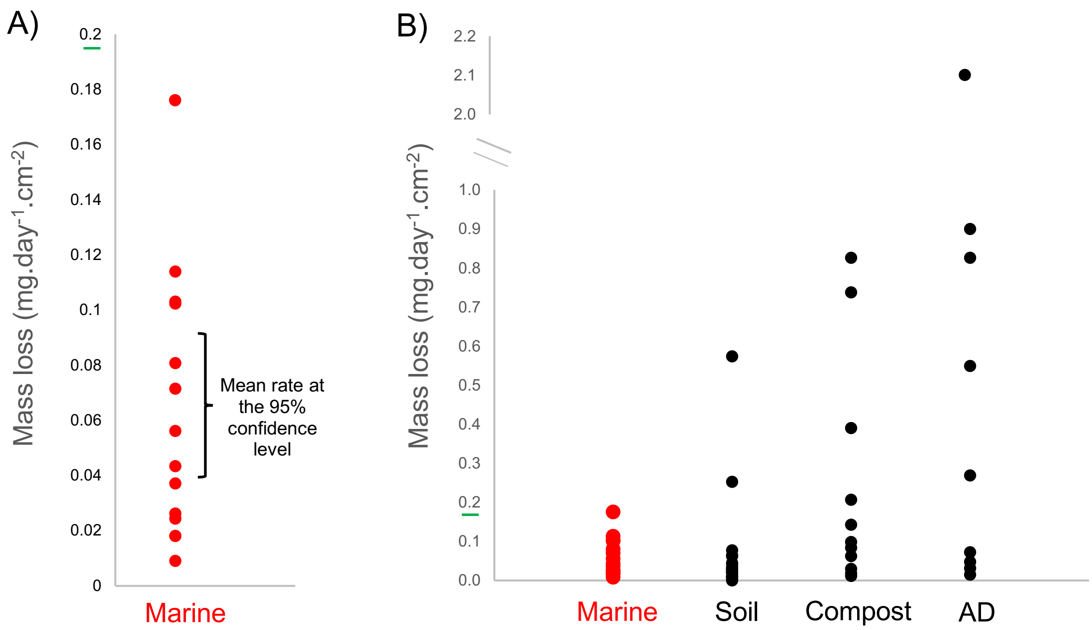


Figure 5-2: Normalised biodegradation rate of PHA in different environments

Note to Figure 5-2: A) Marine data in detail including the 95% confidence interval of the mean. B) Degradation rate in different environments (marine data is the same as in A). Note the break in the y-axis for B.

To add context to these results, the 95% confidence interval was used to calculate the potential lifetimes of different PHA items in the marine environment (**Figure 5-3**). As would be expected, the time required for complete biodegradation of a product and the range of uncertainty increases with thickness of the material. A 35 µm PHA bag, for example, could be expected to last between 25 days and two months before it has completely biodegraded. On the other hand, a PHA bottle, with a wall thickness of 800 µm, is expected to take much longer, with the shortest lifetime being one and a half years, but the upper limit being approximately three and a half years. These are currently the best predictions that can be made. However, there is clearly more research required to reduce the level of uncertainty, as well as to tailor the predictions to the many different ocean environments that exist. **Figure 5-3** is useful in that it defines upper and lower bounds for what could be expected for the mean of lifetime estimation of different PHA objects. However, factors such as temperature, nutrient availability, and location in the water column (explored in **Table 5-3** and **Table 5-4**) will influence what the mean lifetime for a specific material in a specific environment could be expected to be.

A key limitation with this value, which is unavoidable given the available data, is that it heavily draws on work performed on thin films. It could be expected that thin films would degrade faster than thicker objects due to the propensity for the formation of pores and cracks, increasing surface area (Tsuji and Suzuyoshi, 2002a, 2002b) and enabling more rapid fouling and fragmentation (Volova et al., 2010; Wang et al., 2018). This means that the biodegradation rates calculated in the majority of the reviewed papers have the potential to over-estimate biodegradation rates when extrapolated to solid objects. This said, thicker objects (that have a longer lifetime) will be less influenced by the uncertainty surrounding lag time than thinner objects (with shorter lifetimes). Thus, there is also a margin of error on the degradation times for the thinner objects such as plastic bags and this is related to the time taken for biofilm formation.

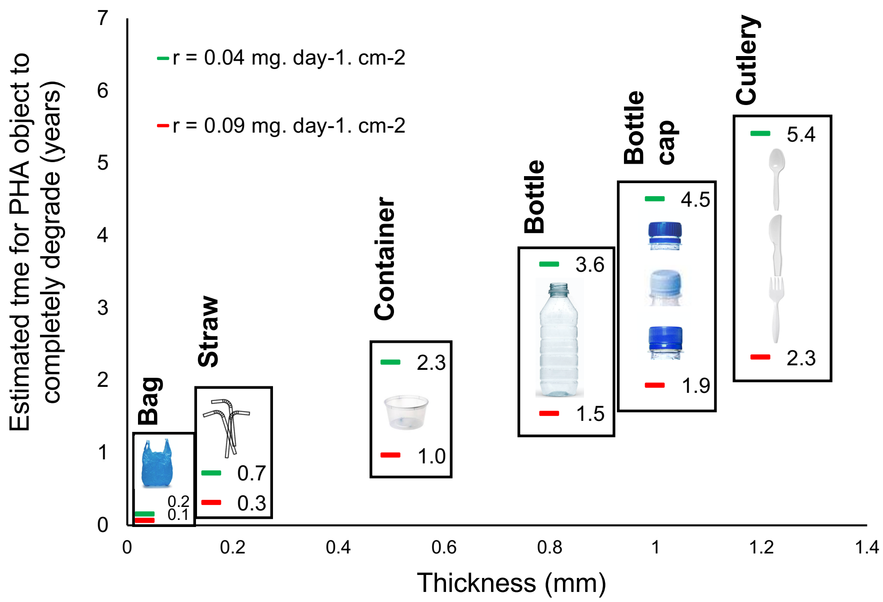


Figure 5-3: How long will it take a PHA item to degrade in the marine environment?

Note to Figure 5-3: Lifetime values estimated using the 95% confidence interval for the mean of the rate of biodegradation of PHA in the marine environment.

It is known that biodegradation kinetics can vary depending on the environment. In order to understand how the biodegradation rates of PHA in the marine environment compare to those in other environments, some key papers relating to the biodegradation of PHA in soil, compost or anaerobic digestion were identified and, where possible, results normalised in the same way as described for the marine studies to allow for comparison (**Figure 5-2B**). The ranges for biodegradation rate in both compost and anaerobic digestion (AD) were much larger than for the soil and marine environments, indicating the capacity for a much higher rate of biodegradation in these systems. This would be expected given that both compost and AD are controlled systems, with higher concentrations of microbes and higher operating temperatures, designed to provide optimal rates of breakdown (Haider et al., 2018). On a within-study basis, Rutkowska et al. (2008) found that the weight loss of PHBV was different between different environments and decreased in the expected order with anaerobic sludge > aerobic sludge > river sediment > seawater. Manna and Paul (2000) also found the biodegradation rate of anaerobic digestion to be faster than soil, which in turn was faster than in compost or marine settings. However, more targeted research with controlled samples across the different environments will be required to confirm these results.

In an attempt to develop a more nuanced understanding of the biodegradation rate of PHA in a marine environment, the factors that have the potential to influence the rate of biodegradation were identified and evidence relating to their influence were collated from all of the PHA marine biodegradation studies. The results of this detailed analysis are recorded in **Table 5-2**. The benefit of this collation is that it allows a rapid assessment of which factors have received considerable exploration and which factors have not.

Each of the data points for biodegradation rate of PHA in the marine environment presented in **Figure 5-2** were then coded for the four factors that all of the studies reported on. Namely, material type (PHB or PHBV), environment (freshwater or marine), thickness of the sample (>2 mm or film) and temperature of the test environment (<0°C, 3-10°C, 11-25°C and >25°C). The analysis (not shown) did not reveal an identifiable relationship between biodegradation rate and any of these four factors. However, this finding is obviously limited in its significance due to the limited data available.

Given that no clear relationship emerged, the information available on the effect of each of the controlling factors presented in **Table 5-2** was qualitatively reviewed. Both structure and property of the polymer as well as location, weather and climatic conditions substantially influence biodegradation rates (Volova et al., 2006), so the factors have been divided into two groups - those that are a property of the material being studied (presented in **Table 5-3**) and those that are a property of the environment being studied (presented in **Table 5-4**).

Table 5-2: Detailed description of all factors considered in marine biodegradation of PHA studies

	Material		Location			Test Method		Shape					Molecular mass	Crystallinity	Temp (°C)		UV	Nutrients	Bacteria and fungi		Mechanical property changes considered?	Length of study (days)
	PHB	PHBV	Lab	Field	Sediment considered?	Weight loss	CO_2 evolution	Film	Thickness (µm)	Solid	Thickness (cm)	Powder	Changes Considered?	Changes Considered?	Range (°C)	Investigated as key point?	Considered?	Measured or added?	Counted?	Identified?		
Brandl and Puchner (1992)		✓ (8%)		✓*	✓	✓		✓	not stated	✓	Bottle				< 6 6 - 8							245
Deroine et al. (2014a)		✓ (8%)	✓	✓		✓				✓	0.4		✓	✓	4, 13, 25, 40	✓			✓		✓	360
Deroine et al, (2015)		✓ (8%)	✓	✓	✓	✓	✓	✓	200			✓	✓	✓	11 – 20				✓			180-600
Doi et al. (1992)		✓ (0-61%)		✓		✓		✓	50-150	✓	0.2		✓		13 - 26					✓	✓	365
Greene (2012)	✓		✓		✓		✓	✓	not stated						30							365
Ho et al. (2002)	✓		✓			✓	✓	✓	not stated						28			✓ (added)	✓			86
Imam et al. (1999)		✓ (12%)		✓		✓		✓	510						Various 25 – 29 32 - 36				✓		✓	365
Kasuya et al. (1998)		✓ (14%)	✓			✓		✓	100						25			✓ (added)				28
Mayer (1990)		✓ (8, 24%)	✓		✓	✓		✓	not stated						30							84
Mergaert et al. (1995)	✓	✓ (10, 20%)		✓		✓				✓	Dog bone		✓		Marine 6 – 14					✓	✓	180
Muhamad et al. (2006)	✓	✓ (20%)	✓			✓		✓	not stated						37							Not clear
Rutkowska et al. (2008)		✓ (12%)		✓		✓		✓	115				✓		17 – 20						✓	42
Sashiwa et al. (2018)		Other	✓				BOD	✓	20			✓	✓		27							28
Sridewi et al. (2006)	✓	✓ (5%)		✓	✓	✓		✓	not stated						32							56
Thellen et al. (2008)	✓	✓ (5 - 12%)	✓	✓	✓		✓	✓	190						Field 12 - 22			✓ (both)				49-100
Tsuji and Suzuyoshi (2002a)	✓			✓		✓		✓	50				✓		19 - 26						✓	150
Tsuji and Suzuyoshi (2002b)	✓		✓			✓		✓	50				✓		25						✓	300
Tsuji and Suzuyoshi (2003)	✓		✓			✓		✓	50-200						25						✓	35
Volova et al. (2010)	✓	✓ (11%)		✓		✓		✓	100	✓	0.5		✓	✓	27 – 30				✓	✓		160
Wang et al. (2018)		Other	✓				✓			✓	0.1				Ambient			✓ (added)		✓		195

* Only a freshwater environment was considered, ** studies highlighted in red are those used in Figure 5-2 A.

Table 5-3: Factors that are a property of the material

Properties of the material	
Polymer type	Within the polyhydroxyalkanoate family, most marine biodegradation studies have focused on PHB and PHBV (most commonly within the range of 5 – 20 mol% HV content). When directly comparing the biodegradation rates of PHB and PHBV (11% HV) films, Volova et al. (2010) noted no difference between the biodegradation rates. However, Thellen et al. (2008) noted faster biodegradation rates for PHBV (12%) when compared to PHB, as did Mergaert et al. (1995) for PHBV (10% and 20%). Doi et al. (1992) had inconsistent results with the 21% HV content sample degrading faster than all of the other samples (4% HV content sample, PHB, and 61% HV content sample which all had similar rates to each other). In a review on polymer lifetime prediction, Laycock et al. (2017) reports that copolymers consistently degrade faster than homopolymers.
Shape and Surface morphology	There has not been a great deal of work into understanding the influence of shape on biodegradation rates. Most of the studies of PHA biodegradation in the marine environment focus on the biodegradation of films, in the range from 20 – 510 µm. Two papers analysed dog-bone specimens (Deroiné et al., 2014a; Mergaert et al., 1995) whilst only three papers experiment with both films and 3D forms (Brandl and Puchner, 1992; Doi et al., 1992; Volova et al., 2010). Volova et al. (2010) found that films (100 µm) degraded faster than compacted pellets. However, no other study specifically comments on this. In regard to consideration of surface morphology, Tsuji and Suzuyoshi (2003) produced PHA films with pores on the surface and found that this significantly enhanced the biodegradability.
Crystallinity	None of the papers interrogate crystallinity as a controlling factor. In regard to changes in crystallinity during biodegradation, Volova et al. (2010) and Deroine et al. (2014a) found that the crystallinity index does not change with biodegradation of PHA which is consistent with a surface erosion mechanism.

Table 5-4: Factors that are a property of the environment

Properties of the environment	
Location in the water column	Although only considered in a few studies, it appears that contact with sediment plays a significant role in influencing rates of biodegradation. Mayer (1990), found that biodegradation with sediment contact is faster than just in water and Sridewi et al. (2006) found that objects on a sediment surface degraded slower than those that were completely buried. Deroine et al. (2015) found the biodegradation kinetics to be slower in just sand compared to a saturated sand and seawater combination, proposing that this is due to degree of surface contact. Thellen et al. (2008) also considered sediment addition and concluded that sediment and the associated microbes play a role in influencing rates of weight loss but could not establish a simple relationship.
Temperature	A range of different temperatures have been considered in the studies reviewed. When temperature was considered within a study, it was normally found that biodegradation is faster when water temperature is higher. Mergaert et al. (1995) observed that biodegradation of a sample monitored in the environment was faster over summer and Brandl (1992) found that biodegradation of a PHA bottle was faster closer to the surface of a lake. Both related these results to the temperature of the water, with higher water temperature leading to faster biodegradation rates. Doi et al. (1992) also observed that the rate of surface erosion was markedly dependent on the temperature of the sea-water. Thellen et al. (2008) suggests that the effect of colder water temperatures (and limiting nutrient supply) is what slowed the rate of weight loss in a natural environment compared to standard laboratory methods. Deroine et al. (2014a) is the only study to consider controlled trials of different water temperatures as part of accelerated ageing experiments designed to assess the validity of lifetime estimation based on the Arrhenius relationship. The temperatures used were 4, 25 and 40 ºC. They found that the increase in temperature did not have a substantial impact on weight loss. This needs further investigation.
Nutrients	None of the studies performed in the natural environment report the nutrient levels of the water or discuss it as a controlling factor. This is an oversight given that nutrient levels have been known to influence bacterial populations for many years (Zobell and Grant, 1943) and are likely an important and variable factor across different field settings.
Microbes	Seven of the studies quantify the concentration of bacteria in their study (Deroiné et al., 2015, 2014a; Ho et al., 2002; Imam et al., 1999; Mayer, 1990; Volova et al., 2010; Wang et al., 2018) whilst a few take this a step further and perform sequencing to identify the organisms present (Doi et al., 1992; Mergaert et al., 1995; Volova et al., 2010; Wang et al., 2018). Some of the studies measure concentrations before addition of the PHA whilst others measured the concentration after. None looked at a change in the microbial community over time.
UV light exposure	The only paper that mentions UV in a marine setting when considering PHA biodegradation (Tsuji and Suzuyoshi, 2002a) doesn't measure UV radiation, but states that they believe it is having no influence given that no changes in molecular mass are observed. PHA is denser than water, so it is not expected to be found on the surface in a marine environment and therefore UV exposure will probably be minimal.
Dissolved oxygen and Salinity	Dissolved oxygen and salinity are recorded but not discussed as a controlling factor in any of the studies.
pH	No studies discuss the influence of the pH of the natural environment on PHA biodegradation. The only study that focuses on pH considers hydrolysis in the absence of enzymes under controlled conditions (Muhamad et al., 2006). The mass loss of PHB and PHBV samples in pH 7.4, 10.0 and 13.0 at 37 °C, were monitored and it was found that degradation proceeded faster in an alkaline medium. However, the mechanism was not delved into further.

Ultimately, it is hard to isolate the influence of any of these factors in the natural environment and it is the influence and interplay of each of these identified (and potentially other unidentified) factors that need to be understood.

In regard to the influence of environmental factors, it is important to consider the ultimate location of PHA in the marine environment. PHA contains heteroatoms in its backbone and is denser than water, meaning it is more likely to sink than a conventional polyolefins. This suggests that PHA is likely to be in contact with sediment rather than be free-floating, and that its dispersal via ocean currents will be different to a conventional polyolefin (potentially remaining closer to its point of entrance to the ocean versus being distributed to the open ocean). Any sort of light exposure and UV degradation will probably be minimised if it settles in the sediment, removing one of the most important factors initiating the abiotic photodegradation of conventional polymers (Gewert et al., 2015). In addition, sediment, and in particular deeper sediment layers, are suggested to host a larger consortium of microorganisms and will have low dissolved oxygen concentrations (Andrady, 1994). Furthermore, if PHA remains close to shore it would likely be exposed to higher temperatures and more active bacterial populations (Deroiné et al., 2014a; Rutkowska et al., 2008) given that the bacterial population in deeper, colder water can be at least one order of magnitude lower than in shallower testing environments (Deroiné et al., 2014a; Imam et al., 1999). This suggests that tests conducted in sediment with nutrient and temperature profiles similar to a shoreline are more likely to be reflective of the biodegradation of PHA than those conducted as suspended samples in the open ocean.

In regards to the influence of the material characteristics, surface phenomena, particularly roughness, can influence bacterial attachment and enzymatic action (Woolnough et al., 2013), as can porosity (Chan et al., 2019; Tsuji and Suzuyoshi, 2003) and crystallinity (Spyros et al., 1997). The type of polymer processing (solvent cast, melt pressed, extruded), post processing treatment, and surface chemistries (e.g. orientation effects) can also influence biodegradation rate. For example, Sridewi et al. (2006) suggested that the increased surface porosity of a poly(3-hydroxybutyrate-*co*-5 mol% 3-hydroxyhexanoate) film compared to other films contributed to its increased biodegradation rate whilst Boyandin et al. (2013) suggested that due to the presence of micropores at inter-particle boundaries in pellets formed through a pressing mechanism, they degraded faster than samples produced through casting.

A deeper understanding of these factors influencing biodegradation rates will require studies to investigate targeted comparisons between samples as opposed to just considering biodegradation in general. In particular, studies are required that look at the influence of shape, surface morphology, porosity, additives and processing techniques as they have the potential to be used as controlling factors (Chan et al., 2019; Sridewi et al., 2006; Tsuji and Suzuyoshi, 2003). There has also been no targeted consideration of the method of production of the PHA objects tested, the processing additives, or the post-processing methodology, although this influences the material properties and potentially the biodegradation rate (Cherpinski et al., 2017; Follain et al., 2014; Laycock et al., 2017). Most of the literature to date has focused on thin films (< 200 μm) which may behave differently than thicker objects and this affects lifetime estimations.

It is also important that more studies be conducted in the natural environment, as there are issues with transferability of results from laboratory studies (Deroiné et al., 2014a) and in the natural environment the polymer may not be a preferred substrate relative to other available materials (Haider et al., 2018). As discussed, the likely sinks (e.g. sediment) must also be considered and should be the target locations for biodegradation studies (Nauendorf et al., 2016).

Of the literature reviewed, many papers failed to report the critical information that is required to make a comparison between the different pieces of research, limiting the utility of the body of research to date. Initial mass and surface area or sample dimensions should always be reported to allow for standardisation between studies.

5.4 **Conclusion**

The aim of this research was to determine the mean biodegradation rate and lifetime estimation of PHA in the marine environment, dependent on the information available to date. The key result is the determination of the mean rate of biodegradation of PHA in the marine environment as 0.04 – 0.09 $mg.day^{-1}.cm^{-2}$ ($p = 0.05$). This was used to estimate the average lifetime of various PHA products in the marine environment. For example, using the calculated biodegradation rate a PHA bottle could be expected to take approximately one and a half to three and a half years to completely biodegrade. No single environmental or morphological factor emerges as the key factor influencing biodegradation rate and there is not enough information to understand their individual effects or develop a robust understanding of how an individual factor would affect lifetime estimation. This in itself is an important contribution, guiding future research and demonstrating that more targeted studies are required that directly compare the influence of different factors (particularly properties of the test sample) as well as ones that consider the ultimate location of the PHA. The calculated rate can be updated once these controlling factors are more thoroughly understood.

CHAPTER 6 Life-cycle assessment

Chapter Summary

Within the environmental impact assessment theme, the objective was to '*Assess the environmental impact of PHA/TPS food packaging*'. This chapter presents the results of a life-cycle assessment conducted to address this objective. Food waste was included in the system boundary and the impact of landfill methane capture efficiency was considered. A key result was that, when food waste is included in the system boundaries, it contributes over 50% of the greenhouse gas (GHG) emissions associated with the system, regardless of whether the package is biodegradable or not. The overarching result was that a PHA/TPS food packaging only delivers positive GHG outcomes if it reduces food wastage or increases the viability of biological food waste processing.

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6.1 **Introduction**

Whilst recycling is viewed as the primary mechanism to reduce the environmental and waste management issues associated with the use of plastics, 30% of plastic packaging materials may never be eligible for recycling or reuse without fundamental redesign of the materials used (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). This is due to a variety of reasons. Some niche materials are used in too small a quantity to justify the recycling infrastructure, some plastic products are considered too small for practical sorting, and some packaging products are prone to being contaminated with organics (particularly food) or chemicals (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016).

Another category of materials that pose a particular challenge for recycling, are those that comprise a combination of different plastic types – whether that be blended plastics or multi-layered materials (with a different plastic type in each layer). In either case, the cost and technology constraints of separating and recycling the different plastic polymers can be prohibitive and alternative strategies are required to address the waste management challenges associated with plastic disposal (Barlow and Morgan, 2013; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). On land, the focus of materials redesign and waste management is to reduce the reliance on landfilling for waste disposal, as well as reduce the amount of plastic that is disposed of in open-dumps and on the streets in developing countries (Hoornweg and Bhada-Tata, 2012; Hopewell et al., 2009). This will then help to reduce the amount of plastic waste that enters the oceans. It has been estimated that ~30% of all packaging is not disposed of appropriately, and thus has the potential to accumulate in the world's oceans (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). Given this, it is also important that materials design focuses on reducing the marine life impact of the plastics that inevitably make their way to the oceans.

Food packaging is a particularly problematic part of the global challenge with plastics waste management as it represents the largest demand for plastic packaging (PlasticsEurope, 2016) and comprises a large portion of the objects identified in coastal surveys (Andrady, 2015). This seems likely to increase, as urbanisation and dietary change in developing countries is leading to an increased global reliance on processed foods (World Packaging Organisation, 2008). Improved recycling systems can only go so far to solving this problem, in part because of food contamination, and in part because the food industry is increasing its use of multi-layered packaging materials. This is because multi-layer materials can provide a much higher resistance to water and gas transfer than single-layered materials, and thus reduce food spoilage (Barlow and Morgan, 2013).

Given that materials redesign has a useful role to play for food packaging, a multi-layered combination of thermoplastic starch (TPS) and polyhydroxyalkanoate (PHA) could warrant inclusion in the list of materials being considered (Fabra et al., 2016a; Shogren, 1997). TPS is one of the best oxygen barriers of all polymeric materials (Krochta and De Mulder-Johnston, 1997), while it has been shown that PHA exhibits the best water barrier properties of biodegradable polymers (Shogren, 1997). The combination might therefore offer impressive barrier properties, with the potential to lower food spoilage rates compared to more conventional packaging materials.

The biodegradation characteristics of a PHA/TPS combination also seem promising. The appeal of biodegradable food packaging is that it might broaden the waste management options for materials that can't easily be recycled (by including biological processing). However, while there are a number of plastic polymers that exhibit high biodegradation rates in landfill or composting, TPS and PHA are two of the very few materials that also show potential to biodegrade in sea-water (O'Brine and Thompson, 2010; Volova et al., 2010).

A mixed-layer PHA/TPS material for food packaging might therefore be rather unique in its potential to both reduce food spoilage rates and alleviate the marine pollution impacts caused by recalcitrant plastics making their way to the ocean. The PHA/TPS combination has been successfully formulated at lab-scale using a number of different techniques (Dilkes-Hoffman et al., 2018b; Fabra et al., 2016a; Martin et al., 2001) (**Chapter 3** & **4**) although further research is required to better understand its food preservation and marine-degradation performance.

The potential PHA/TPS combination also offers an intriguing mix of greenhouse gas (GHG) implications in a carbon-constrained world. Focusing just on the production processes, some studies suggest the energy demand to produce biopolymers can be higher than for conventional products, but the overall carbon footprint can be lower depending on assumptions about the biological feedstock used (Hottle et al., 2013; Yates and Barlow, 2013). Furthermore, biodegradability is not always viewed in the literature as a GHG benefit, depending on the assumptions used for degradation rates and waste handling systems (Yates and Barlow, 2013). Importantly, while most carbon-footprint studies of food packaging pay no attention to the food itself, the few that do consider the role of packaging choice on food preservation show that this could be the single biggest influence on overall GHG results (Conte et al., 2015; Williams and Wikström, 2011).

Here, the aim was to identify those design characteristics most useful for minimising environmental trade-offs associated with a multi-layered PHA/TPS food packaging material. The primary focus for this research is on GHG implications, although life-cycle water demand is also considered, given the potential importance of food production to the conclusions. The analysis covers the full life-cycle of the biodegradable packaging and the food it contains. To our knowledge, this is the first study to discuss the implications of including food wastage when assessing biodegradable food packaging materials. The study also considers the impacts of landfill methane capture efficiency.

Two very different food products (beef and cheese) that would likely benefit from a packaging material with improved water and oxygen barrier properties were considered (Barlow and Morgan, 2013). Sensitivity analysis was used to consider potential variation in those parameters found to be influential in previous literature (Clune et al., 2017; Yates and Barlow, 2013). A key outcome is that food waste is shown to contribute at least 50% of the GHG emissions in a food packaging life-cycle analysis regardless of whether the package is biodegradable or not, and that the differences in the relative impact of the two types of packaging are minimal compared to the overall, particularly when including food waste in the calculation. As a consequence, reducing food waste is a key design consideration for PHA/TPS food packaging. Such packaging was only found to deliver positive GHG outcomes if it reduced food wastage or contributed to building the viability of biological food waste processing (e.g. anaerobic digestion). Reducing food wastage also has the potential to offset the emission released from biodegradable packaging in inefficient landfills.

6.2 **Methods**

6.2.1 *Life-cycle assessment*

Life-cycle assessment (LCA) is the most popular tool for estimating the environmental impact of a product or process throughout its entire life-cycle (Reap et al., 2008) and guidelines for its implementation are defined by the International Organisation for Standardisation 14040 series (ISO, 2006). An LCA involves collating information on the inputs and outputs of a system and subsequently converting these to environmental consequences (such as global warming potential). It was thus selected as the tool to explore the research question. Although not a perfect tool (e.g. it is unable to capture the marine impacts of plastic litter), nor able to provide definitive answers, it is a transparent way in which to assess specific environmental trade-offs (Schnoor, 2009).

6.2.2 *Literature review*

In food packaging LCAs, the default has been to analyse the packaging in isolation, overlooking the implications of the food system (Madival et al., 2009; Toniolo et al., 2013). However, there is a movement towards including the function of the package (i.e. delivering food to the consumer) and associated indirect effects (e.g. food waste) within the system boundaries (Wikström et al., 2016). Results of such studies have demonstrated the importance of using this expanded system boundary to reduce total environmental impact when considering development of alternate packaging materials (Williams and Wikström, 2011). In particular, Conte et al. (2015) assessed single layer and multi-layer conventional packaging and showed that multi-layer materials emerge as environmentally superior only when food waste is included in the system boundaries. To further develop the idea, Grant et al. (2015) have developed a packaging design tool that includes product loss in the assessment of packaging materials. Whilst the impact of food waste in a packaging LCA has been documented, a biodegradable packaging option has not been considered. Thus, the expanded LCA methodology was selected for this study.

6.2.3 *Methodology of the study*

Impact factors

Climate change and water use were calculated as the impact factors. The study was limited to these impact factors as there was not sufficient data available over the entire scope of the analysis for the consideration of other impact factors.

- *Global Warming Potential (GWP)*

Global warming potential using a 100-year time horizon (GWP100) was used as the default impact factor across the entire study. However, the need to check the sensitivity of an LCA's conclusions to different modelling choices is discussed by Levasseur et al. (2016). It was for this reason that a global warming potential using a twenty-year time horizon (GWP20) was also calculated for selected scenarios to consider impacts in the nearer term. Methane is a short-lived GHG, and because of this using a twenty-year time horizon places greater emphasis on its emissions and its associated global warming potential than using a 100-year time horizon (Levasseur et al., 2016). The GWP is reported as CO_2-equivalent (CO_2e) emissions.

- *Water use*

There is a considerable amount of water use associated with food production (Mekonnen and Hoekstra, 2010). Thus, a water use analysis was conducted for the food and packaging system as it was predicted that it would be an important factor when considering the potential sustainability impact of a package in the context of the food system. Water use was added on a volumetric basis across the life-cycle from available inventory flows for water inputs from water storages, rivers and lakes. Rainwater inputs to agriculture were not included and no account was included for water stress in the regions where water was taken from. Water stress was not included in an attempt to keep the results independent of location.

Scope and functional unit

The entire system of a hypothetical supply chain from production of the raw materials to end-of-life (EOL) was considered for the **functional unit** '1kg of packaged product at the house' as presented in **Figure 6-1**. Mass flows for the figure are recorded in **Table 6-1**. Production of the polymers occurred in China, with subsequent transport to Victoria (Australia) where the package is produced and combined with a food product, followed by transport to Queensland (Australia) where the product is sold in a supermarket, consumed at the house and the waste disposed of.

The influence on the environmental outcomes of changing packaging types, food types, waste disposal scenarios and food wastage levels were explored.

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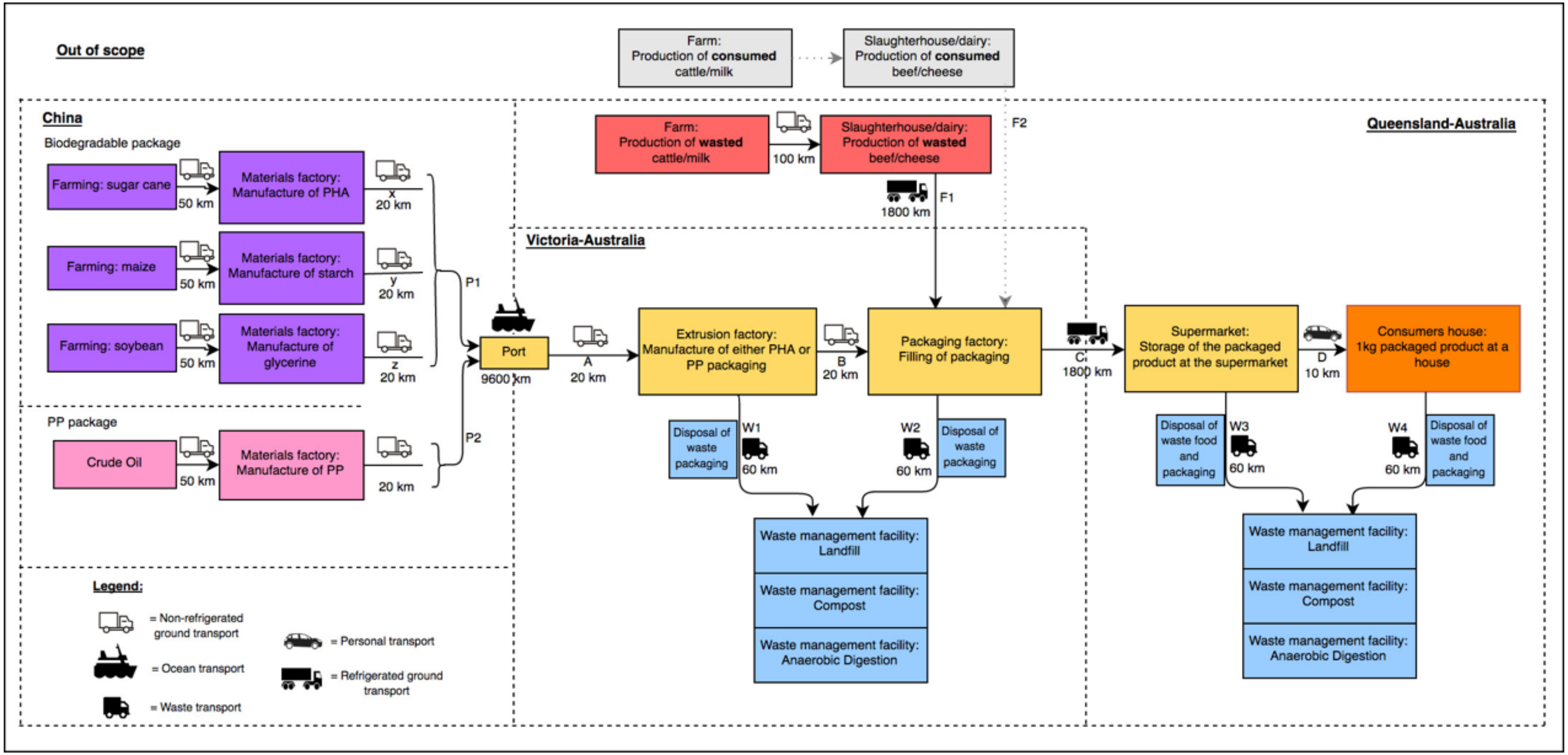

Figure 6-1: System boundaries for the LCA study

Note to Figure 6-1: Production of food consumed at the house (and associated transport and refrigeration) is not included in the system boundaries

Table 6-1: Mass flows associated with **Figure 6-1**

Material flows as per Figure 6-1	**Beef** Packaging (g)	**Beef** Food (g) Wasted	**Beef** Food (g) Consumed	**Beef** Total (g)	**Cheese** Packaging (g)	**Cheese** Food (g) Wasted	**Cheese** Food (g) Consumed	**Cheese** Total (g)	**Calculation**
P1 (x)	11.5				11.6				Total PHA = 20% of A
P1 (y)	27.6				27.8				Total Starch = 60% of 80% of A
P1 (z)	11.5				11.6				Total glycerol = 25% of 80% of A
				50.6					Note: The remainder of the total of the biopolymer packaging is water (15% of 80%).
P2	57.7			57.7	58.0			58.0	Total PP = 100% of A
F1		110.8				92.3			Fraction of the food wasted = W3 + W4
F2			878.8				902.5		Fraction of food consumed = Total food in 1000g of packaged product minus fraction of food wasted at the house (Beef = 950-(950 x 7.5%), cheese = 950- (950 x 5%))
A	57.7				58.0	0			Packaging (beef or cheese) = B + 5% wastage
B	54.8				55.1	0			Packaging (beef or cheese) = C + 5% wastage
C	52.1	110.8	878.8		52.4	92.4	902.5		Beef: packaging = D + 4% wastage, food = total food D + 4% wastage, Cheese: packaging = D + 4.5% wastage, food = total food D + 4.5% wastage.
D (functional unit)	50	71.2	878.8	**1000**	50	47.5	902.5	**1000**	Packaging: total packaging for 1000g = 5%. Food: Total wasted food at house for 1000g (Beef = 950 x 7.5%. cheese = 950 x 5%). Total consumed food for 1000g (Beef = 950 x 92.5%, cheese = 950 x 95%).
W1	2.9	0			2.9	0			Packaging = 5% of A
W2	2.7	0			2.8				Packaging = 5% of B
W3	2.1	39.6			2.4	44.8			Beef: 4% of packaging and total food in C, Cheese: 4.5% of packaging and total food in C
W4	50	71.2			50	47.5			Total packaging in D. Beef: 7.5% of total food in D. Cheese: 5% of total food in D
Mass balance In				168.5				150.3	**Total mass of food and packaging used within the system boundaries** =Total packaging + total waste food = A + F1
Mass balance Out				168.5				150.3	**Total mass of food and packaging disposed of within the system boundaries** =Total packaging disposed of + total food disposed of = W1+W2+W3+W4

- *Packaging types*

Two types of food packages were compared, the proposed biodegradable PHA/TPS layered material and the most commonly used meat packaging material, polypropylene (PP) (based on a visual survey in local shopping centres and reports published by the industry).

PHAs are produced through bacterial fermentation of a carbon source under limited nutrient conditions with intracellular accumulation of the polymer. This can then be purified, usually through solvent extraction (Jiang and Zhang, 2013). The carbon sources are most commonly sugars derived from an agricultural feedstock (Braunegg et al., 1999) although the use of other feedstocks has also been explored (Khardenavis et al., 2005; Strong et al., 2016). Production of PHA from sucrose (from sugar-cane) was used as the default method in this study following the inventory of Harding et al. (2007). Thermoplastic starch is produced through treatment of native starch with either heat or shear in the presence of plasticisers (Jiang and Zhang, 2013). The starch can come from a variety of sources including maize, wheat, potato and cassava. Production of thermoplastic starch from maize starch and glycerol was used as the default method in this study. PP is produced from crude oil via naphtha (PlasticsEurope, 2014).

- *Food types*

Beef and cheese were selected as model food types as they are typical food types that require high-barrier packaging to prevent food spoilage. Cheese represents a mid-range impact product for emissions associated with food production and beef represents a high-range impact product (Clune et al., 2017). Australian production was modelled for both food types, with the CO_2 emissions associated with production close to the world mean in both cases (Clune et al., 2017). The allocation approach was as defined by the relevant EcoInvent databases. Further details are provided in **Table 6-2**. Only the environmental impact due to the production, transportation and storage of the food WASTE associated with the functional unit was included (this waste can occur at the supermarket or the house) as this is the portion of food production which packaging can actually influence.

- *Waste disposal scenarios*

Landfill, anaerobic digestion and composting were considered as waste disposal scenarios for the PHA/TPS package and the food so that the benefits of biological waste disposal options could be explored. It was assumed that landfill was the only waste disposal option for PP due to a range of barriers still slowing the uptake of recycled materials in food contact applications (Australian Packaging Covenant, 2014). Incineration was not considered, as it is not a common practice in Australia (Randell et al., 2013).

- *Food wastage levels*

It was assumed that the packaging could play a role in increasing the shelf life of the food through providing improved barrier properties. This was assumed to lead to a decrease in food wastage levels at both the supermarket and the house. However, there is no specific numerical relationship that describes how increased shelf life translates into reduced food wastage.

Table 6-2: Inventory inputs and GWP100 results for the modeled system

Process	Description	kg CO_2e/kg processed
Production of the packaging	**PHA/TPS:** The production of PHA from maize was based on data published by Harding et al. (2007). Thermoplastic starch production was from maize starch, glycerol and water. Maize and glycerol production processes were from Ecoinvent.	3.35
	PP: Polypropylene granulate production and extrusion process were from Ecoinvent.	3.41
Production of the food	**Beef:** The inventory for cattle production was from personal correspondence (Tim Grant, Lifecycles). Water use was taken from the report by Mekonnen and Hoekstra (2010).	Beef fillet = 26.0
	Cheese: The inventory for milk production was from a Dairy Australia report (Dairy Australia, 2012). Water use was taken from the report by Mekonnen and Hoekstra (2010).	9.06
Production of the packaged product	A model was created to represent the inputs for production of the packaged product. Waste was taken as 5% packaging waste and 0% food waste.	
Storage at the supermarket	Food waste at the store was modelled as 4% meat waste or 4.5% dairy waste according to a report by the European Comission (European Commission, 2015).	
Storage at the house	Food waste at the house was modelled as 7.5% meat waste or 5% dairy waste according to a report by the European Comission (European Commission, 2015).	
Waste processing	**PP:** The Ecoinvent process for disposal of PP to a sanitary landfill was used.	0.133
	PHA/TPS: **Landfill:** The AusLCI process for foodwaste in landfill was used as the model with minor adjustments. **Anaerobic Digestion:** The AusLCI process for AD of foodwaste was used as a model with the production of compost and methane. Methane combustion was assumed to offset electricity (average electicity from the Victorian grid) whilst compost use offset fertiliser application. **Composting:** The AusLCI process for aerobic composting of foodwaste was used as the model and compost use offset fertiliser application.	**Landfill:** 2.13 **AD:** -1.86 **Composting:** -0.0337
	Food: **Landfill:** The AusLCI process for foodwaste in landfill was used as the model. **Anaerobic Digestion:** The AusLCI process for AD of foodwaste was used as a model with the production of compost and methane. Methane combustion was assumed to offset electricity whilst compost use offset fertiliser application. **Composting:** The AusLCI process for aerobic composting of foodwaste was used as the model and compost use offset fertiliser application.	**Landfill:** 1.29 **AD:** -0.119 **Composting:** -0.0591

6.2.4 *Inventory inputs*

Mass flows and inventory inputs were collected for each section of the supply chain from the databases Ecoinvent (version 3.2), AusLCI (version 1.26) and AustralasianLCI (version 2011.8) as well as a variety of publications, reports and personal correspondences. Energy use was adjusted to reflect the Australian grid for all background processes taken from Ecoinvent. Simplified descriptions of the inputs for the modelled system as well as selected GWP100 results (for comparative purposes) are presented in **Table 6-2** (A more detailed description of the inventory inputs can be found in Appendix B, **Table B2**). Transport was included for all processes and descriptions for the vehicle models are presented in **Table 6-3** whilst distances are shown in **Table 6-1**. All transport distances were estimates from Google Maps.

Table 6-3: Descriptions for the transport models

Transport type	Description
Non-refrigerated ground transport	A model for articulated truck freight with 100% backhaul, 90% rural driving from AusLCI was used.
Ground transport	A model for articulated truck freight with 100% backhaul, 90% rural driving from AusLCI was modified by including additional fuel consumption for refrigeration of 2.875 L/h.
Ocean transport	A model for transoceanic freight ship from AusLCI was used.
Personal transport	A model from AusLCI for the average fuel use of a car per km travelled was used with 30% of the trip to the store allocated to the functional unit and 100% of the return trip was allocated to alternative activities.
Waste transport	A model for municipal waste collection service from Ecoinvent was used.

6.2.5 *Sensitivity analysis inputs*

Low and high range scenarios for the emissions associated with production of the packaging, production of the food and the waste processing were included for sensitivity analysis and are presented in **Table 6-4**. The results obtained when using these alternative scenarios are then presented as ranges in the main results. The sensitivity analysis is particularly important given that the data for each of these different processes were taken from different sources, which may not have had consistent allocation approaches or boundary conditions.

- *Production of the packaging*

Both PHA and starch can be produced from a variety of different sources as described in section 2.3.2.1. This can introduce a large variability in the GHG emissions associated with production. Furthermore, choices concerning farming methods, production technologies, energy use, and energy source can also influence the environmental outcome (Yates and Barlow, 2013).

PHA: Based on a review of biopolymer LCAs by Yates and Barlow (2013), the lowest and highest GWP100 values were selected to demonstrate the possible range. The PHA production modelled in the main system corresponded to mid-range when compared to this review. The most extreme range was associated with genetically modified plant production of the polymer, with environmental impacts varying based on the type of electricity input used (biomass vs. coal) (Kurdikar et al., 2000).

Starch: Fewer reports have been published on the LCAs of pure starch materials and the range recorded is much smaller than for other biopolymers (Hottle et al., 2013; Yates and Barlow, 2013). Due to the lack of literature, the range for TPS production was determined from the range of starch production processes present in Ecoinvent.

- *Production of the food*

The environmental impact of food production can vary substantially due to differences in either physical factors such as geography and production methods or methodological factors such as system boundaries and allocation method (Clune et al., 2017). Based on a comprehensive meta-analysis by Clune et al. (2017) (which collated all the reported GHG footprints of production for different food types) a range for the impact of food production was tested. The upper quartile and lower quartile values were used as representation of the most probable range.

- *Waste processing*

The influence to the results due to disposal of the packaged product in either a sub-optimally functioning composting or AD facility, or state-of-the-art landfill facility were explored. To achieve this, offsets (i.e. compost and electricity generation) were removed from composting and AD scenarios, whilst 97% methane capture (Scheutz et al., 2009) was modeled for the landfill. These extreme scenarios, although perhaps unrealistic, were selected so that 'what-if' situations could be understood. For example, it allows an understanding of whether landfilling is an inherently poor option for biodegradable plastics (from a GHG emissions perspective) or whether it would be on par with AD if gas capture infrastructure could be improved. These extreme scenarios also allow for a visualisation of the entire range of results.

Table 6-4: Description of the scenarios tested as part of the sensitivity analysis for GWP100

Process	Parameter	Results from the LCA (kg CO_2e/kg processed)	High alternative value (kg CO_2e/kg processed)	Low alternative value (kg CO_2e/kg processed)	Reference for alternative values
Production of the packaging	Emissions: PHA production process	2.87	5.7	-4.0	Yates and Barlow (2013), Harding et al. (2007) and AusLCI
	Emissions: TPS production process	1.35	2.31	1.24	Rossi et al. (2015) and Ecoinvent.
Production of the wasted food	Emissions: beef production	26.0	31.6 (upper quartile value)	22.26 (Lower quartile value)	Clune et al. (2017)
	Emissions: cheese production	9.06	9.58 (Upper quartile value)	7.79 (Lower quartile value)	Clune et al. (2017)
Waste processing	AD: Compost offsets	340 (food), 150 (biopolymer)	N/A	0	N/A
	Compost: Compost offsets	450 (food), 420 (biopolymer)	N/A	0	N/A
	Landfill: gas capture efficiency	0	0.97	N/A	Scheutz et al. (2009)

6.3 Results and discussion

6.3.1 *Comparison of packaging materials (not including food impacts)*

Firstly, the CO_2e emissions of the different food packaging materials in isolation (no food included in the system boundary) were considered so that different contributors within packaging production and disposal could be analysed (**Figure 6-2**). The results of this study fall within the range of life-cycle impacts for biodegradable polymers reported in the literature (Hottle et al., 2013). The production of the polymers emerges as a significant GHG contributor relative to polymer processing and distribution, although there is little difference between the production of a PP package and the alternative biodegradable package. Of note though, as indicated by the range bars which represent the emissions associated with alternative scenarios, is that because biopolymer production is a fairly young industry, there is large variability for the GHG emissions associated with their production depending on production method and feedstock (Hottle et al., 2013). This means there is the potential that as the biopolymer industry matures, production emissions will be reduced. Methane emissions in landfill lead to the biggest difference between the two packages, as although the PHA/TPS packaging is biogenic and produced from materials that absorb CO_2 during growth, landfilling of these materials results in methane emissions which result in a higher global warming potential than the release of CO_2 (Heyde, 1998). The base scenario presented here is for a landfill with 0% methane capture whilst the range shows an upper-value scenario where 97% of methane is recovered. In practice there are only a few state-of-the-art landfills which demonstrate a 97% methane capture efficiency (Scheutz et al., 2009) and the Australian average is 30% methane capture efficiency (Australian Greenhouse Office Department of the Environment and Heritage, 2011). These results indicate the large variability in environmental impact biodegradable plastics will have depending on methane capture rates during waste processing. Transport (excluding personal car transport) was a minor part of the life-cycle emissions with fuel use allocated on a mass basis (e.g. if the packaging accounted for four percent of a food and packaging product by mass, then four percent of the fuel use was allocated to the packaging).

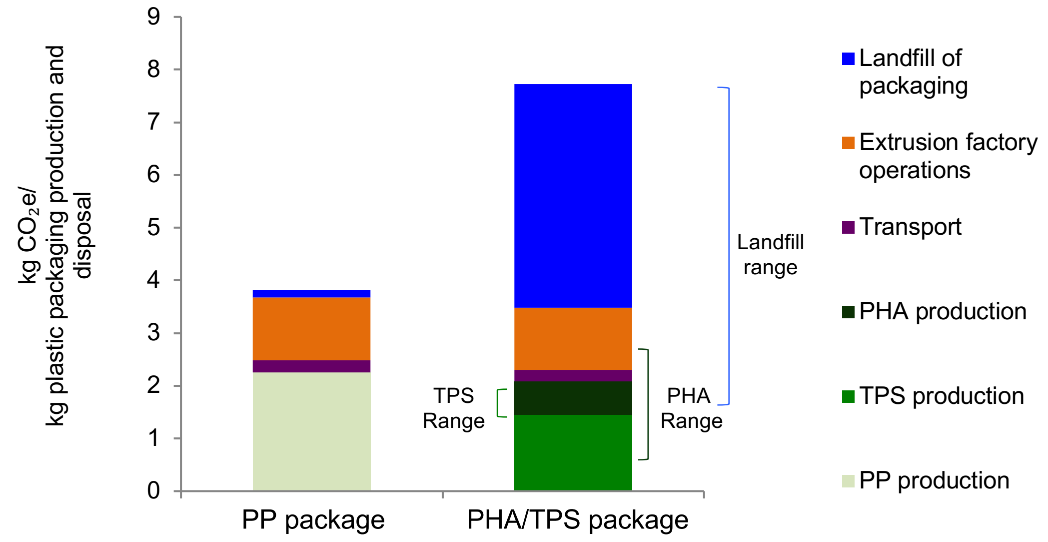

Figure 6-2: kg CO_2e emissions (GWP100) for the production and disposal of 1kg of PP or PHA/TPS food packaging

Note to Figure 6-2: The ‘use’ phase (i.e. food production and transport and storage at the supermarket and house) of the packaging is not included. The range bars indicate the variability for emissions associated with alternative starch and PHA production methods as well as indicating the reduction in landfill emissions if a 97% methane capture efficiency was achieved.

6.3.2 *Including food waste in the system boundary*

Whilst it is important to understand the specifics with regards to the production of the plastic packaging, it is also important to understand its impact in the context of the food packaged within it. As shown in **Figure 6-3**, once the wasted food is included in the system boundary, the impact of the production of the packaging becomes a small part of the impact of the total system. This finding has been reported in previous studies (Conte et al., 2015; Williams and Wikström, 2011), and the considerable impact of food production and wastage has also been noted for other indicators such as acidification and eutrophication (Silvenius et al., 2014). Calculating either GWP100 or GWP20 delivers the same message, although it is more pronounced when using a twenty-year time horizon. Due to methane emissions associated with both food production and waste disposal in landfill, these sectors show an increase in the magnitude of the impact relative to the other aspects of the system when moving to a shorter time horizon.

Figure 6-3A shows a beef scenario with disposal in landfill whilst **Figure 6-3B** shows a cheese scenario with disposal in landfill. Whilst in both cases, production of the food that is wasted is the greatest area of impact; it is evident that different food types change the balance between the impact of the food versus the impact of the packaging versus the impact of waste disposal. Cheese production dominates the graph to a lesser extent than beef production. Given that food production has such a large associated impact, there was the potential that variation in food production footprint would drastically change the results, so alternative food production scenarios were considered, and the range bars represent these results in the figures. However, in every scenario wasted food production still remains the most dominant footprint.

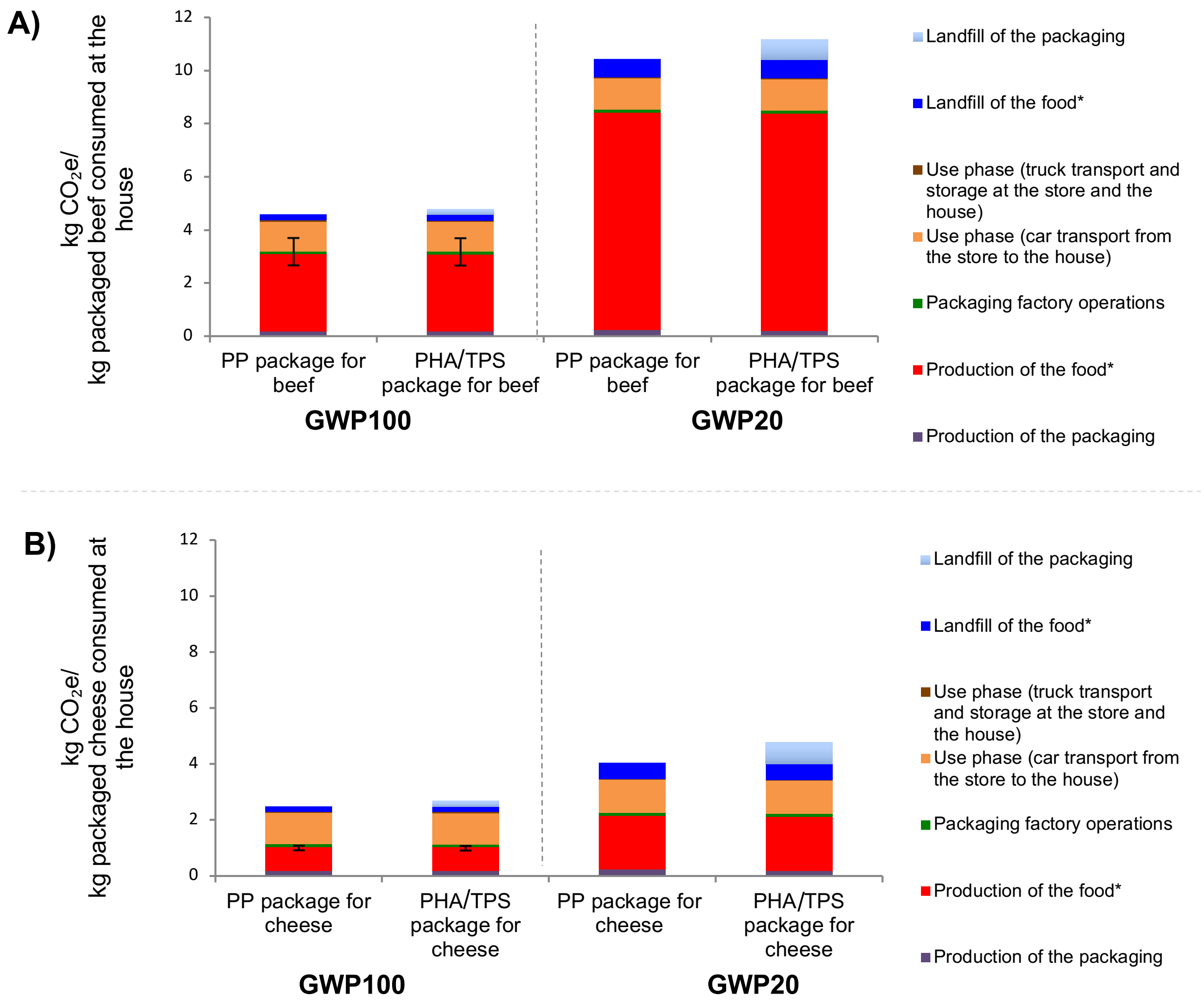

Figure 6-3: kg CO_2e emissions for the full system boundary for both twenty-year (GWP20) and 100-year (GWP100) time horizons

Note to Figure 6-3: A) for 1kg of packaged beef consumed at the house; **B**) for 1kg of packaged cheese consumed at the house. The range bars indicate variation associated with changing food production.
*Only the impact of the production and disposal of the wasted food associated with 1kg of the packaged food product at the house is included in the system boundary.

This analysis reveals two sectors that have substantial associated GHG emissions which can be reduced through packaging systems design and should be further investigated if the aim is to understand how a biodegradable, multi-layered packaging could offer sustainability improvements. The associated opportunities are (i) to improve waste management and (ii) to reduce food wastage. Personal car transport contributes a large percentage of the systems GHG emissions and this is supported by literature (Gruber et al., 2016). However, this cannot be influenced by packaging systems design and is thus not considered further.

6.3.3 *Considering waste management options*

Biodegradable packaging has an inherent value compared to non-biodegradable packaging as it presents an opportunity to direct food away from landfill (Razza and Innocenti, 2012). This is important as it has been reported that the waste sector accounts for 3% of total GHG emissions with food waste in landfill contributing to half of this (DEFRA, 2011). Anaerobic digestion and composting are considered to be the best options for food waste disposal (Eriksson et al., 2015). The greatest value for this would be in situations where there is a higher proportion of unopened, packaged food that needs to be disposed of, such as from supermarkets. Legislation has been introduced to encourage more effective waste management, with countries such as Sweden setting environmental targets for 50% of food waste from supermarkets and bulk food preparation to be biologically treated by 2018 (Eriksson et al., 2015).

Thus, the implications of different waste handling technologies were considered and are presented in **Figure 6-4** (with the assumption that the alternatives can only be accessed for the biodegradable packaging). Results are similar for the cheese scenarios and are presented in Appendix B (**Figure B1**). Each of the alternative options are benchmarked to the emissions from '1kg of PP packaged product at the house, disposed of in landfill with 0% methane capture' and the results are shown as percentage differences. All of the scenarios perform better than the benchmark but it is interesting to note that the two most commonly proposed alternatives for food waste handling (composting and AD) do not perform better than recovering full methane from landfill (although, as this is just a GHG focused study other factors such as nutrient recovery have not been considered, also, as noted previously, 97% methane capture in a landfill is very optimistic). However, this demonstrates that it is avoidance of methane emissions that is important, not necessarily the specific way in which this is achieved.

The breakdown within the AD and composting scenarios show that a majority of the benefit of these alternative processes is derived from diverting food waste from a 0% methane capture landfill.

Brancoli et al. (2017) recently reported that if food is disposed of in its packaging from supermarkets to an AD facility, up to 40% by mass of this food will then be rejected along with the plastic fraction. This sort of scenario could be avoided if the disposed food was packed in biodegradable packaging, leading to a reduction in GHG emissions from the waste sector.

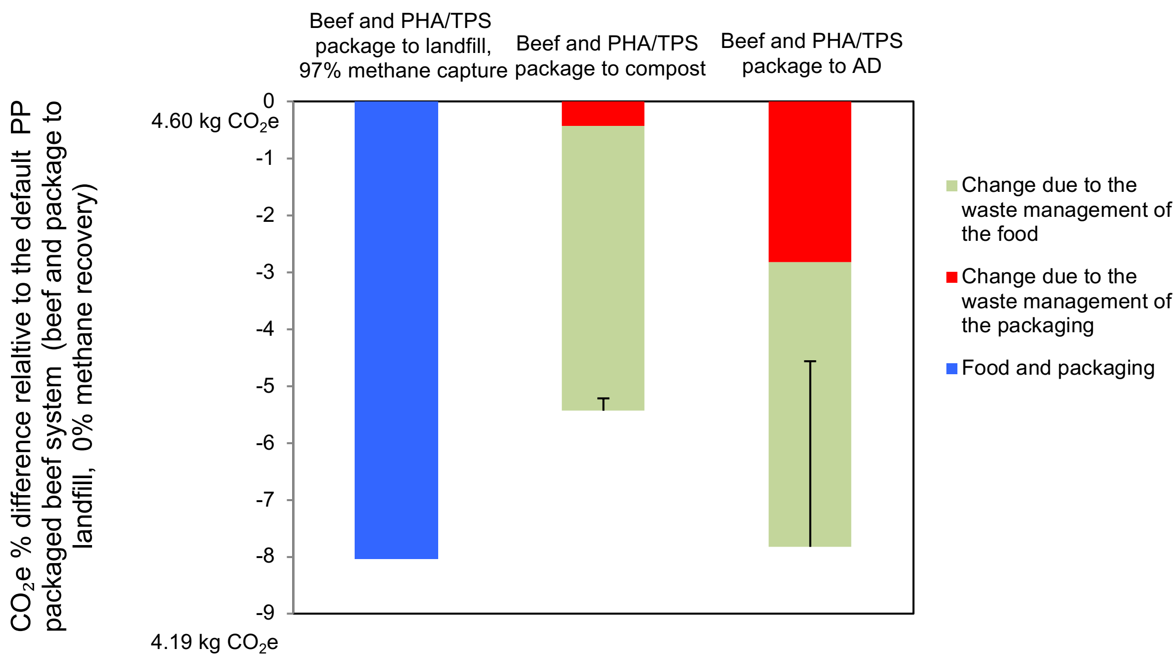

Figure 6-4: Implications for the GWP100 of a PHA/TPS beef packaged product under different waste disposal scenarios

Note to Figure 6-4: The scenarios are benchmarked to '1kg of PP packaged product at the house with disposal in landfill with 0% methane capture'. The range bars indicate the changes associated when material offsets (i.e. compost and electricity production) are not included and demonstrate a 'worst-case' scenario.

6.3.4 *Considering food waste prevention*

Redirecting inevitable food waste to a more appropriate waste management system is important but there is general agreement that prevention of food waste should be a top priority when designing sustainable food packages (Grönman et al., 2013). This redesign also carries an economic incentive for food distributors. There are a variety of behavioural factors that would play the central role in reducing food waste, but as discussed, packaging can play a role through limiting the exposure of the food to oxygen and water.

Figure 6-5 and **Figure 6-6** present the associated difference in GWP100 for a variety of beef waste scenarios and disposal scenarios. In **Figure 6-5** the scenario is benchmarked relative to 'a PP package disposed of in landfill with 0% methane recovery' whilst in **Figure 6-6** the scenarios are benchmarked to 'a PP package disposed of in landfill with 97% methane recovery'. **Figure 6-5** indicates that when landfill methane is not captured, the GHG emissions of the biodegradable scenario are ~7% higher than a PP scenario when food wastage is considered to be equal. However, if the package can reduce food wastage by ~6% (through reducing food spoilage) the emissions associated with the biodegradable package in landfill can be negated. It is therefore possible that the GHG benefits in reducing food spoilage could more than outweigh the GHG burdens of making and disposing of a PHA/TPS food-packaging product for beef, even with disposal in a 0% methane capture landfill. When using a twenty-year time horizon (results presented in Appendix B, **Figure B2**) a slightly greater reduction in food wastage is required to overcome the impact of packaging in landfill (~8%).

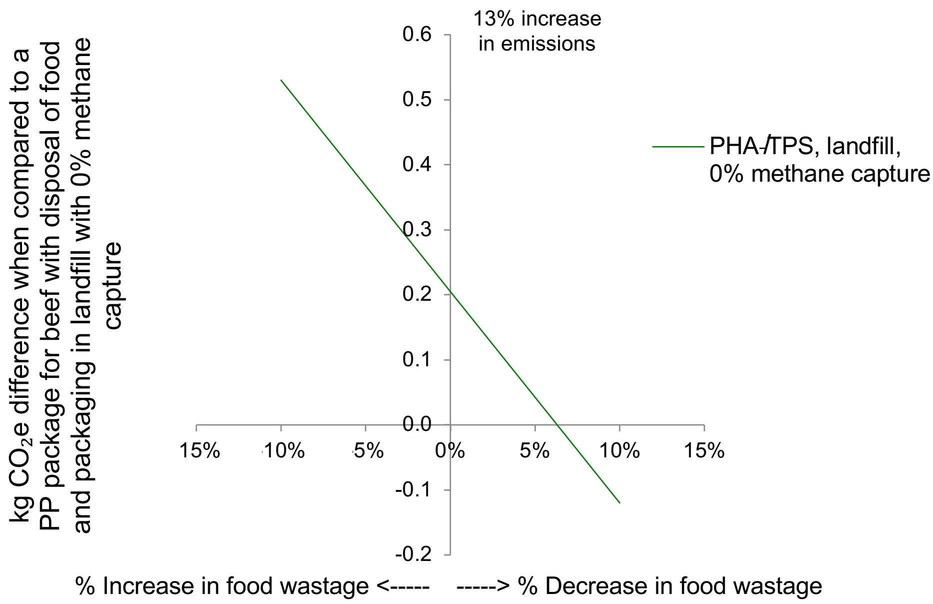

Figure 6-5: kg CO_2e difference (GWP100) for a variety of beef wastage scenarios with disposal in a 0% methane capture landfill

Note to Figure 6-5: The results are calculated relative to a 'PP beef package disposed of in landfill with 0% methane capture'.

On the other hand, as presented in **Figure 6-6**, even under conditions of maximum methane recovery, if a biodegradable packaging does not match the functional performance of a PP package, a small increase in food waste can negate any benefits obtained by disposal in optimised waste management infrastructure.

The combination of these results demonstrates that if all methane emissions associated with waste management can be captured, AND food wastage can be reduced then the reduction in GHG emissions of using a PHA/TPS material relative to a PP material can be maximised. It is reasonable to believe that for the proposed PHA/TPS package these levels of food waste reduction could be achieved. A 15% increase in the shelf life of beef has been reported for other starch-based packaging materials compared to traditional systems (Cooper, 2013), although it is not exactly known what percentage reduction in food waste this correlates to. The benefit of a biodegradable package is also that it enables more flexibility with regards to waste processing options.

Results for a cheese scenario, presented in **Figure B3** in Appendix B, show similar trends, however, because cheese has a lower GWP associated with its production, a greater reduction in food waste would be required to overcome the emissions of biodegradable plastic in landfill. In this case alternative waste management systems are of greater importance.

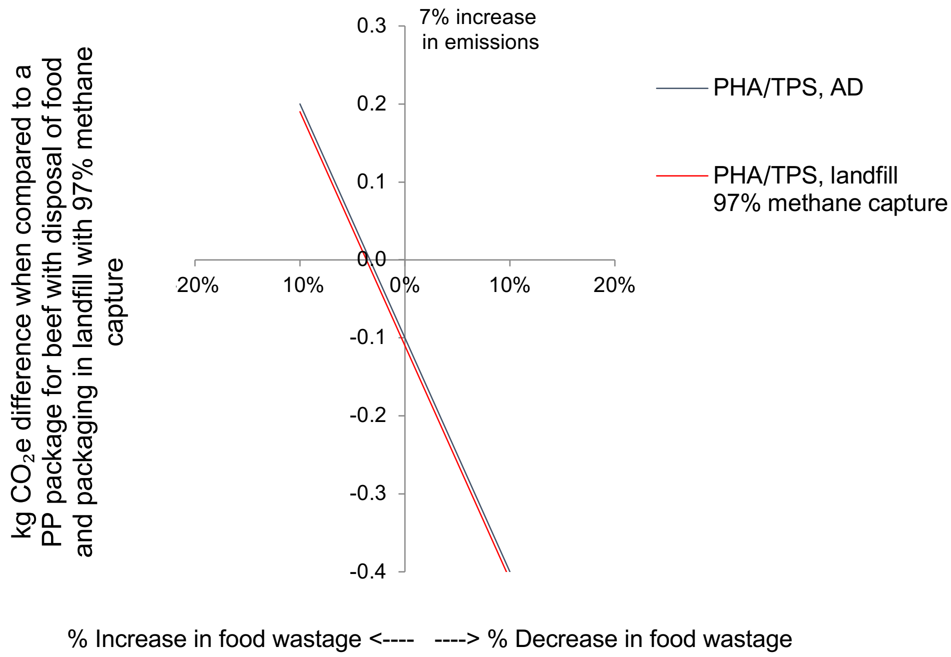

Figure 6-6: kg CO_2e difference (GWP100) for a variety of beef wastage scenarios with disposal in a 97% methane capture landfill or AD

Note to Figure 6-6: The results are calculated relative to a 'PP beef package disposed of in landfill with 97% methane capture'.

6.3.5 *Considering potential impact*

Further to the results presented, the opportunities should be considered in light of relative consumption levels of the food products. In Australia, approximately 580,000 tonnes of beef (Meat and Livestock Australia, 2016) and 260,000 tonnes of cheese (Dairy Australia, 2017) are domestically produced and consumed per year. Given that more beef than cheese is consumed, and also that it has a higher wastage rate and carbon footprint of production, it appears that beef would be the more immediate focus for high-barrier, biodegradable packaging applications. However, to provide quantitative conclusions as to the total potential impact of the new packaging material, the following parameters would need to be known:

-The frequency of each food type being disposed of in packaging;

-What percentage of food is wasted due to reaching its 'use-by-date';

-What the quantitative relationship is between increased food lifetime and reduced food wastage.

Confirmation of the PHA/TPS packaging's ability to reduce food spoilage is also required.

6.3.6 *Water use*

Although the initial scope was limited to GHG emissions, it is useful to know whether considering other resources common to both the food and packaging sectors would change the conclusions of the study. The results for a water use analysis are presented in **Figure 6-7**. Approximately 60% more water is required to produce the PHA/TPS packaging when compared to a PP packaging, however, similar to the GHG analysis, this is dwarfed by the water use for the production of the wasted food. Thus, the change in the consumption of water in producing the packaging is minor, but any reductions in food wastage could substantially reduce the water use of the system. This supports the original conclusion that reducing food wastage should be a key consideration in biodegradable packaging design.

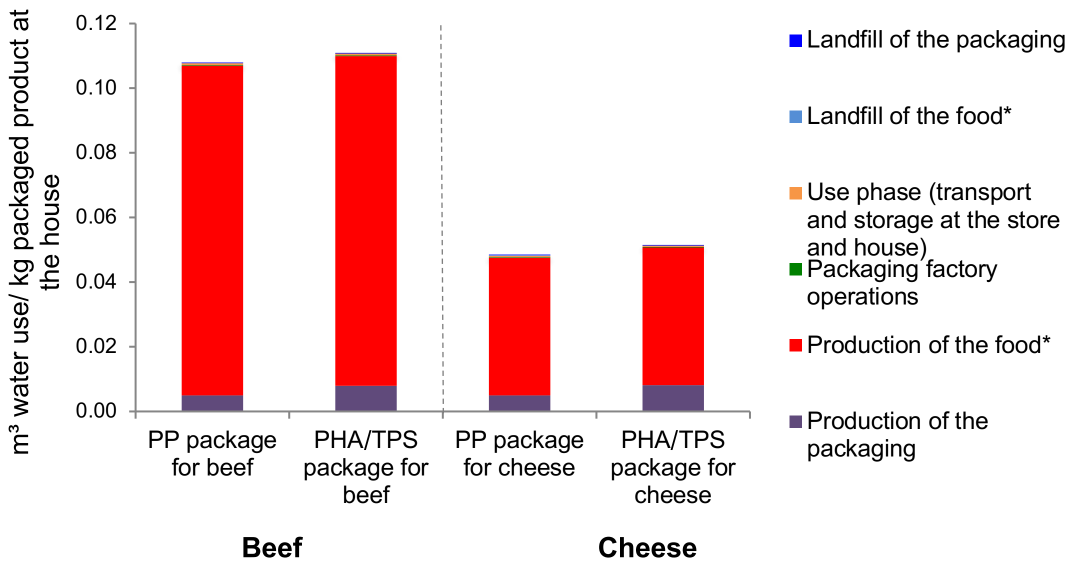

Figure 6-7: Water use by sector for the full system boundary

Note to Figure 6-7: *Only the impact of the production and disposal of the wasted food associated with 1kg of the packaged food product at the house is included in the system boundary.

6.4 Conclusion

On a basic level, the results of this work show that the main differences in GHG emissions between a PP and PHA/TPS food packaging are due to landfill emissions. However, this only holds true until food waste is included in the system boundary, at which point differences become dominated by changes in the quantity of food waste. This then leads to three main insights. Firstly, that food packaging design needs to focus on the reduction of food waste (e.g. focus on high barrier properties), even if a biodegradable material is used. Secondly, that the GHG emissions associated with disposal of a PHA/TPS packaging in landfill can be offset if the package reduces beef wastage by approximately 6% (demonstrating the viability of a high-performing biodegradable packaging providing GHG benefits even if disposed of in landfill). Thirdly, that a biodegradable packaging could provide GHG benefits through increasing the amount of food waste available for biological processing (e.g. anaerobic digestion with subsequent biogas processing). As a final note and word of caution, it should be acknowledged that whilst this LCA has provided some interesting comparisons between biodegradable and conventional plastic packaging, it can only tell a small part of the story. LCA is a useful tool but is currently not configured to quantify many of the important environmental impacts associated with plastics, particularly accumulation in the oceans.

CHAPTER 7 Public attitudes towards plastics

Chapter Summary

Within the social attitudes theme, the objective was to '*Examine public beliefs and attitudes towards plastics and bioplastics in Australia'*. This chapter presents the first of two research activities that relate to this research objective, focusing just on conventional plastics, with bioplastics the subject of **Chapter 8**. All data for the social attitudes theme were collected through a survey; the full set of questions are included in Appendix E. Overall, the survey results indicate that the public view plastics as a serious environmental issue. Plastic in the ocean had the highest mean rating for seriousness out of nine environmental issues – including climate change – followed by two other issues relating to plastic waste production and disposal. However, the public place the bulk of the responsibility for reducing the use of disposable plastic on industry and government.

This chapter was published in full as a journal article during the candidature.

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Consistent with the requirement for research involving human participants, ethical approval was granted by the University of Queensland's 'Engineering, Architecture and Information Technology, Low and Negligible Risk Ethics Sub-committee' on 27 April 2018, with an amendment approved on 15 May 2018. Copies of the approval letters are included in Appendix F. This process ensures that the research complies with the provisions contained in the *National Statement on Ethical Conduct in Human Research* and complies with the regulations governing experimentation on humans.

7.1 Introduction

There is growing attention worldwide towards reducing the use of disposable plastics and transitioning towards a circular economy for plastics (Dauvergne, 2018; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). For example, at the Our Ocean Conference in 2017, there was a pledge from six major international companies to "use 100% recyclable, compostable, or reusable packaging by 2025" (Ellen MacArthur Foundation, 2017; European Commission & European External Action Service, 2017). The number of companies committing to this pledge rose to eleven in early 2018 (Ellen MacArthur Foundation, 2018a). Later in 2018, over 280 organisations signed up to a global commitment aiming to eliminate plastic waste and pollution (Ellen MacArthur Foundation, 2018b). At the same time, the European Commission adopted the first ever Europe wide strategy on plastics (European Commission, 2018a). There is also a growing body of literature demonstrating the environmental presence and impact of plastics which underpins this political focus (Browne et al., 2011; Gall and Thompson, 2015; Jambeck et al., 2015; Zalasiewicz et al., 2016).

While it is clear that there is a sense of urgency and increased attention on plastics as an environmental issue by industry and governments, and that this focus is supported by science, there is a gap in the data documenting the attitudes of the general public towards plastics that warrants consideration. Whilst securing commitment from government and industry is key to solving the plastics issue, given that a large portion of plastic is used in consumer applications (PlasticsEurope, 2017), the public is also an important part of the system. It is the public who will need to interact with any changes in the plastic system and who are often responsible for disposing of plastic waste. It is clear that for any changes in the plastic system to be successful, they must not only be economical and technically viable but also socially acceptable (Gelcich et al., 2014; Pahl et al., 2017).

To date, whilst there is certainly an extensive body of literature on public attitudes towards plastic waste, most of the research has focused on attitudes towards specific actions. For example, there have been focused considerations of topics such as: the outcomes of trialling a new recycling initiative in a university setting (Cheung et al., 2018); the factors influencing an individual's motivation to recycle including the role that the convenience of a recycling scheme and general environmental attitudes have on participation levels (Best and Kneip, 2011; Huffman et al., 2014; Saphores and Nixon, 2014); behaviour relating to the reuse/recycling of plastics (Babader et al., 2016; Khan et al., 2019) including what difference there is in the factors that motivate waste minimisation behaviour versus recycling behaviour (Barr et al., 2001; Tonglet et al., 2004); and factors leading to lifestyle changes such as a switch to using cloth bags from plastic bags (Ari and Yilmaz, 2017).

These more targeted studies are important. However, in light of the global movement to more effectively manage the plastic waste issue, it is also important to understand the broader backdrop of societal attitudes towards plastics. To achieve at scale the changes required to improve the sustainability of the plastics system, significant disruption will probably be required — as is already evident in the growing focus on bans on certain single-use products. A focus on the public's attitudes towards the broader topic of plastics as an environmental issue is important if we are to understand whether there is acceptance from the public for this focus on plastics and whether they might support such disruptive solutions.

Published literature on the broader scope of public attitudes towards plastics is extremely scarce. This type of research can be found for topics such as public perception and understanding of environmental issues like climate change (Chilvers et al., 2014; Nisbet and Myers, 2007) and waste (European Commission, 2017), but not for plastics. Globally, one of the only relevant examples for understanding public attitudes towards plastics is the 2017 Eurobarometer Survey (European Commission, 2017). However, its relevance and insight are limited, as it only asked a single question on plastics, with results showing that 87% of respondents at that time were worried about the impact of plastic products on the environment.

Thus, given the absence of public attitudinal research towards plastics from a non-targeted perspective, but rapidly increasing interest at government and industry level, this research was undertaken to obtain the much-needed data on public attitudes towards plastics. The central aim of this research is to identify whether the general public views plastics as a serious environmental issue. Secondary aims include to understand what factors influence attitudes towards plastics, and to explore whether those attitudes motivate any personal reduction in plastic use. These aims also feed into understanding whether there is support for an industry and government intervention that focuses on reducing plastic use, and to what extent the public holds them responsible for addressing the issue. These aims were achieved by undertaking a survey of the Australian public and provide a comparison point for other countries' attitudinal research.

7.2 Methodology

The formal data collection was performed by Survey Sampling International, a market research company, from 16th May 2018 to 22nd May 2018. In total, 3028 respondents started the survey with

2529 completing it (the retention rate was 83.5%). Of the completed responses 2518 were retained for the analysis (with the data cleaning protocol being described in Section 7.2.3). All response collection was performed by the market research company via their standard practices which included emailing the survey to a sample of their database and rewarding respondents for completing the survey with a small voucher. Recipients of the initial email that had not responded were emailed several times to try and encourage them to respond. The market research company selected respondents using the quota method, meaning that the sample selected was representative of gender, age and state for the Australian population. The result is that the survey can be considered to be representative of the Australian population with all demographics collected closely matching those from Australian census data (results presented in Appendix C)(Australian Bureau of Statistics, 2018, 2016). The gender split was 47% male, 53% female; all age groups were adequately represented; each of the states and territories received even representation relative to population; educational level, occupational status and income bracket were all evenly represented; and there was an even spread of political leanings with the majority of respondents placing themselves as centrist.

7.2.1 *Australia in context*

Australia has an internationally competitive, advanced market economy and in 2017 was the 13th largest national economy by nominal GDP (The World Bank, 2019). It is a significant exporter of goods (including food) (Central Intelligence Agency, 2019). It has a population of 23 million and a multi-cultural population base (Central Intelligence Agency, 2019). Although attitudes will vary across nations (Herbes et al., 2018), given the status of Australia as an important global presence, a study of the Australian population provides globally relevant insights.

7.2.2 *Survey development*

This research work was motivated by two factors - a suspicion that there is growing public interest in plastics as an environmental issue, and a realisation that there was little reliable literature in the space of broader attitudes towards plastics as a material, not specific interventions. Between 2015 and 2018 there were a number of prominent projects and seminal publications that aimed to catapult plastics and waste onto the main stage, as evidenced by the Ellen MacArthur Foundation's 'New Plastic Economy' report (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016), David Attenborough's 'Blue Planet II' documentary, and the Science publication by Jambeck et al. (2015). The quantification of the production, use and fate of all plastics ever produced (Geyer et al., 2017) as well as the recent Chinese import ban on plastic waste (Brooks et al., 2018) also contributed to increased attention on this issue.

Despite anecdotal evidence that public interest in the impacts of plastics was increasing, a dedicated research tool for capturing these attitudes did not exist. Hence, this survey was developed. Inspiration was taken from a variety of periodical environmental surveys (including: an OECD survey on Environmental Policy and Individual Behaviour Change which included questions on the seriousness of different environmental issues (but not plastics), questions relating to general environmental concern, and questions relating to waste production and recycling behaviour (OECD, 2014); a European waste survey that included questions relating to plastics and personal health as well as the role of the individual, governments and businesses in addressing environmental issues (European Commission, 2017); and an Australian 'Who cares about the environment?' report which asked questions about the environment in general (Department of Environment and Conservation NSW, 2017)).

The survey was refined through several rounds of prototyping within the authors' research groups as well as selected members of the public, meaning a diverse range of knowledge levels as well as age groups were consulted. This helped with the development of the focus of the survey and the ensuring accuracy of interpretation of the questions. After this, the survey was piloted with a random sample of 250 members of the general public by Survey Sampling International (the market research company). After minor changes during the prototyping, the amended survey was used for formal data collection. The full list of questions relevant to this chapter can be found in Appendix E.

The survey included a mix of Likert scale, multiple-choice, and some open-ended questions. The open-ended questions were positioned at the beginning of the survey with the aim of giving respondents the opportunity to present their opinions on plastics without being influenced by the wording and context of the survey. This method enabled the issues that first came to mind with immediacy and availability to be captured (Chilvers et al., 2014) and has been used in other large-scale studies for the same purpose (Chilvers et al., 2014; Lorenzoni and Pidgeon, 2006; Sherry-Brennan et al., 2010).

7.2.3 *Data cleaning and analysis*

Data were cleaned and analysed using SPSS version 25.0.0.0. Initial data cleaning was performed by Survey Sampling International which included removing any respondents who completed the survey in less than one third of the median time (22 minutes) and removing anyone that responded with unintelligible answers for the open-ended responses. This is to ensure data quality (Zhang and Conrad, 2014). Secondary cleaning was performed by the authors and included removing any respondents that had no variation in their pattern of response selection (e.g. had selected the highest option for every question, even if this led to a contradiction). This is known as nondifferentiation and can affect both validity and reliability of the response (Yan, 2008). After all of the data cleaning, 2518 data points remained for the final analysis.

Chi-squared tests for independence were used to determine whether there were significant differences between categorical variables using a cut-off of $p = 0.01$. Cramer's V was used to determine effect size with the following interpretation: $\varphi C < 0.1$ (negligible effect), $\varphi C < 0.2$ (weak effect), $\varphi C < 0.3$ (moderate effect) (Rea and Parker, 1997). Significant differences between means were calculated using a paired t-test ($p = 0.01$) and Eta squared (η^2) was used to determine the effect size with the following interpretation: $\eta^2 < 0.01$ (negligible effect), $\eta^2 = 0.01$ (small effect), $\eta^2 = 0.06$ (moderate effect) and $\eta^2 = 0.14$ (large effect) (Pallant, 2016). Open-ended questions were analysed through inductive content analysis (Vaismoradi et al., 2013). This was performed manually by L.D.H (main author), with coding consistency being checked by P.L and B.L (co-authors).

Factor analysis

In order to explore the underlying structure of attitudes towards plastics, exploratory factor analysis was initially performed on 13 items from the questionnaire according to the method outlined in Pallant (2016) but using a principal axis factoring method. Any variables that had a loading of below 0.45 on all factors or had a communality below 0.4 were removed (Comrey and Lee, 1992; Tabachnick and Fidell, 2007). The final factor analysis was performed on 10 variables. This technique was deemed to be appropriate as Bartlett's test of sphericity was statistically significant (at $p < 0.001$) and the KMO measure of sampling adequacy was 0.806 (exceeding the recommended value of 0.6) (Pallant, 2016). To aid in interpretation of the factors, oblique (direct oblimin) rotation was used (as there was found to be correlation between the factors). The decision to extract three factors (which explained a total of 62% of the variance) was based on an initial two factor solution producing two variables with equivalent loadings on both of the factors. For the final three factor solution, all of the factors had a number of strong loadings, with all variables only loading on one factor (all > 0.7 loading).

7.3 Results and Discussion

7.3.1 *Plastics are viewed as a serious environmental issue*

The central aim of this research was to understand whether the public view plastics as an environmental issue of particular concern, with the Australian public used as the data set. To explore this, one of the first questions in the survey asked respondents to indicate how serious they thought nine environmental issues were using a scale of 1 (not serious) to 10 (extremely serious) (refer to

Table 7-1). Plastic in the ocean had the highest mean score, with almost 70% of participants rating it as either 9 or 10, with only 3.3% of respondents rating it as 5 or less. All items relating to plastics or waste had the three highest mean scores. Forty-eight percent (48%) of respondents also gave plastic in the ocean a higher rating than climate change. Climate change was actually the environmental issue with the lowest mean score, and with the greatest range of responses. A large number of respondents (~45%) still rated climate change as a 9 or a 10, but the mean score is reduced by almost 16% of respondents rating it less than 5. Given previous work showing divided opinions on the topic of climate change in Australia, with 7% of respondents believing that climate change is not happening, and 15% of respondents being unsure (Ashworth et al., 2011), this spread in responses is not surprising.

Table 7-1: Responses to '*please indicate how serious you think each of the following environmental issues are'*

Environmental Issue	Response selection (%)											Mean rating	Standard Deviation
	Not serious 1	2	3	4	5	6	7	8	9	Extremely serious 10	Don't know		
Plastic in the ocean	0.4	0.4	0.1	0.4	2.0	3.3	8.6	14.7	20.4	48.5	1.2	8.9	1.49
The amount of plastic waste produced	0.5	0.2	0.6	0.8	2.6	4.3	11.3	19.4	20.9	37.9	1.5	8.59	1.58
The amount of general waste going to landfill[a]	0.4	0.4	0.4	0.7	2.9	6.3	12.1	21.2	20.4	34.0	1.1	8.45	1.61
Water pollution[a,b]	0.4	0.3	0.4	1.2	3.5	5.5	14.2	22.8	19.7	30.5	1.5	8.34	1.62
Endangered species and biodiversity[b,c]	0.5	0.2	0.6	1.3	4.3	6.6	15.3	20.4	16.4	31.5	3.0	8.26	1.71
Natural resource depletion (forest, water, energy)[c]	0.5	0.4	0.7	1.4	5.2	7.4	14.9	22.3	15.5	29.3	2.4	8.14	1.75
Air pollution[d]	0.8	0.6	1.1	1.4	4.1	8.3	18.7	24.1	15.1	23.2	2.7	7.93	1.78
Water shortages[d]	0.6	0.8	1.2	2.5	5.4	9.0	16.7	18.3	15.6	26.8	3.1	7.93	1.91
Climate change (global warming)[d]	3.3	1.6	2.0	2.1	6.8	7.3	12.8	18.0	13.7	29.7	2.7	7.73	2.34

[a,b,c,d] = Mean rating for the issues are not significantly different or are negligibly significantly different to each other at the p = 0.01 level. Results of statistical tests are available in Appendix C, **Table C3**.

The influence of demographics on ratings for the environmental issues of plastic and waste were then considered (**Table 7-2**). Given the absence of previous research on attitudes towards plastics, climate change was also included as a way to check the validity of this study relative to previous studies. Previous studies have found that views on climate change are significantly influenced by a variety of factors including age, gender, education, income (Department of Environment and Conservation

NSW, 2017), scepticism of environmental claims, and trust in scientific experts (OECD, 2014). Thus, the results of the current study, finding that age, gender and political leaning (political leaning being self-reported on a scale of 1 = left to 10 = right) have a significant and non-negligible influence on ratings of climate change, are in line with previous research. However, gender emerges as the only demographic that has a statistically significant and non-negligible effect on the rating an individual will give to issues related to plastic and waste. Both males and females rate the issues as serious, but females are more likely to rate them as 10 (for example, 53.4% of females rate plastic in the ocean as a 10 compared to 45.4% of males, Appendix C, **Figure C4**). These results clearly indicate that Australians currently consider plastic and plastic waste as serious environmental issues and that this concern appears to be independent of any key demographics other than gender. Even then, both genders rate it as a serious environmental issue, gender just influences the likelihood of rating the seriousness as a ten out of ten.

Table 7-2: Influence of demographics on rating of environmental issues

	Influence of demographics on response*					
	Political leaning	**Gender**	**Age**	**Education**	**Income**	**State**
Plastic in the ocean	Negligible $p < 0.001$ $\varphi C = 0.087$	**Weak** $p < 0.001$ $\varphi C = 0.117$	Negligible $p < 0.01$ $\varphi C = 0.061$	None $p = 0.173$	None $p = 0.620$	None $p = 0.536$
The amount of plastic waste produced	Negligible $p < 0.001$ $\varphi C = 0.082$	**Weak** $p < 0.001$ $\varphi C = 0.117$	Negligible $p < 0.01$ $\varphi C = 0.061$	None $p = 0.140$	None $p = 0.097$	None $p = 0.915$
The amount of general waste going to landfill	None $p = 0.020$	**Weak** $p < 0.001$ $\varphi C = 0.178$	None $p = 0.257$	None $p = 0.314$	None $p = 0.585$	None $p = 0.178$
Climate change	**Weak** $p < 0.001$ $\varphi C = 0.159$	**Moderate** $p < 0.001$ $\varphi C = 0.204$	**Weak** $p < 0.001$ $\varphi C = 0.106$	Negligible $p < 0.001$ $\varphi C = 0.079$	None $p = 0.211$	Negligible $p < 0.01$ $\varphi C = 0.064$

* Measured at the p = 0.01 level

In the absence of plastics attitudinal research, to set these results in context, literature regarding attitudes towards waste is used as a proxy. Considering the global scale, a 2011 survey of OECD countries found that waste generation was in the top three environmental issues in only a few countries (not including Australia) (OECD, 2014). The results presented in this chapter suggest that concern for plastics and plastic waste may have grown, but this remains to be confirmed by further research.

7.3.2 *Plastics are associated with food packaging, convenience and environmental concern*

Open-ended questions were included as the first questions in the survey to gather respondents' attitudes before any bias towards environmental concern was potentially introduced. The very first question asked respondents to '*please record the first two words/phrases that come to mind when you hear the word 'plastic'*'. In total, 5057 responses were received and coded. For ease of interpretation the results are presented as a word cloud (**Figure 7-1**). Most of the responses (80% of them) were categorised into three groups; *positive connotations (23%)* (including: 'recyclability' (340), 'cost (cheap)' (123), 'convenience' (100) and 'usefulness' (103)), *negative connotations (38%)* (including: 'waste/rubbish' (380 mentions), 'pollution' (242), and 'environment' (190)) and *packaging items (24%)* (including: 'bag (411), 'bottles' (344) and 'container' (115)). The remaining 20% of responses were either neutral ('barbie', 'toy', 'manufactured', 'hard') or deemed ambiguous ('disposable', 'artificial', 'plastic', 'nothing').

Figure 7-1: Responses to '*Please record the first two words that come to mind when you hear the word plastic'*.

Note to Figure 7-1: Blue = food related, red = negative connotation, green = positive connotation, grey = neutral/ambiguous, size of word relates to frequency.

The second and third questions asked respondents to list two positive or two negative words related to plastics (the order of the two questions were randomised to reduce bias). General environmental concern (754), association with waste (620) and pollution (512) and ocean impacts (500) were top of the list of why respondents viewed plastics negatively (**Table 7-3**), while convenience (509), useful material properties (463), affordability (401) and recyclability (389)/reusability (310) were the key reasons that respondents viewed plastics positively (**Table 7-4**). The association of plastics with packaging was further probed through the question '*Please choose the three product categories you most immediately associate plastic materials with*'. Eighty-eight percent (88%), 73% and 41% of respondents selected ‘food related packaging’, ‘single-use carrier bags’ and ‘all other non-food packaging’ respectively while 27% of the population chose all three (Appendix C, **Figure C1**).

The themes emerging from these three unprompted questions and the product category question show that Australians strongly associate plastics with packaging and food related items. They are also simultaneously aware of the negative aspects of plastic use (namely, the environmental impacts), and the positive aspects of plastic use (namely, the ease they bring to their lives). This suggests that environmental concerns are not the only influencer of attitudes towards plastics.

Table 7-3: Responses to ‘*Please record the first two negative words that come to mind when you hear the word plastic*’

Negative connotations	Count
General environmental mentions	754
Waste	620
Pollution	512
Ocean impacts	500
Does not break down (or similar wording)	420
Animal impacts	325
Total responses	4505

Table 7-4: Response to ‘*Please record the first two positive words that come to mind when you hear the word plastic*’

Positive connotations	Count
Convenient	509
Useful material properties (light, tough, durable, strong, waterproof, rigid, elastic, soft, hard, flexible, colourful, clear, transparent, malleable, permeable, unbreakable)	463
Affordable	401
Recyclable	389
Reusable	310
Easy	294
Total responses	4017

7.3.3 *Factor analysis supports the qualitative findings*

The underlying structure of attitudes towards plastics were further explored using a wide variety of questions throughout the survey followed by a factor analysis. The question wordings and the item loadings within the pattern matrix are displayed in **Table 7-5** whilst **Table 7-6** shows the associated descriptive statistics for each of the items. Three distinct factors underpinning attitudes towards plastics were identified during this process which are described below.

Factor 1. *Concern regarding plastic waste production and fate*. This factor captures concern regarding production and disposal of plastic waste as well as views on the severity of the environmental issues associated with plastic waste. The highest loading items relate to concern for plastic waste disposal in landfill, concern for the volumes of plastic waste production and perceived severity of plastic in the ocean as an environmental issue. Examining the descriptive statistics, it can be seen that whilst the majority of respondents indicate worry on all of the variables, the strongest responses for both concern and severity relates to plastic in the ocean (44% of respondents feel extremely concerned about plastic pollution in the oceans and 69% of respondents rate plastics in the ocean as very serious). It can also be seen that a belief in the severity of an issue is not directly related to concern as on average, levels of concern are consistently lower than perceived severity.

Factor 2. *Recognised utility of plastic food packaging*. Two items load onto this factor and focus on the convenience and usefulness of food packaging. The vast majority of respondents rate food packaging as convenient and useful, demonstrating that they recognise the personal utility of plastic food packaging.

Factor 3. *Negative perception of plastic food packaging.* This factor combines two items and captures responses relating to the perception of plastics as 'good/beneficial' or 'bad/harmful'. In general, when specifically questioned, respondents skew towards viewing plastic food packaging negatively. Interestingly, this decision appears to be distinct from environmental concern. One of the items that did not load onto this factor, indicating a different underlying construct, is rating plastic food packaging specifically on the trait 'good for the environment' or 'bad for the environment'. This receives a much stronger negative response than the two questions forming the factor. This indicates that something mediates peoples negative environmental view of plastics – and from the free-word associations this could be assumed to be either convenience or price.

Table 7-5: Pattern matrix for the factor analysis after oblimin rotation*

Question	Factor 1	Factor 2	Factor 3
Please indicate your level of concern for each of the following: Plastic being disposed of in landfill	**0.774**	-0.076	0.022
Please indicate your level of concern for each of the following: The amount of plastic waste produced daily in Australia	**0.770**	-0.083	0.006
How serious do you think the following environmental issues are? Plastic in the ocean	**0.767**	0.071	-0.001
How serious do you think the following environmental issues are? The amount of plastic waste produced	**0.766**	0.055	-0.039
What is your level of concern for the following? Plastic pollution in the oceans	**0.737**	0.019	-0.010
How serious do you think the following environmental issues are? The amount of general waste going to landfill	**0.724**	0.008	0.005
Please rate plastic food packaging against the following traits: Inconvenient/convenient	0.015	**0.796**	-0.030
Please rate plastic food packaging against the following traits: Not useful/useful	-0.021	**0.784**	0.055
Please rate plastic food packaging against the following traits: Bad/good	0.009	-0.006	**0.890**
Please rate plastic food packaging against the following traits: Harmful/beneficial	-0.012	0.015	**0.831**
Cronbach's alpha (for items included in the factor)	0.884	0.777	0.853

Note to Table 7-5: *The factors represent a grouping of the items that reflects an underlying structure across all of the items. High loadings indicate that the item is strongly related with that factor. It can be seen that all items only load onto one factor. The proposed factor structure based on high loadings is highlighted in grey. Positive and negative loadings just represent positive and negative associations respectively.

Thus, the factor analysis reveals a few interesting points. Firstly, the factors begin to give structure to the types of words volunteered in the qualitative questions at the beginning of the survey (Section 7.3.2). People are concerned about the volume of plastic waste being produced and its impacts on the land and ocean. At the same time, they recognise the utility that plastic items such as food packaging provide them individually. The complexity of our attitudes probably then arises from the interplay between these understandings.

Table 7-6: Descriptive statistics for questions included in the factor analysis

Item	% 1	2	3	4	5
Factor 1	'Not concerned' 'Not serious'				'Extremely concerned' 'Very serious'
Concern: Plastic waste disposed of in landfill	2.5	7.6	25.3	36.1	28.4
Concern: The amount of plastic waste produced daily in Australia	2.0	6.1	25.9	35.3	30.7
Seriousness: Plastic in the ocean	0.8	0.6	5.3	23.2	68.9
Seriousness: The amount of plastic waste produced	0.6	1.4	6.9	30.7	58.5
Concern: Plastic pollution in the oceans	1.2	4.2	18.1	32.5	44.0
Seriousness: The amount of general waste going to landfill	0.8	1.1	9.3	33.7	55.0
Factor 2	'Inconvenient' 'Not useful'				'Convenient' 'Useful'
Rating of plastic food packaging: Inconvenient/convenient	5.5	5.0	12.0	34.2	43.4
Rating of plastic food packaging: Not useful/useful	6.9	7.5	19.5	39.9	26.3
Factor 3	'Bad' 'Harmful				'Good' 'Beneficial'
Rating of plastic food packaging: Bad/good	27.8	34.2	23.6	12.1	2.3
Rating of plastic food packaging: Harmful/beneficial	27.1	30.8	24.6	13.7	3.8
Rating of plastic food packaging: Good for the environment/bad for the environment	61.2	22.7	10.4	3.7	1.9

7.3.4 *Australians show higher interest than action for reducing plastic use*

Next, it was investigated whether attitudes towards plastic translate into a desire to reduce plastic use and whether this desire is then translated into action. The desire to reduce plastic use was examined in two survey questions (1 = strongly disagree, 5 = strongly agree): '*I would like to reduce my use of disposable plastics* ($\bar{x}$ = 4.05, σ = 0.87)' and '*I would like to reduce my use of plastics used in longer-term applications (buy items made from alternative materials)* ($\bar{x}$ = 3.97, σ = 0.84)' (Appendix C, **Table C1**). The results reveal strong support for reducing the use of plastics and also show that this support is not only restricted to the case of disposable plastics but extends to plastics used in longer term applications as well. Respondents also appear to welcome external intervention aimed at reducing plastic use, with 80% of respondents agreeing with the statement *'measures should be taken to reduce the use of single-use plastic items (e.g. shopping bags, straws…)'* (Appendix C, **Table C2**).

In fact, reducing plastic use appears to trump other packaging considerations for many Australians. Many people do not support increased use of plastic packaging even if it provides the well-documented (Dilkes-Hoffman et al., 2018a; Williams and Wikström, 2011) (**Chapter 6**) environmental function of reducing food waste, with 37% of respondents disagreeing with the statement '*If plastic food packaging reduces food wastage that justifies its increased use*' (Appendix C, **Table C2**). These sentiments most likely stem from the fact that plastics are not currently viewed favourably as a material. Eighty percent of respondents agreed with the statement *'These days, too many items are made out of plastic'* (Appendix C, **Table C2**), whilst alternative materials received support (with paper and glass being rated as more environmentally friendly food packaging materials when compared to plastics by 69% and 64% of respondents respectively (Appendix C, **Figure C2**).

Following the questions relating to attitudes towards reduced plastic use, respondents were asked to indicate how frequently they performed activities to reduce their use of: a) 'on-the-go' plastics, b) packaging (through buying at packaging free, zero waste stores) and; c) non-disposable plastics (**Table 7-7**). Similar to above, there was very little differentiation between responses for disposable versus non-disposable plastics. However, the numbers for those actually taking action are smaller than for those indicating that they would like to take action. The results indicate that whilst the public perceive plastics as a serious environmental issue and aspire to reduce their plastic use, this does not always translate into action. This attitude-behaviour gap is not uncommon, with many authors discussing it in relation to environmental issues (Boulstridge and Carrigan, 2000; Moraes et al., 2012).

Whilst there is an attitude-behaviour gap in relation to reducing plastic use, this does not extend to recycling. In-line with recycling being one of the most mentioned words in the open-ended questions, Australians report being heavily invested in recycling with 80% saying that they recycle 75% or more of metals, plastics, glass and paper (Appendix C, **Figure C3**). Whilst support of recycling is a positive outcome, it could actually be having unintended consequences for the 'reduce' aspect of the waste hierarchy (and it also must be noted that many plastics are downcycled and that all plastics have a limited mechanical recycling lifetime (Rahimi and Garciá, 2017)). Although most respondents (46%) disagree with the statement '*If all plastic is recycled there is no need to reduce my use of it'*, almost one-third (29%) do still subscribe to this notion (Appendix C, **Table C2**).

Taken together, these results reveal support for reducing plastic use, and a negative sentiment towards plastics as a packaging material. However, there is a distinct attitude-behaviour gap, with many people not acting on their expressed desire to reduce plastic use. Intervention to reduce plastic use appears to be widely supported, although more targeted research on the exact measures people are willing to accept is required. There are also relatively widely held misconceptions about recycling and the role of plastic as a packaging material.

Table 7-7: Self-reported activity to reduce personal plastic use

	Never	Rarely	Roughly 30% of the time	Roughly 50% of the time	Roughly 70% of the time	Roughly 90% of the time	Always
Reduce your use of 'on-the-go' plastic (e.g. bring your own take-away coffee cup, bring your own take-away container)	12.4	23.0	14.0	17.0	14.5	11.5	7.6
Reduce your use of packaging (e.g. buy at a 'packaging-free, zero-waste' store; avoid packaged personal care products)	12.7	28.8	17.2	18.1	12.2	6.8	4.2
Reduce your use of non-disposable plastic (e.g. replace plastic containers with glass, buy wooden household goods as opposed to plastic goods)	7.3	22.6	18.0	20.2	15.8	9.5	6.5

7.3.5 *Australians predominantly place responsibility for reducing plastic use on industry and government*

To understand the Australian public's views on who should be responsible for changing the way plastic is used in society, respondents were asked to *'please indicate the level of responsibility of each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic'***.** The results indicate that companies/industry are perceived to hold the highest level of responsibility with 64% of respondents selecting mostly or completely responsible for companies/industry, and 29% selecting moderately responsible (**Figure 7-2**). This is followed by government and then the individual.

Whilst no party is absolved of responsibility, with 54% of respondents disagreeing with the statement *'I have no control over how much disposable plastic I use'* (Appendix C, **Table C1**), the public clearly view industry as being responsible for reducing the use of disposable plastics through thinking about the type of packaging that goes to market, closely followed by government through legislation. This bodes well for initiatives that are being implemented both around the world and in Australia. The move by two major Australian supermarket chains to cease their distribution of free plastic bags at checkouts is one example of industry playing a role in reducing plastic use whilst worldwide, container deposit schemes are examples of industry and government collaboration to improve plastic collection.

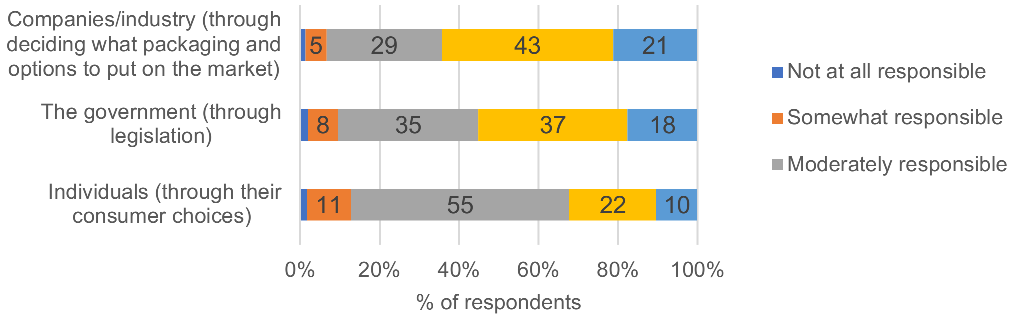

Figure 7-2: Response to *'Please indicate the level of responsibility of each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic'*

7.4 Conclusion

This research, interpreted in the context of the worldwide trends noted in the introduction, indicates that Australians view plastics as a serious environmental issue, and support a reduction in the use of disposable plastics. They place the main responsibility for achieving this on industry.

Plastics in the ocean, and environmental topics relating to plastic waste production and disposal were rated as the most serious environmental issues out of a list of nine topics. This clearly shows that the general public is concerned about the environmental impacts of plastics. On the whole, despite open-ended responses and factor analysis showing that the public values the utility of plastics in relation to food packaging and recognises the convenience that plastics provide, it is apparent that the public view the use of plastics in food packaging negatively.

As a consequence, a large majority (80%) of respondents express a desire to reduce their personal plastic use, view alternative materials as more environmentally friendly packaging options and support measures to reduce the use of disposable items. However, this research also shows that many do not translate these beliefs into consistent action to reduce personal plastic use. Like other environmental challenges, it is clear that an attitude-behaviour gap exists. This is consistent with the finding that whilst almost all respondents attributed at least some responsibility for reducing the use of disposable plastics to the individual, the majority placed the highest responsibility to take action on companies/industry through controlling the type of packaging that goes to market. There is also an expectation that governments will take responsibility through implementing specific legislation.

Further research is now required to develop an action-oriented understanding of the public's attitudes towards plastics. For example, research is required to understand the specific actions and changes the public would be willing to support from industry and government. More research is also required to build on the research that attempts to understand what the main factors are that inhibit people from reducing their personal plastic use despite concern for its environmental impacts. This would involve further exploring what balance of value they place on the convenience that plastic provides versus reduced plastic use for environmental benefit. Price will also play a role and needs to be considered. The use of focus group methodology would be beneficial to allow the complexity of peoples' opinions to be captured and to better understand any concerns or opportunities that emerge. There is also current work being undertaken to understand attitudes towards more specific and novel subsets of plastics such as bioplastics.

CHAPTER 8 Public attitudes towards bioplastics

Chapter Summary

This chapter presents the second of two research activities that relate to the objective of the social attitudes theme: '[To] *Examine public beliefs and attitudes towards plastics and bioplastics in Australia'*. The same methodology was used as presented in **Chapter 7**, but this chapter focuses on bioplastics. The results indicate that the Australian public's knowledge of bioplastics is low, but perception, particularly of biodegradable plastics, is positive. Biodegradable plastics were perceived as better for the environment than 'normal plastics' and 'easily recyclable plastics', although similar to paper and glass.

This chapter is currently under review in the journal Resources, Conservation and Recycling.

Consistent with the requirement for research involving human participants, ethical approval was granted by the University of Queensland's 'Engineering, Architecture and Information Technology, Low and Negligible Risk Ethics Sub-committee' on 27 April 2018, with an amendment approved on 15 May 2018. Copies of the approval letters are included in Appendix F. This process ensures that the research complies with the provisions contained in the *National Statement on Ethical Conduct in Human Research* and complies with the regulations governing experimentation on humans.

8.1 Introduction

Recent research based on a survey of 2,518 members of the general public has identified that the Australian public are highly concerned about the environmental impact of plastics, particularly in relation to ocean plastics (Dilkes-Hoffman et al., 2019c) (**Chapter 7**). Respondents report wanting to reduce their plastic use as a result. In **Figure 8-1**, the basis of this concern is clear. In it, a snapshot is presented to show where all of the plastics produced in 2015 will be by the year 2035 if there is no change to the way we produce, use and dispose of plastics. Over 300 million tonnes of plastic is being produced per year, but very little of it is re-entering the system, instead ending up in landfill or as litter, some of which makes its way to the oceans. To ensure that the plastics system does not look like this in 20 years will require some disruptions, one of which may lie in the small material flows seen at the bottom of **Figure 8-1** - bioplastics.

Although currently a low volume, bioplastics are slowly gaining market share, with the major use being in consumer packaging applications (European Bioplastics, 2018). If they have not already, members of the general public will soon be coming into contact with bioplastics and, against this back-drop of concern for the impacts of conventional plastics, will be needing to make judgements about them. The benefits and the pitfalls of the use of bioplastics can be a complex topic to understand. The aim of this research was to determine public understanding and perceptions of bioplastics. This contributes to understanding the context in which these materials enter the market and the opportunity for their use, as well as identifying the potential for negative outcomes, such as greenwashing.

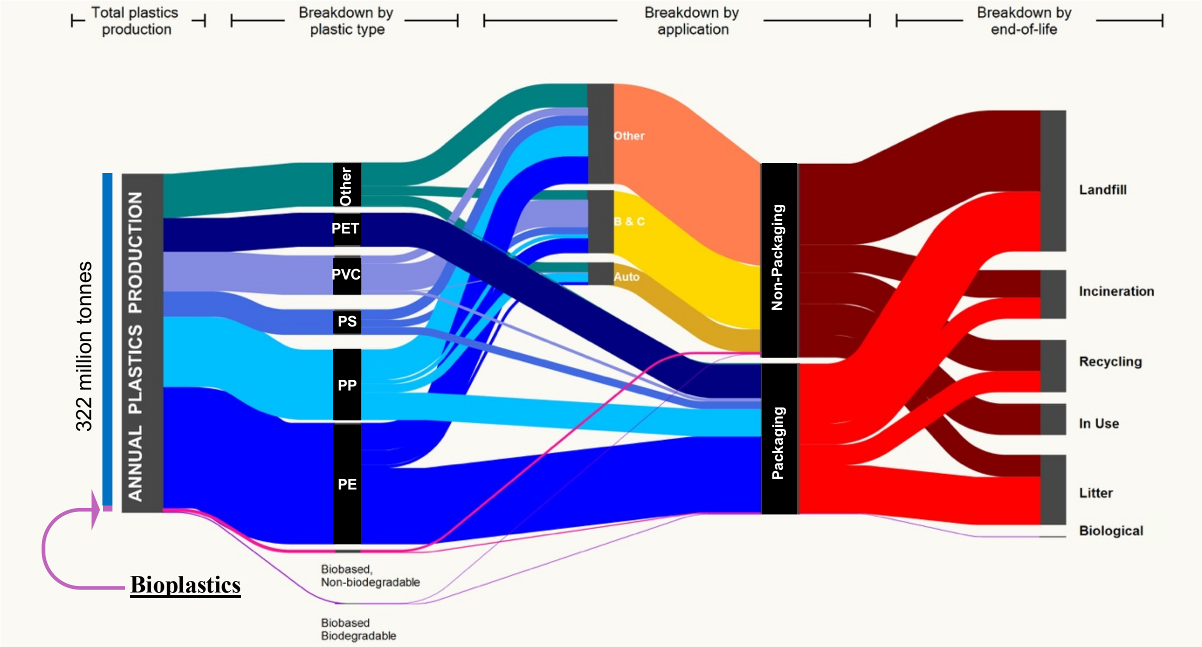

Figure 8-1: Predicted destination of all the plastics produced in 2015 in the year 2035 (plastic fibres not included)

Note to Figure 8-1: Polymer type breakdown based on Plastics Europe (2016) and European Bioplastics (2018). End-of life destinations based on literature estimations (Geyer et al., 2017; Hoornweg and Bhada-Tata, 2012; Jambeck et al., 2015; PlasticsEurope, 2016; World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016). 'Long-term use' is taken to mean longer than a 20-year lifetime. Litter includes all directly littered items as well as mismanagement and illegal dumping of waste. It should be noted that it is hard to draw a definitive distinction between a poorly managed landfill and litter. Data collated by L. S. Dilkes-Hoffman.

Bioplastics are a complex topic to understand because the word 'bioplastic' refers to a broad group of plastics and defines a plastic that is biobased *and/or* biodegradable, with the key point being that the material needs to be one of these, but not necessarily both. For example, the word bioplastic encompasses plastics from biobased but not biodegradable plastics (e.g. bioderived polyethylene (BioPE)); to non-biobased, biodegradable plastics (e.g. polycaprolactone (PCL)); to biobased and biodegradable plastics (e.g. starch, polyhydroxyalkanoates (PHAs), and polylactic acid (PLA)) (Dilkes-Hoffman et al., 2019b; Shen et al., 2009). Within these subsets, not all are biodegradable under the same conditions. For example, PLA should technically be referred to as industrially compostable and not biodegradable as it requires specific higher temperature conditions in compost and degrades through abiotic hydrolysis (Gorrasi and Pantani, 2018). In addition, whilst bioplastics may initially appear to be environmentally superior alternatives to conventional plastics, this isn't necessarily true (Dilkes-Hoffman et al., 2019b; Haider et al., 2018) — their environmental credits can be influenced by their ability to perform their function to an equivalent degree to conventional plastics (Dilkes-Hoffman et al., 2018a) (**Chapter 6**); there needs to be adequate waste management infrastructure for processing them at end-of-life; and recycling often has a better environmental profile when compared to biodegradation, even assuming carbon capture and reuse in the natural production of these biopolymers (Cosate de Andrade et al., 2016; Piemonte, 2011). Reuse is also seen as a more desirable outcome from a circular economy perspective (World Economic Forum; Ellen MacArthur Foundation; McKinsey & Company, 2016), although this does not necessarily discount the longer term need for chemical and mechanical recycling.

The research most similar to the current work was conducted over ten years ago for the Waste, Resource, Action Program (WRAP) in the UK (WRAP, 2007). In that study, consumer perceptions of biopolymers were captured through focus groups and product identification tests. It was found that, overall, (i) knowledge or awareness of biodegradable and compostable plastics was very low but, (ii) many consumers felt positively towards both biodegradable and compostable plastics (although the second point was not examined in depth). However, whilst WRAP's study is a useful backdrop for the current work, it is hard to know how transferrable the results are to today's environment. For example, in setting the context for the WRAP study it was stated that in some parts of the country plastics recycling was still relatively new. This means that both the general attitudes towards plastics and the understanding of associated issues have likely shifted significantly. This said, Sijtsema (2016) recently reported that knowledge of biobased products, including biobased plastics, was still very low in five European countries. Participant questions and responses during focus group sessions in that study highlighted the complexity as well as lack of familiarity with the concept of biobased materials. It can be presumed by extension that the concept of biodegradable plastics likewise remains unfamiliar (Sijtsema et al., 2016).

The larger body of recent work focuses on a consumer's willingness to pay for 'green' materials. Multiple studies have found that consumers are willing to pay more for green/sustainable packaging (Hao et al., 2019; Martinho et al., 2015; Singh and Pandey, 2018). However, it must be noted that the terms 'green' and 'sustainable' were used in a very broad sense, so the result does not necessarily imply anything about the knowledge of the consumers. For example, in Hao (2019) green packaging was defined as '[a] *packaging product that conserves resources or energy, is easy to recycle or reuse, can be incinerated or degraded'.* Drawing on other recent research, it can be assumed that most consumers interpret green/sustainable packaging as referring to its end-of-life attributes, namely that it is 'biodegradable', 'recyclable' or 'reusable' (Herbes et al., 2018; Scott and Vigar-Ellis, 2014). It has been shown that other attributes referring to the raw materials (e.g. 'made from renewable resources', 'made from recycled material') or the production process (e.g. 'energy efficiency in production'), although equally deserving of the title green packaging, are less often associated with the term (Herbes et al., 2018). Thus, the research on willingness to pay for green materials is probably more accurately described as a willingness to pay for 'sustainable' end-of-life options. This assumption is supported by Orset (2017) who more specifically found that there was a willingness to pay a premium for recyclable or biodegradable bioplastic for water bottles. It is also supported by Herbes (2018), who showed that biodegradability is generally perceived as positive, but that being biobased is less so (Herbes et al., 2018). Positive sentiment towards ‘sustainable’ packaging is also demonstrated by Rokka (2008), who found that one-third of their participant consumers favoured environmentally friendly packaging, stating it as a key factor influencing product choice.

Many of these studies extended their research into perceptions of biobased and green packaging materials so as to compare and contrast consumers’ subjective evaluations against the objective outcomes of a life-cycle assessment (LCA). What is clear in all of the studies is that there is a conflict between LCA results and consumers’ perceptions (Boesen et al., 2019; Herbes et al., 2018; Steenis et al., 2017; Van Dam, 1996). Consumers appear to understand ‘green’ packaging to only relate to its end-of-life options, not to its function during production or use, and thus judge packaging sustainability on this basis (Steenis et al., 2017). This was again made obvious recently when it was found that the public in general does not believe that increased food packaging is justified, even if it reduces food wastage (Dilkes-Hoffman et al., 2019c) (**Chapter 7**). Basically, from a consumer’s point of view, environmental friendliness is judged based on the waste left post consumption, not considering the production or use of the material (Van Dam, 1996). In some ways, an LCA does not capture what a consumer is judging the packaging on – i.e. concerns about plastic accumulation, both on land and in the ocean (Dilkes-Hoffman et al., 2019c) – and what a consumer defines as ‘sustainability’ in packaging will differ from the definition used by a sustainability or packaging expert (Van Dam, 1996). Overall, it appears that there is not a clear understanding from many consumers as to what the terms “biobased” or “biodegradable” actually means. Despite this, there is a general positive perception towards such packaging, probably based on a consumer’s focus on end-of-life impacts when evaluating the environmental friendliness of plastics. The current work builds on these previous results but assesses consumers’ knowledge and perception of biobased and biodegradable plastics in a more targeted manner. In particular, perceptions of conventional plastics versus biodegradable plastics versus alternative materials are explicitly probed as well as the understanding of the relationship between biodegradable and biobased materials.

8.2 Methods

The data used in the current research was gathered through the same survey as detailed in an earlier publication by Dilkes-Hoffman et. al. (2019c) (**Chapter 7**) although it draws on a different set of questions than have previously been presented. Readers are referred to the cited work for full details of the survey development and analysis methodology. The questions relevant to the current chapter are outlined in Appendix E.

The survey was developed, refined and prototyped by the authors prior to formal data collection performed by Survey Sampling International, a market research company, between 16th May 2018 and 22nd May 2018. In total, 3028 respondents started the survey with 2529 completing it (the retention rate was 83.5%). Of the completed responses, 2518 were retained for the analysis following data cleaning. The survey can be considered to be representative of the Australian population with all demographics collected closely matching those from Australian census data (results presented in supplementary information of Dilkes-Hoffman et. al. (2019c), Appendix C) (Australian Bureau of Statistics, 2018, 2016).

The section of the survey that focused on bioplastics included a mix of Likert scale, multiple-choice, and open-ended questions. The open-ended question was positioned at the beginning of the section giving respondents the opportunity to present their opinions on bioplastics before being influenced by any information provided.

Statistical analyses were the same as in Dilkes-Hoffman et. al. (2019c). Chi-squared tests for independence were used to determine whether there were significant differences between categorical variables and paired t-tests were used to determine if there were significant differences between means. Open-ended questions were analysed through inductive content analysis (Vaismoradi et al., 2013). This was performed manually by L.S.D.H (main author), with coding consistency being checked by P.L and B.L (co-authors). Eighty-four percent of open-ended responses could be coded, with the remaining 16% being ambiguous or not obviously relevant to the question.

8.3 Results

8.3.1 *Knowledge: Respondents' understanding of what bioplastics are and their characteristics*

At the start of the bioplastics section of the survey, respondents were asked a free word association question so that knowledge of bioplastics could be assessed before any bias was triggered: '*Please record the first two words that come to mind when you hear the word 'bioplastic''*. Knowledge of bioplastics appears to be relatively low, with 30% of the responses being 'Don't know' or 'Nothing' (**Figure 8-2**). When respondents did suggest words, biodegradable (13%) emerged as a more common connection than 'biobased' (5.5%). These are both valid responses, with the word 'bioplastic' encompassing both biodegradable AND biobased plastics, as outlined in the introduction. Many people also associate bioplastics with recyclable/reusable (4.7%) (only mentioned slightly less than biobased) indicating a higher tendency to focus attention on end-of-life outcomes. In general, responses also revealed that respondents relate bioplastics to positive outcomes (8.7%) and better outcomes for the environment (5.8%) (presumably compared to conventional plastics).

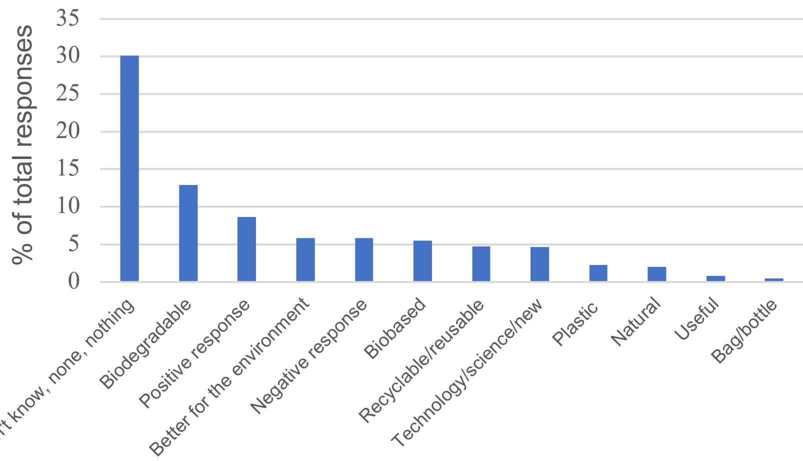

Figure 8-2: Frequency of responses supplied for the word 'bioplastic'

A lack of knowledge about bioplastics was further revealed through more targeted questions included in the survey (**Table 8-1**). The majority of people are unsure whether all bioplastics are biodegradable (70%) and whether all plastics made from plants are biodegradable (69%), and when they do make a decision to agree or disagree, twice as many people choose the incorrect answer as choose the correct answer (judged relative to the definition of a bioplastic as specified in the introduction). Also, respondents are highly uncertain about whether they have come in contact with a bioplastic, indicating that they are not perceived as being widely used. In terms of potential environmental impact, one-third of the respondents indicate that they think biodegradable plastics can have negative environmental impacts, 9% of the respondents don't think biodegradable plastics can have environmental impacts and 58% of respondents again indicate that they are unsure.

Table 8-1: Responses to questions regarding knowledge of bioplastics

	Disagree	Unsure	Agree
All bioplastics are biodegradable	7.7 ✓	70.4	21.9
All plastics made from plants are biodegradable	10.2 ✓	68.8	21.0
I have used an item made from a bioplastic before	10.7	69.1	20.2
Biodegradable plastics can have negative environmental impacts	9.0	58.1	32.9✓

✓ Indicates the correct response

8.3.2 *Perception: Respondents' perceptions of environmental impact and utility*

Respondents were asked to rate three different types of plastic materials (plastic food packaging, durable plastics (containers, furniture and stationery) and biodegradable plastic food packaging) against a variety of traits (**Figure 8-3**). All results were significantly different at the p = 0.05 level apart from two (indicated in **Figure 8-3**; results of the paired t-test can be found in Appendix D, **Table D1**). What emerged was that, on average, biodegradable plastic food packaging is viewed as significantly better for the environment than either of the other materials (although the rating was still closer to neutral than actually being positive). Durable plastic was then considered superior compared to regular plastic food packaging (single-use). Biodegradable plastic food packaging also rated significantly higher on positive words such as good and beneficial. However, these trends changed for the categories of usefulness and convenience. Biodegradable plastic food packaging was seen as being significantly less convenient than durable plastic and regular plastic food packaging, and less useful than durable plastic packaging (although the latter might be related to a single-use assumption more than the material type, as it was not significantly different to regular food packaging). Biodegradable plastic packaging was also perceived as being slightly better compared to conventional plastic packaging with regards to reducing food waste.

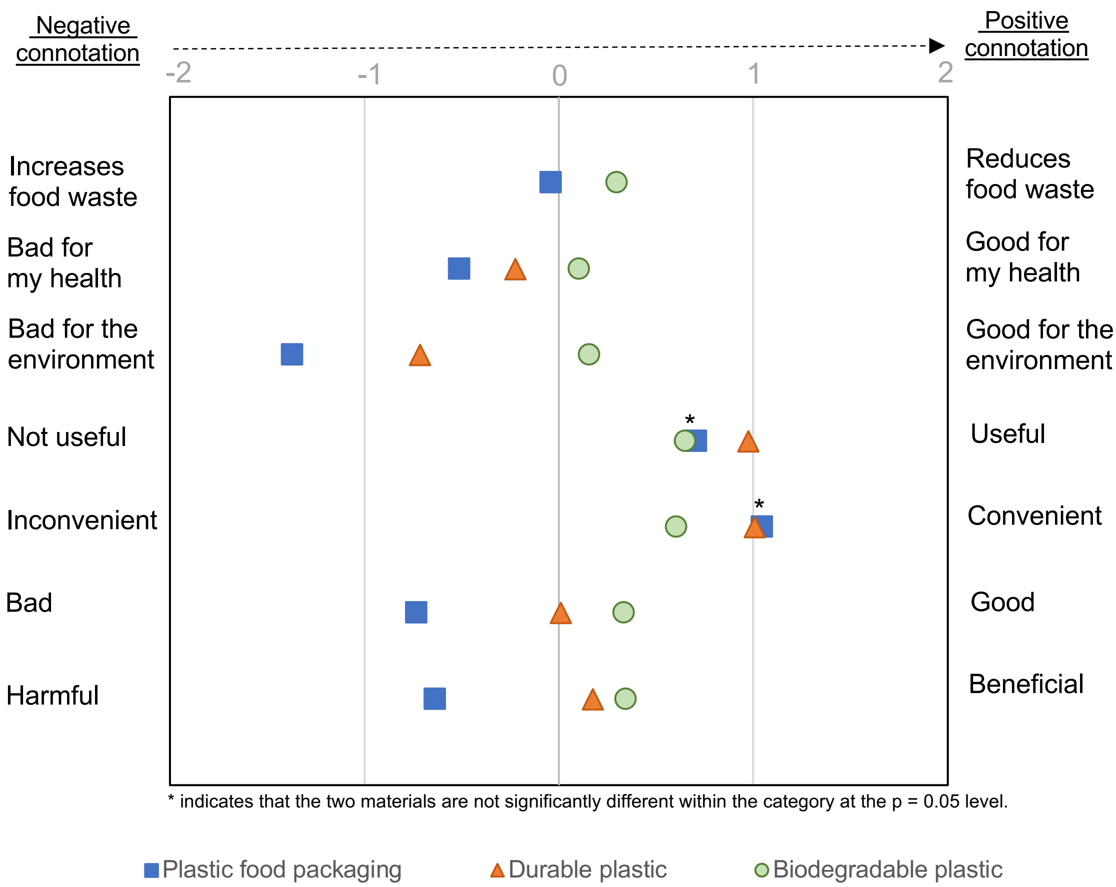

Figure 8-3: Rating of plastic food packaging, durable plastics (containers, furniture and stationery) and biodegradable plastic food packaging for a variety of traits

When asked to compare a variety of alternative materials (biodegradable plastic, biobased plastic, degradable plastic, paper and glass) to conventional plastic food packaging, '*Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? (1 = much worse, 5 = much better)*' all of the alternative materials were perceived as environmentally superior on average (**Table 8-2**), with very few people considering any of the alternative materials to be worse. All of the alternative materials were rated similarly (p = 0.05). Paper was given on average the highest rating compared to conventional plastics, although this was not significantly different to the rating for biodegradable plastics. Biodegradable plastic, biobased plastic and glass were not significantly different to each other. The only consistently lower rated material was degradable plastic. Even then, whilst degradable plastic was given an average rating lower than the other comparison materials, only 7% of people viewed it as worse than conventional plastics and 61% of people viewed it as better.

Table 8-2: Response to the question "Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? (1 = much worse, 5 = much better)"

	% response						
	Much worse 1	**Somewhat worse 2**	**About the same 3**	**Somewhat better 4**	**Much better 5**	**Mean**	**Standard deviation**
***Paper[a]**	1	4	26	33	36	4.0	0.9
Biodegradable plastic[a,b]	1	3	26	46	24	3.9	0.8
***Glass[b]**	2	6	28	33	31	3.8	1.0
Biobased plastic[b]	1	2	29	46	21	3.8	0.8
Degradable plastic	2	5	33	44	17	3.7	0.9

*Results adapted from Dilkes-Hoffman et al. (2019c); [a,b] = Having the same superscript indicates that the mean rating for the items are not significantly different or are negligibly significantly different to each other at the p = 0.05 level. Results of statistical tests are available in Appendix D, **Table D2**.

Respondents were then asked a similar question, but this time asking them to specifically compare biodegradable materials to recyclable materials: 'On a scale from 1 = Much worse to 5 = Much better; *Do you think biodegradable plastics are better for the environment or worse for the environment when compared to easily recyclable plastics?"*. In this case the mean response ($\bar{x}$) = 3.90 (σ = 0.90) is similar to the results above and indicates that even when prompted with the possibility of recycling, many people lean towards seeing biodegradability as an even better outcome. It appears that this positive perception of biodegradability leads to the majority of people (68%) wanting more of their items to be biodegradable (**Figure 8-4**).

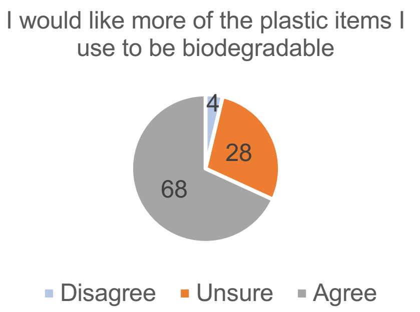

Figure 8-4: Response regarding support of biodegradable plastic items

Any differences in responses to all the questions based on age or gender were negligible (p = 0.01). Differences based on concern for environmental problems were assessed by asking '*In general, are you concerned about environmental problems? (1 = No, not concerned, 4 = Yes, a great deal)*' and were significant but small, with those that were more concerned about environmental problems more likely to agree that they would like more of the plastic items they use to be biodegradable (see Appendix D, **Figure D2**).

8.3.3 *End-of-life management*

If biobased and biodegradable plastics are used on a more regular basis, then their disposal needs to be considered. Most people (62%) indicate that they would dispose of a bioplastic (assuming they were able to identify it) in the regular recycling bin '*Please select how you would currently dispose of a biodegradable plastic material (e.g. a food package or a take away container); response options – recycling bin, regular bin, home compost, don't know, other*' (**Figure 8-5**). This is potentially an emerging trend that needs to be managed. When considering drop-in biobased plastics, such as bioPE, there is no issue as they are chemically identical to petroleum derived counterparts and can be recycled in the same stream. However, biodegradable plastics, which are chemically distinct from plastics currently on the market, represent a new stream of materials that will need to be sorted into different streams at a plastics recovery facility. Given that volumes of these materials are currently low, it is unlikely that they will be recycled, at least in the current waste management system.

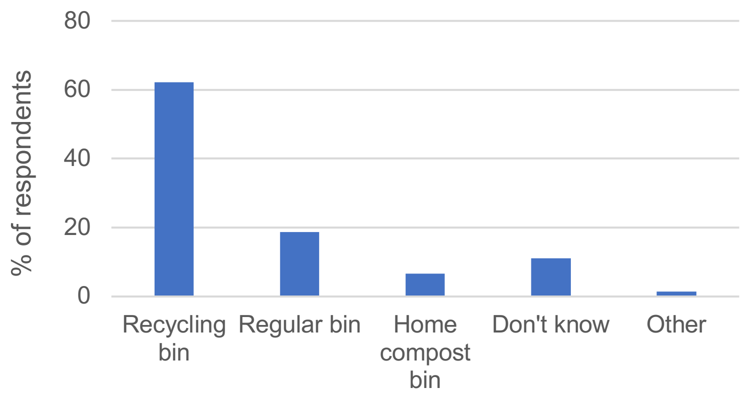

Figure 8-5: Response to the question "Please select how you would currently dispose of a biodegradable plastic material (e.g. a food package or a take away container)"

Regarding disposal, another issue that requires consideration is whether consumers will be more careless with littering if they are told their item is biodegradable. On a positive note, here we see that the majority of people (68%) think that littering still applies even if a plastic material is biodegradable (**Figure 8-6A**). Many people (59%) also still express concern for biodegradable plastics entering the ocean. However, there is also a large group (30%) that are unsure about whether they should be concerned about biodegradable plastics entering the ocean (**Figure 8-6B**). This is reflective of the results presented in **Table 8-1**, which showed that 58% of respondents were unsure about whether biodegradable plastics can have negative environmental impacts. This then led to uncertainty regarding how concerned they should be about biodegradable plastics entering the marine environment, with 41% of respondents that selected 'unsure' about negative environmental impacts selecting 'unsure' for concern about entering the ocean. This is significantly different to the response profiles for respondents that selected 'agree' or 'disagree' for the negative environmental impacts question (see Appendix D, **Figure D3**).

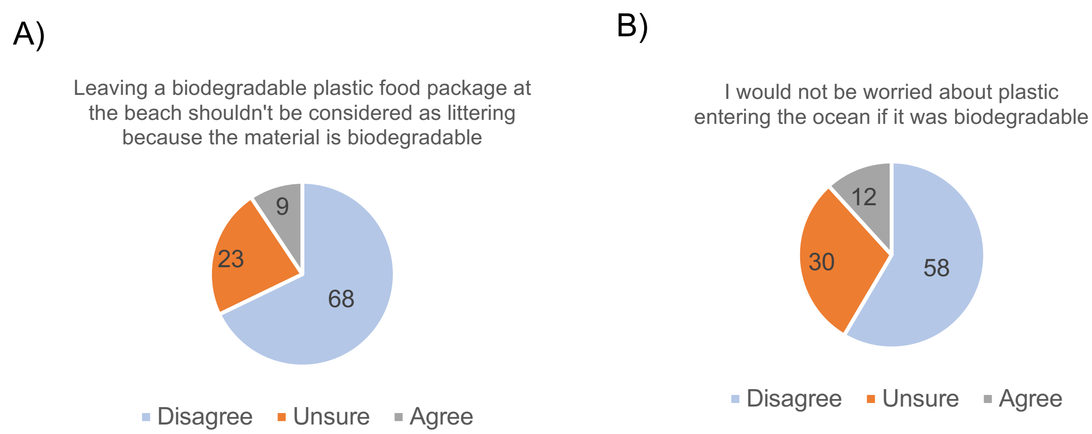

Figure 8-6: Attitudes towards littering of biodegradable plastics or their impact upon entering the environment

8.4 **Discussion and conclusion**

Bioplastics are a class of materials that the Australian public are unfamiliar with. As revealed through open-ended questions as well as targeted knowledge questions, knowledge of what a bioplastic is and the relationship between biodegradable and biobased plastics is poor. This reflects the results found for biodegradable plastics in the UK ten years ago (WRAP, 2007) and the results found for biobased materials more recently in Europe (Sijtsema et al., 2016).

Whilst knowledge of the material characteristics of bioplastics may be low, the public generally hold clear expectations and perceptions of them. What becomes apparent through the open-ended question is that, in general, the public are more likely to relate bioplastics to end-of-life outcomes (such as biodegradable, recyclable, reusable) as opposed to production characteristics (e.g. made from plants). They are also likely to associate bioplastics with positive words and positive environmental outcomes. This focus on end-of-life characteristics when thinking about bioplastics and the tendency to view them positively highlights the connection the public draws between plastics and end-of-life environmental impacts (Dilkes-Hoffman et al., 2019c; Van Dam, 1996) (**Chapter 7**), with bioplastics appearing to be perceived as an alternative.

Biodegradable plastics are shown to be more positively perceived than normal plastic food packaging or durable plastics, specifically with regard to environmental impact. Again, it could be inferred that this is linked more broadly to the public's focus on end-of-life impacts as biodegradable plastics, paper and glass were all rated as better for the environment when compared to normal plastic food packaging. Although biodegradable and biobased plastics were not rated significantly differently in this case, it is unclear whether most participants knew that biobased materials are not necessarily biodegradable. Paper and glass were specifically included in this question as a benchmark for the biodegradable and biobased plastics because they are existing, alternative packaging materials that are not linked to marine pollution. Paper was rated the best on average compared to conventional plastic but was not rated as being significantly different to biodegradable plastics. This potentially indicates that biodegradable plastics are perceived as of similar biodegradability to paper (which one would assume is regarded as a completely biodegradable material). However, this would need to be confirmed by asking the public to directly compare the materials. Biodegradable plastics **were** directly compared to recyclable plastics and significantly, biodegradability was on average perceived as better for the environment than recyclability. Degradable plastics having the lowest average rating indicates that the public may have some level of knowledge that 'degradable' plastic is different to 'biodegradable' plastic. However, on average, they were still rated as better than normal plastics, which is a perception that will need to be addressed if other countries follow suit with the European Union and move against oxo-degradable plastics (European Commission, 2018b).

Whilst perceptions clearly skew positively towards biodegradability in Australia, it should be noted that perceptions do differ to some extent across cultures. Herbes (2018) showed that Germans also consider biodegradability most important in terms of environmental attributes, whilst the French and Americans focus on recyclability. Whilst relative rankings between materials may change, what does appear to be constant across cultures, as well as the current study, is a consumer's strong focus on end-of-life attributes when considering sustainability (Dilkes-Hoffman et al., 2019c; Herbes et al., 2018) (**Chapter 7**).

Positive perception of biodegradable plastics does not appear to extend to utility, with biodegradable plastic packaging not perceived as being as convenient as durable plastics or regular plastic packaging. However, this does not appear to be a dominating factor, as the public still responds that they would like to see more of the plastic items they use to be biodegradable.

This work confirms that the Australian public's knowledge of bioplastics is low, but perception, particularly of biodegradable plastics, is positive. This presents a basis for concern – firstly, that some businesses will engage in greenwashing, making claims about the biodegradable/biobased nature of their packaging and products, in order to capitalise on the positive perception of biobased packaging; secondly, that businesses will switch to biodegradable/biobased plastic materials for their single-use and packaging items, due to their positive sentiment towards bioplastics, but without putting in place appropriate mechanisms for their capture and disposal; thirdly, that in terms of lifetimes in the environment, the expectations of bioplastics do not match their reality. The waste management of biodegradable plastics requires dedicated collection of organic waste streams which are taken to composting or anaerobic digestion facilities that have indicated they are happy to have biodegradable plastics included in the process. Australia, and many other countries, don't currently have the waste management mechanisms to appropriately process biodegradable plastics. There is also still a lot of uncertainty regarding the lifetime that can be expected for biodegradable plastics under uncontrolled conditions (i.e. when they enter the environment) (Dilkes-Hoffman et al., 2019a; Harrison et al., 2018; Kale et al., 2006) (**Chapter 5**). Until lifetimes are better understood, it cannot be assumed that biodegradable plastics will reduce impacts compared to conventional plastics when entering the environment.

In light of these results − highlighting the confusing nature of this topic, public interest in bioplastic materials and potential for undesirable outcomes − governments and local councils are encouraged to play a greater role. There is a need for clear education, standards, and labelling to be coupled with the introduction of bioplastic packaging materials, as well as better understanding and communication of the fates of biodegradable plastics and associated lifetimes. One of the reasons to set clear standards for biobased and biodegradable plastics, and uphold them, is to protect consumers so that companies cannot falsely capitalise on the positive sentiment that exists towards biodegradable and biobased materials. Also, at both local and national levels, appropriate waste management systems need to be developed concurrently with the rise in the availability of biodegradable plastic packaging materials. As it stands, the public are not aware (and probably do not have available to them) appropriate disposal options for biodegradable materials meaning their use is not having the sustainability benefit the public would be expecting.

CHAPTER 9 Summary and future outlook

9.1 Summary and synthesis of results

The aim of this Ph.D. thesis was to explore the role of biobased, biodegradable plastics in a new and sustainable plastics economy. Although this aim was initially approached from a material properties perspective, it soon became apparent that to satisfactorily explore the role of biodegradable plastics, the broader system they are used within would need to be considered. Thus, research based across three different themes, each of which are important drivers of the use of biodegradable plastics (A: material properties, B: environmental impact assessment and C: social attitudes), was undertaken. A summary of the outcomes in relation to the research questions is presented in **Figure 9-1**.

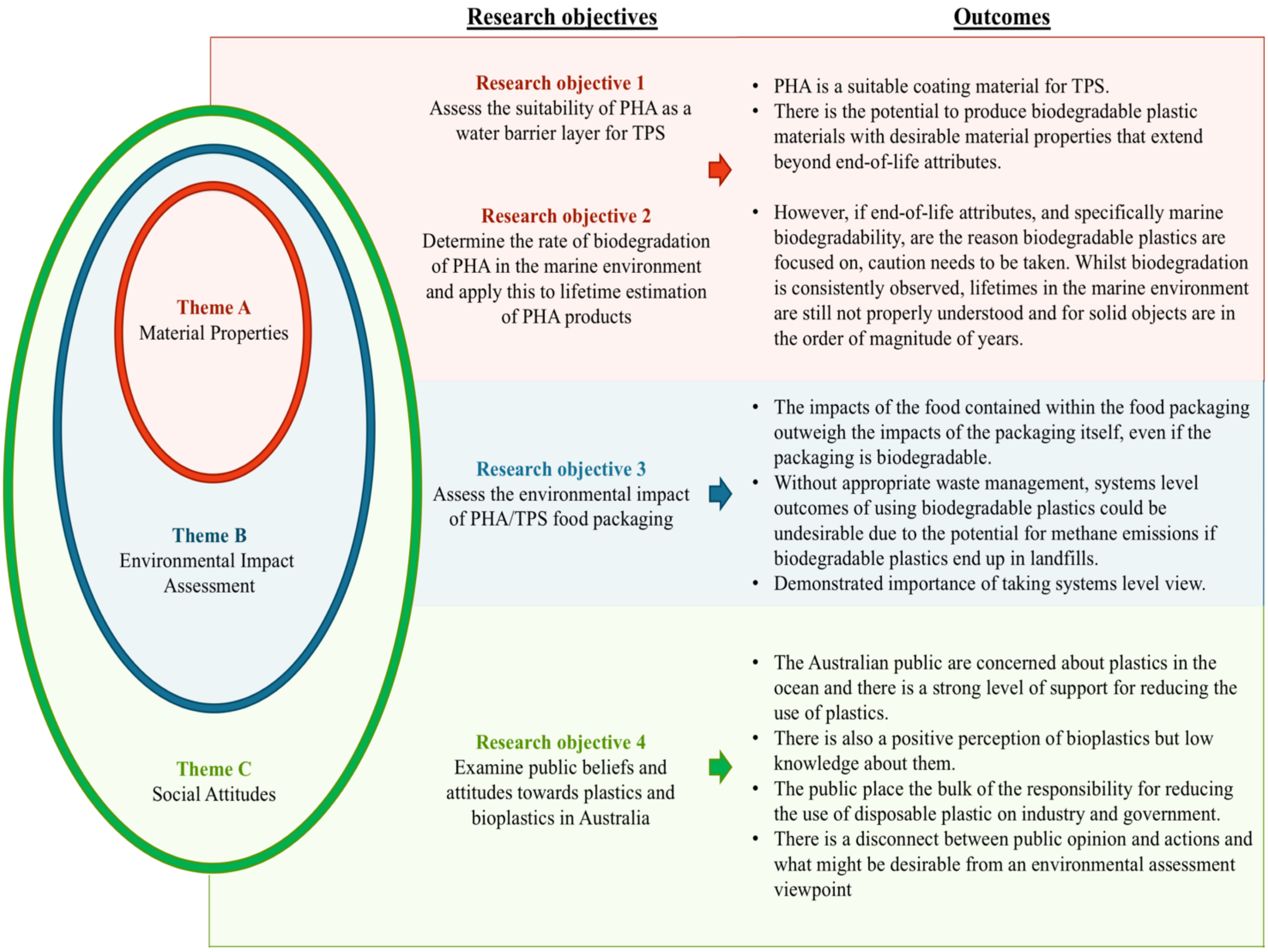

Figure 9-1: Summary of research results

The first research objective within the material properties theme explored the possibility of producing a 100% biodegradable multi-layer film by testing the suitability of PHA as a water barrier layer for TPS. The goal was not to produce a product of commercial quality, or test all possible formulations, but to explore the possibility of making a biodegradable material that could replace a problematic conventional packaging material. This was driven by the idea that unless it is possible to produce biodegradable plastics that offer comparable or enhanced material properties compared to conventional plastics, the role they will play in the plastics system will be limited.

The research showed that PHA is a suitable coating material for TPS, reducing moisture uptake and helping it to maintain its barrier properties (**Chapter 3**). This research also explored and demonstrated the feasibility of using a flow-induced migration technique with the potential to overcome adhesion issues encountered in the first part of the research (**Chapter 4**). Together, these activities demonstrate the possibility of producing biodegradable plastic materials with desirable material properties that extend beyond end-of-life attributes. In fact, in the case of these PHA/TPS multi-layered films, the properties are good enough that they warrant consideration regardless of the focus on trying to produce biodegradable materials. This is an example of biodegradable plastic materials providing interesting opportunities for innovation. With further research it is believed the material properties could be controlled to produce desired outcomes for a variety of applications.

However, as discussed in **Chapter 2**, simply demonstrating the feasibility of producing a biodegradable plastic version of a product does not guarantee improvements from a sustainability perspective. This realisation influenced the trajectory of the thesis, and prompted the research presented under the environmental impact assessment theme (**Chapter 6**). Life-cycle assessment methodology (LCA) was used to assess the likely environmental impact of using a PHA/TPS multi-layered material in a food packaging application. This allowed consideration of the broader system surrounding the material, with food waste being included. Whilst there are trade-offs associated with every change in a system, the LCA clearly showed that if end-of-life options for a food packaging were the only consideration underpinning a decision to switch to a biodegradable material, then sustainability efforts would be misguided. In the case of the food packaging, the impacts of the food contained within the packaging outweigh the impacts of the packaging itself, even if the packaging is biodegradable. This means that the key target for any food packaging material is to reduce food waste. The results also revealed that if waste management systems for biodegradable plastics do not develop at the same rate as the material's implementation, then the systems level outcomes of using biodegradable materials could be undesirable. This is due to the potential for methane emissions from biodegradable plastics if they end up in landfills that don't have effective gas capture systems. Not only is the development of appropriate waste management systems important for ensuring biodegradable plastics don't end up in landfill, it also encourages the appropriate processing of food waste, which biodegradable plastic packaging can facilitate. The overarching result was that a PHA/TPS multi-layer food packaging only delivers positive GHG outcomes if it reduces food wastage or increases the viability of biological food waste processing.

The outcomes of the LCA work were enlightening and again prompted a reassessment of the research trajectory. The results demonstrated the importance of taking a systems level view, whilst simultaneously demonstrating that LCA could not actually deliver the full systems view for plastics. Currently, LCA cannot provide insight into many of the environmental impacts associated with plastics, in particular, accumulation of plastics in the ocean. Also, LCA relies on scenarios to reflect different human behaviour outcomes (e.g. disposal in landfill or compost) but can give no insight into the likelihood of the different behaviours. Consideration of these two points both brought the focus back to the material properties theme (this time targeting marine biodegradation), as well as expanded the scope to the research objectives defined in the social attitudes theme (given the need to understand the likelihood of the behaviours represented in the scenarios).

Returning to the material properties theme and picking up on the idea of needing to be able to produce biodegradable plastics with comparable or enhanced material properties compared to conventional plastics, a clear category where one would think biodegradable plastics could offer enhanced functionality is in reducing marine plastics persistence. However, it became apparent that not only could LCA not provide insight into the potential for biodegradable plastics to address marine persistence, neither could the existing materials properties research − which is why the marine biodegradation of PHA was explored. PHA was selected as it was the biodegradable plastic with seemingly the most promising marine biodegradation profile. Whilst a reasonable number of experimental research articles had been published on its marine biodegradation, no synthesis of the results had ever been performed. This meant there was a lack of clarification as to what 'marine biodegradation of PHA' meant in practicality and what lifetimes could be expected for PHA objects in the marine environment. The meta-study presented in this thesis (**Chapter 5**) showed that whilst lifetimes are certainly much shorter than for conventional plastics, and biodegradation is observed in all cases, they are not short enough to present an immediate solution to the leakage of plastics into the marine environment. PHA objects would likely still persist for years in the marine environment (with exact timeframes dependent on a variety of factors) which is still enough time to pose a threat to the ecosystem. In order for this result to be better understood — and for there to be the potential to control/manipulate the biodegradation rate — the factors that influence the lifetimes of these materials need to be further investigated. There is a need for work that focuses on specific properties of the material, for example, different thicknesses, surface morphology, shapes and porosity of samples, as opposed to more research that simply tracks mass loss over time without testing a specific characteristic. Until such research is conducted, and marine biodegradation is properly understood from a materials property perspective, we will remain unable to have an informed discussion about the benefits and trade-offs of using biodegradable plastics. Also, without the materials level understanding, a tool such as LCA will remain unable to provide a true picture of the systems level impacts of biodegradable plastics, even if methodology improvements allowed it to include plastic accumulation/persistence as an indicator.

The outcomes of this material level investigation reinforced the need to expand the scope of the research to include the social attitudes theme — basically, attitudes towards biodegradable plastics and their market potential need to be understood before time and effort are invested in addressing some of the detailed material properties questions. The research undertaken in the social attitudes theme was guided by the question 'even if time is invested in understanding and modifying the material properties of biodegradable and marine biodegradable materials, with the chosen areas of effort based on rigorous environmental impact assessment, what sort of difference in the market will this actually make and what sort of behaviours can be expected towards them?'. This is in recognition that the **way** plastics are used, and **what** will be used, is influenced by consumer, industry and government decisions. One of the steps in answering this question involves understanding the public's beliefs and attitudes towards plastics and biodegradable plastics. This was explored through a survey of the Australian public. It was shown that the public, in general, consider plastics, and specifically plastics in the ocean, as a serious environmental threat (**Chapter 7**) indicating that there is currently public support for the focus on plastics. However, the stated concern for the issue does not necessarily translate into action to reconsider plastic use. The public place the bulk of the responsibility for reducing the use of disposable plastic on industry and government. Perhaps unsurprisingly, in the context of this current concern surrounding plastics, bioplastics were looked upon favourably (**Chapter 8**), although knowledge about them was low. Biodegradable plastics were perceived as better for the environment than 'normal plastics' and 'easily recyclable plastics', although similar to paper and glass and respondents reported that they would like to see more items made from biodegradable plastic materials. In this aspect there was a disconnect between public opinion and actions and what might be desirable from an environmental assessment viewpoint. This potentially both reflects that decisions were made based on attitudes rather than knowledge about the materials, but also again highlights the current inability of methods such as LCA to accurately capture the impacts of relevance to plastics. It also highlights the disconnect between the material properties consumers may be expecting from biodegradable plastics (e.g. marine biodegradability), and what they can actually deliver.

9.2 Future outlook

As is shown in **Figure 1-1** in the introduction, bioplastics, and in particular, biodegradable plastics, only represent a small volume of the total plastics production. It is thus unreasonable to imagine a near-term scenario where all of the current plastic production is replaced by biodegradable plastics. It is also debatable whether this is necessarily desirable. However, it can be expected that biodegradable plastics **will** play some role in the future plastics system given that: annual production is increasing, there is public support for them, they provide the opportunity for the use of non-fossil feedstocks, there are many niche applications that require biodegradability (particularly in agricultural and related industries), and these plastics can otherwise offer interesting material properties. This justifies the need for continued research in this space.

In defining the scope of future work, there is first an exercise in identifying 'design for degradation' applications - where biodegradation as a disposal option would provide benefit, addressing a problem that does not have a clear alternative solution. Time and energy could then be sensibly invested in developing the material properties and building the broader system that is required for the identified applications.

A good place to start this work would be through updating previous literature. In particular, the research by Shen et al. (2009), looking at the substitution potential of bioplastics for the entire plastics system, would be interesting to revisit. This time, the focus would be better placed to quantify the substitution potential for applications that would benefit from biodegradable plastics, rather than considering the substitution potential for all plastics. The aim would be to understand the realistic scale of opportunity for biodegradable plastics.

The following are examples of applications that could be considered in the analysis:

- High leakage items used in marine environments: There will always be a certain level of leakage of materials to the environment, regardless of the improvement in our collection systems. Whilst marine biodegradable plastics don't solve the issue of leakage, it is conceivable that in certain applications they could reduce the impact.
- Items that are hard to process via conventional waste management systems: Some desired material properties necessitate complex materials that cannot be processed by recycling. Biodegradable plastics could present interesting alternatives e.g. multi-layer materials.

- Items where the cost of collection or separation is high: For certain items where it does not make sense to collect or separate the materials, biodegradable plastics could provide a good alternative e.g. organically soiled items, linings for paper and cardboard, agricultural films.
- Closed systems with a high level of food waste but where it is hard to implement reuse systems e.g. airlines, cafeterias.

Once a general group of opportunities are identified, a set of overarching criteria could be developed, to allow one to test whether, for a specific scenario, there could be benefit from the implementation of biodegradable plastics. These criteria could also be used to more specifically map the potential for biodegradable plastics in different countries – acknowledging that the different social and infrastructure contexts of different regions influence the role biodegradable plastics could play.

This mapping of 'design for degradation' applications would provide guidance not only on market potential but also about which properties could be focused on in materials engineering research. A clear need, as discussed in section 9.1 and **Chapter 5**, which would emerge again, is for more targeted research into understanding the biodegradation – and particularly marine biodegradation – of biodegradable polymers. This knowledge would then allow one to explore what outcomes could be expected from using biodegradable plastics in high-leakage applications (e.g. it would be possible to start quantifying the steady-state volume of plastics in the marine environment that could be expected given leakage rate and biodegradation timeframes). This would underpin informed decision making regarding the use of biodegradable plastics in such applications.

A key part of developing such overarching criteria would also be in identifying and defining the conditions that need to be met to ensure that the introduction of biodegradable plastics provides the desired outcomes. One clear condition is that appropriate waste management infrastructure needs to be simultaneously developed with the introduction of biodegradable plastics. Meeting this condition presents a few opportunities for research. Firstly, timeframes for the biodegradation of biodegradable plastic items need to be understood in the context of other organic materials, such as food waste, so that they can be composted or digested in existing facilities. Despite there being composting and anaerobic digestion standards, these do not currently consider the whole system, and do not necessarily match the timeframes for biodegradable plastics to those expected by the facility operators. As with the research presented in this thesis, this activity would require consideration of both the technical and social aspects. In particular, such research should be undertaken in consultation with organic waste handlers (who may not want to introduce biodegradable plastic inputs), to understand what the technical requirements are for biodegradable plastics to be broadly acceptable in such facilities. The research proposed at the start of this section, aiming to quantifying the realistic substitution potential of biodegradable plastics, would be useful as it would scope the likely volumes of biodegradable plastics waste management systems would need to be able to accommodate.

There is also work in ensuring that the materials reach the appropriate facilities at end-of-life. This requires clear and consistent labelling (perhaps even colour coding), certification schemes, improvements in sorting technology and consumer education. Aspects of this work will need to be driven from a policy perspective, whilst aspects of it would benefit from further research. One focus for further research could be designing and understanding labelling schemes from the user's perspective. Such research could compare and contrast different cultural contexts, and explore questions relating to the type of communication tools that are the most effective for ensuring biodegradable plastics are disposed of appropriately and whether on pack labelling schemes are having the desired results.

In terms of the broader plastics issue, and the role of policy, there is the opportunity for interesting research focused on assessing the outcomes of recent legislation relating to plastics (including single-use bans and recycled content targets which are being introduced around the world). Broadly, it is important to understand what impact different types of policies are having, but also more specifically to understand their impact on the role of biodegradable plastics. The research presented in this Ph.D. thesis, as well as the research topics suggested in this future outlook section, would ultimately feed into informing how biodegradable plastics should be dealt with by policymakers.

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APPENDIX A Supplementary information for Chapter 5

Table A1: Corresponding papers and rate for data included in the soil, compost and AD sections of **Figure 5-2**

Paper	Environment	Rate ($mg.day^{-1}.cm^{-2}$)
Arcos-Hernandez et al. (2012)	Soil	0.02
Wen et al. (2012) (max)	Soil	0.08
Wen et al. (2012) (min)	Soil	0.05
Manna et al. (2000) (min, saline 20 deg C)	Soil	0.01
Manna et al. (2000) (max, saline 40 deg C)	Soil	0.03
Mergaert et al. (1993) (min, sandy soil 15 deg C	Soil	0.03
Mergaert et al. (1993) (max, hardwood soil, 40 deg C	Soil	0.57
Calmon et al. (1999) (PHB)	Soil	0.01
Calmon et al. (1999) (max PHBV)	Soil	0.01
Calmon et al. (1999)(min PHBV)	Soil	0.02
Mousavioun et al. (2012)	soil	0.01
Madbouly et al. (2014)	soil	0.06
Altaee et al. (2016)	Soil	0.03
Boyandin et al. (2012) (PHB, root zone of Larch)	Soil	0.03
Boyandin et al. (2012) (PHB, root zone of Birch)	Soil	0.01
Boyandin et al. (2013) (PHB in Hoa Lac)	Soil	0.03
Boyandin et al. (2013) (PHBV in Dam Bai)	Soil	0.00
Boyandin et al. (2013) (PHB pellet in Hoa Lac)	Soil	0.25
Boyandin et al. (2013) (PHBV in Dam Bai)	Soil	0.00
Boyandin et al. (2013) (PHB pellet in Hoa Lac)	Soil	0.25
Boyandin et al. (2013) (PHBV pellet in Dam Bai)	Soil	0.02
Volova et al. (2017) (PHBV)	Soil	0.06
Volova et al. (2017) (PHB)	Soil	0.04
Barragan et al. (2016)	Soil	0.02
Woolnough et al. (2010)	Soil	0.10
Manna et al. (2000) (compost, 20 deg C)	Compost	0.01
Manna et al. (2000) (compost, 40 deg C)	Compost	0.02
Mergaert et al. (1994) (heap B, PHBV)	Compost	0.03
Mergaert et al. (1994) (heap C, PHBV)	Compost	0.08
Luo et al. (2003)	Compost	0.39
Gutierrez-Wing et al. (2011)	Compost	0.14
Gutierrez-Wing et al. (2011)	Compost	0.74
Bucci et al. (2007)	Compost	0.83
Tabasi et al. (2015)	Compost	0.06
Gilmore et al. 1992	Compost	0.10

Rutkowska et al. (2008)	Compost	0.21
Gutierrez-Wing et al. (2010)	AD	0.55
Gutierrez-Wing et al. (2010)	AD	2.10
Gutierrez-Wing et al. (2010)	AD	0.27
Gutierrez-Wing et al. (2010)	AD	0.90
Abou-Zeid et al. (2001) (10% methane sludge)	AD	0.05
Abou-Zeid et al. (2001) (100% methane sludge)	AD	0.07
Bucci et al. (2007)	AD	0.83
Manna et al. (2000) (sewerage sludge, 20 deg C)	Sewerage sludge (AD)	0.02
Manna et al. (2000) (sewerage sludge, 40 deg C)	Ssewerage sludge (AD)	0.03

Table A2: Corresponding papers, rate and brief description relating to the data points for the marine environment in **Figure 5-2**

Paper	**Rate ($mg.day^{-1}.cm^{-2}$)**	**Key notes** **Material, environment, thickness, measurement type, temp**
Volova et al. (2010) - film	0.02	PHB/PHBV (11%) (both materials found to degrade at the same rate so this point is representative of both); Tropical marine environment (South China Sea, 120 cm depth); 100 µm cast film; weight loss; 27-30.5° C
Volova et al. (2010) - solid	0.11	This point is representative of PHB (although the range includes the range of PHBV (11% as well); Tropical marine environment (South China Sea, 120 cm depth); moulded solid; weight loss; 27-30.5° C
Rutkowska et al. (2008)	0.10	PHBV (12%); 2 m below sea surface in Polish harbour; 115 µm solvent cast film; weight change; 17-20.3 °C.
Thellen et al. (2008) - PHBV dynamic aquarium	0.07	PHBV (12%); sea-water tank open to environment; 287 µm extrusion film; weight loss; 12-22 °C
Thellen et al. (2008) - PHB dynamic aquarium	0.04	PHB; sea-water tank open to environment; 157 µm extrusion film; weight loss; 12-22 °C
Deroine et al. (2015) - natural marine	0.02	PHBV (8%); Lorient harbour France (no depth given); 200 µm extruded and calendared; weight loss; 10.9-19.8 °C
Mergaert et al. (1995) - PHB salt	0.10	PHB; 6 m below surface of sea-water harbour (Belgium); 0.2 cm thick injection moulded dog bone; weight loss; 6-14 °C
Mergaert et al. (1995) - PHBV salt	0.18	PHBV (representative of both 10%/20%); 6 m below surface of sea-water harbour (Belgium); 0.2 cm thick injection moulded dog bone; weight loss; 6-14 °C
Imam et al. (1999) - mangrove interior	0.02	PHBV (12%); 0.5m below surface in mangrove interior; 510 µm extruded film; weight loss; 32-36 °C
Imam et al. (1999) - mangrove edge	0.04	PHBV (12%); 0. 7m below surface in mangrove edge; 510 µm extruded film; weight loss; 32-36 °C
Imam et al. (1999) - reef edge	0.03	PHBV (12%); 1.9 m below surface in reef edge; 510 µm extruded film; weight loss; 25-29 °C
Imam et al. (1999) - deep open water	0.01	PHBV (12%); 1 m from bottom of deep water; 510 µm extruded film; weight loss; 26 °C
Tsuji et al. (2002a) - sea	0.06	PHB; 1 m below surface of sea; 50 µm; weight loss; 19-26 °C
Doi et al. (1992)	0.08	PHBV (0-61%), 1.5 m depth off the coast of Japan, 50-150 µm solvent-cast film, weight loss, 13-26 °C

APPENDIX B Supplementary information for Chapter 6

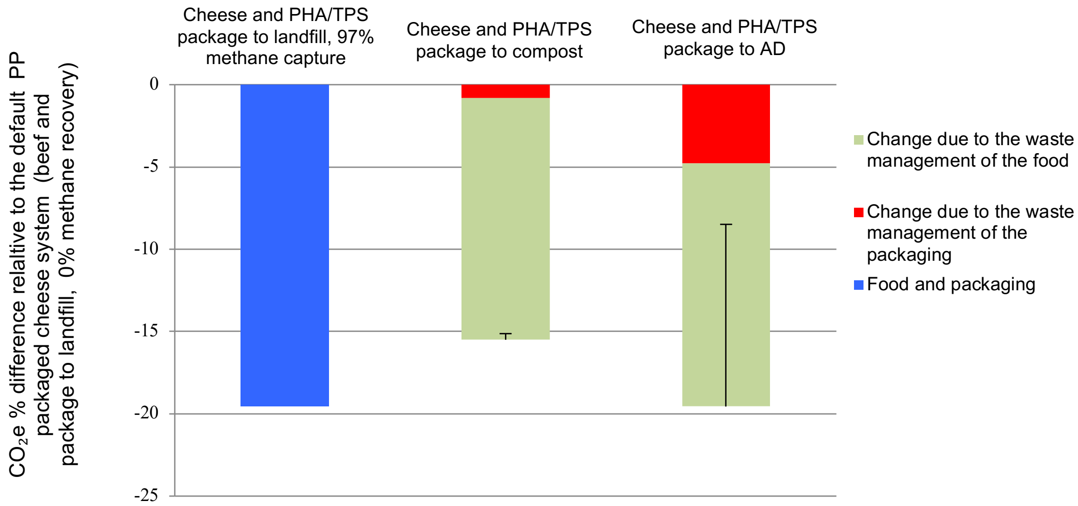


Figure B1 (the cheese equivalent of Figure 6-4): Implications for the GWP100 of a PHA/TPS cheese packaged product under different waste disposal scenarios. The system boundaries are the same as for **Figure 6-3** and the scenarios are benchmarked to '1kg of PP packaged product at the house with disposal in landfill with 0% methane capture'. The range bars indicate the changes associated when material offsets are not included (e.g. Composting: no fertiliser offsets, AD: no electricity or fertiliser offsets)

Notes: Because cheese has a lower GHG footprint of production, changes in EOL lead to a greater % change compared to the system boundary.

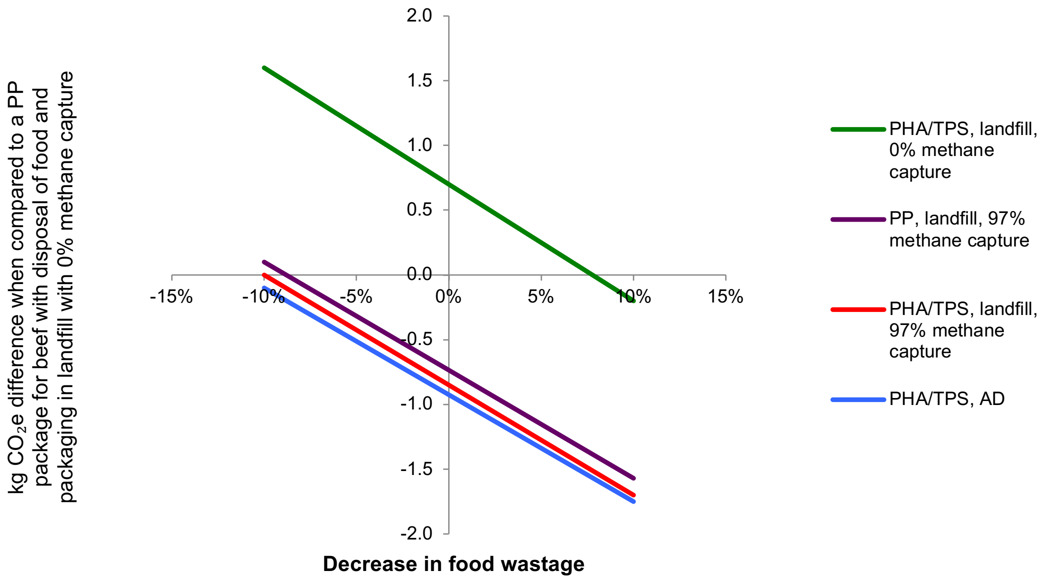


Figure B2 (The GWP20 equivalent of Figure 6-5): kg CO_2e difference (GWP20) for a variety of beef wastage scenarios and waste disposal scenarios. The system boundaries are the same as for **Figure 6-3** and the results are calculated relative to a 'PP beef package disposed of in landfill with 0% methane capture'

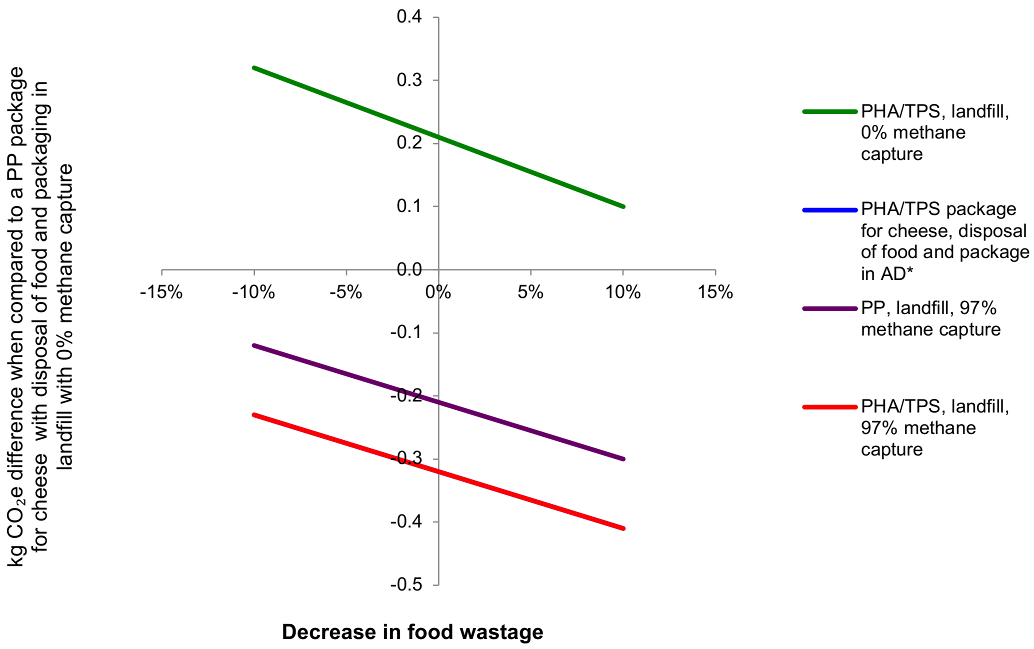


Figure B3 (The cheese equivalent of Figure 6-5): kg CO_2e difference (GWP100) for a variety of cheese wastage scenarios and waste disposal scenarios. The system boundaries are the same as for **Figure 6-3** and the results are calculated relative to a 'PP cheese package disposed of in landfill with 0% methane capture'

* trend line sits underneath the trend line for 'PHA/TPS, landfill, 97% methane capture'.

Note: Because cheese has a lower GHG footprint of production, reducing food wastage does not as effectively negate emissions in landfill. A greater reduction in food wastage is required when compared to beef in order to negate emissions from landfill.

Table B1: Key CO_2 equivalency factors used in the GWP100 and GWP20 scenarios

	Method: IPCC GWP 20a Emissions factor (kg CO_2e/kg)	Method: IPCC GWP 100a Emissions factor (kg CO_2e/kg)
CO_2, fossil	1	1
Methane (biogenic)	82.65	27.75
Methane (non-biogenic)	85	30.5
N_2O	264	265

Table B2: Detailed inventory inputs and GWP100 results for the modeled system

Process	Description	kg CO_2e/kg processed
a) Production of the packaging	**PHA-TPS:** The production of PHA from maize was based on data published by Harding et al. (2007). Thermoplastic starch production was from maize starch, glycerol and water in the ratio 0.6:0.25:0.15 (personal correspondence). Maize and glycerol production processes were from Ecoinvent. A generic extrusion process was used from Ecoinvent with the ratio of PHA:TPS set at 0.2:0.8 (personal correspondence). The waste produced during extrusion was 5% according to Siracusa et al. (2014).	3.35
	PP: Polypropylene granulate production and extrusion process were from Ecoinvent.	3.41
b) Production of the food	**Beef:** The inventory for cattle production was from personal correspondence (Tim Grant, Lifecycles). This dataset was used as an input in an Ecoinvent abatoir process to produce a beef fillet. An economic allocation was used for assigning emissions between co-products. Water use was taken from the report by Mekonnen and Hoekstra (2010).	Beef fillet = 26.0
	Cheese: The inventory for milk production was from a Dairy Australia report (Dairy Australia, 2012). This dataset was used as an input in an Ecoinvent cheese production process to produce the cheese product. An economic allocation was used for assigning emissions between co-products. Water use was taken from the report by Mekonnen and Hoekstra (2010).	Cheese = 9.06
c) Production of the packaged product	A model was created to represent the inputs for production of the packaged product. Energy use was taken as 1.4 kWh/kg packaged product according to Toniolo et al. (2013). Waste was taken as 5% packaging waste and 0% food waste due to an assumption that a biodegradable package can not influence food waste in the packaging process. The packaged product was assumed to be 5% packaging by mass from personal measurements. This aligns well with the values used by Eriksson et al. (2015).	
d) Storage at the supermarket	Food waste at the store was modeled as 4% meat waste or 4.5% dairy waste according to a report by the European Comission (European Commission, 2015). These values align well with other studies (Eriksson et al., 2015; Food and Agriculture Organisation of the United Nations, 2011; Verghese et al., 2013). The energy use for refrigeration was modeled as 0.017 kWh/kg food according to Bernstad Saraiva Schott and Andersson (2015) and was assumed to be the same for both food types.	
e) Storage at the house	Food waste at the house was modeled as 7.5% meat waste or 5% dairy waste according to a report by the European Comission (European Commission, 2015). These values align well with other studies (Eriksson et al., 2015; Food and Agriculture Organisation of the United Nations, 2011; Verghese et al., 2013). The energy use for refrigeration was modeled as 0.56 MJ/kg according to Gruber et al. (2016) and was assumed to be the same for both food types.	
f) Waste processing	**PP:** The Ecoinvent process for disposal of PP to a sanitary landfill was used.	0.133
	PHA-TPS: **Landfill:** The AusLCI process for foodwaste in landfill was used as the model with the following adjustments: Moisture content = 0.15, Degradable organic content (DOC) = 0.59, fraction of the degradable organic content which dissimilates (DOCf) = 0.5 according to Vidal et al.(2007) . Methane capture was set to 0% and sequestered, non-degraded biogenic carbon was accounted for.	**Landfill:** 2.13
	Anaerobic Digestion: The AusLCI process for AD of foodwaste was used as a model with the production of compost and methane. Methane combustion was assumed to offset electricity produced from coal and lignite whilst compost use offset fertiliser application. Methane combustion was adjusted to 380 kg/tonne biopolymer and compost production was adjusted to 150 kg/tonne biopolymer according to Rossi et al. (2015). Sequestered, non-degraded biogenic carbon was accounted for.	**AD:** -1.86
	Composting: The AusLCI process for aerobic composting of foodwaste was used as the model and compost use offset fertiliser application. Compost production was adjusted to 420kg/tonne biopolymer according to Rossi et al. (2015). Sequestered, non-degraded biogenic carbon was accounted for.	**Composting:** -0.0337
	Food: **Landfill:** The AusLCI process for foodwaste in landfill was used as the model. Methane capture was set to 0% and sequestered, non-degraded biogenic carbon was accounted for.	**Landfill:** 1.29
	Anaerobic Digestion: The AusLCI process for AD of foodwaste was used as a model with the production of compost and methane. Methane combustion was assumed to offset electricity produced from coal and lignite whilst compost use offset fertiliser application. Sequestered, non-degraded biogenic carbon was accounted for.	**AD:** -0.119
	Composting: The AusLCI process for aerobic composting of foodwaste was used as the model and compost use offset fertiliser application. Sequestered, non-degraded biogenic carbon was accounted for.	**Composting:** -0.0591

APPENDIX C Supplementary information for Chapter 7

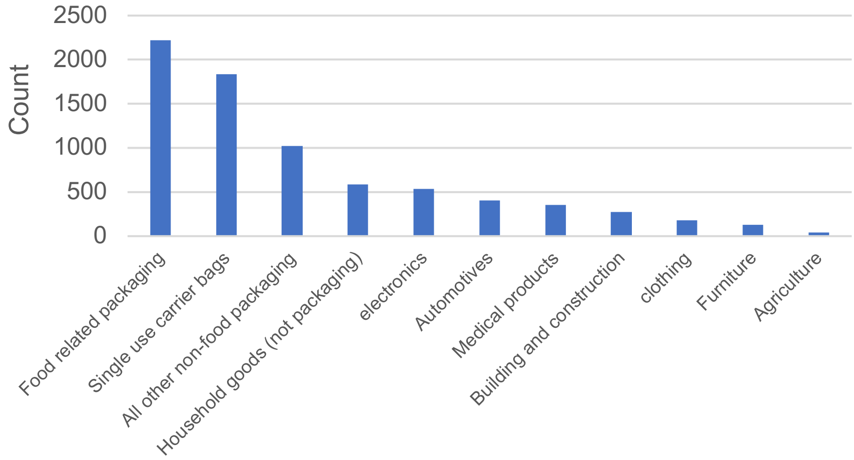


Figure C1: Please choose the three product categories you most immediately associate plastic materials with

Table C1: Desire to reduce personal plastic use

	Strongly disagree	Disagree	Neither agree or disagree	Agree	Strongly agree
I would like to reduce my use of disposable plastics	2.2	2.7	13.9	50	31.3
I would like to reduce my use of plastic used in longer-term applications (buy items made from alternative materials)	1.3	3.5	17.9	50.9	26.4
I have no control over how much disposable plastic I use	14.0	40.4	24.3	16.5	4.7

Table C2: Please indicate your agreement with each of the following statements

	Strongly disagree	Disagree	Neither agree or disagree	Agree	Strongly agree
Measures should be taken to reduce the use of single-use plastic items (e.g. shopping bags, straws...)	4.9	3.4	11.5	32.5	47.7
If plastic food packaging reduces food wastage, that justifies its increased use	8.5	28.9	39.0	19.9	3.7
These days, too many items are made out of plastic	1.2	3.6	15.7	49.4	30.1
If all plastic is recycled, there is no need to reduce my use of it	11.4	34.7	25.0	22.1	6.8

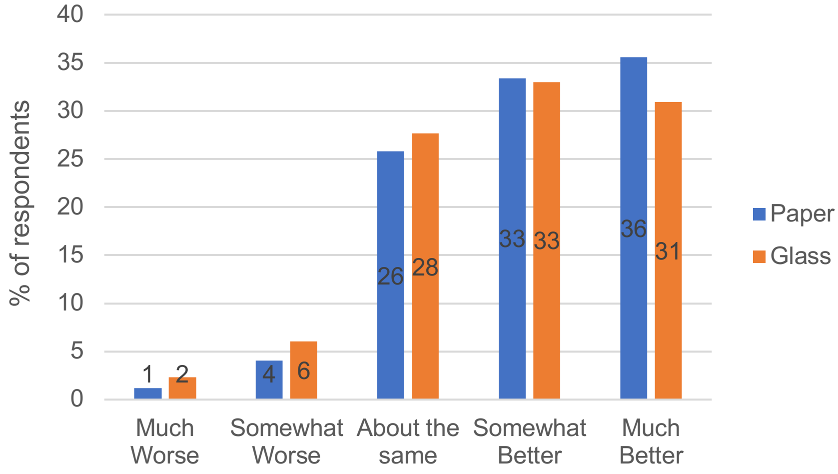


Figure C2: Considering food packaging applications and bags: compared to normal plastics do you think paper and glass are better for the environment or worse for the environment?

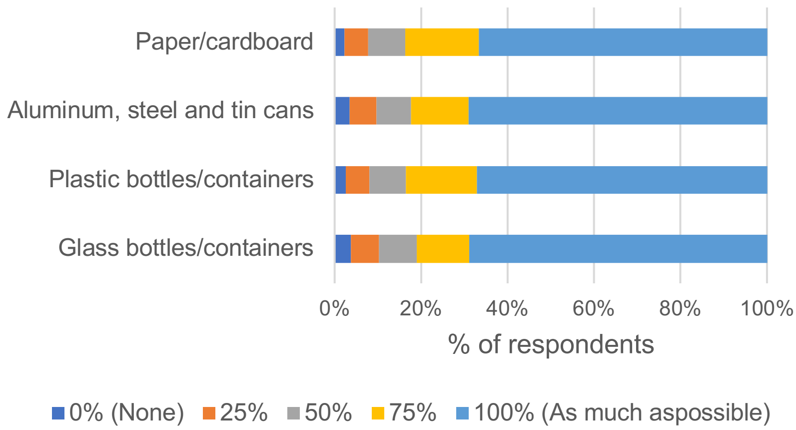


Figure C3: For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year

Table C3: Results of the paired t-test for determining if ranking of the environmental issues was significantly different (results deemed negligible or not significantly different highlighted in grey)

		t	df	Sig. (2-tailed)	Eta-squared
Pair 1	Q6_1_recode Air pollution - Q6_2_recode Water pollution	-14.910	2438	0.000	0.084
Pair 2	Q6_1_recode Air pollution - Q6_3_recode The amount of plastic waste produced	-19.425	2433	0.000	0.134
Pair 3	Q6_1_recode Air pollution - Q6_4_recode Plastic in the ocean	-27.849	2439	0.000	0.241
Pair 4	Q6_1_recode Air pollution - Q6_5_recode The amount of general waste going to landfill	-14.901	2440	0.000	0.083
Pair 5	Q6_1_recode Air pollution - Q6_6_recode Climate change (global warming)	4.657	2415	0.000	0.009
Pair 6	Q6_1_recode Air pollution - Q6_7_recode Natural resource depletion (forest, water, energy)	-6.594	2417	0.000	0.018
Pair 7	Q6_1_recode Air pollution - Q6_8_recode Endangered species and biodiversity	-9.327	2408	0.000	0.035
Pair 8	Q6_1_recode Air pollution - Q6_9_recode Water shortages	0.507	2407	0.612	0.000
Pair 9	Q6_2_recode Water pollution - Q6_3_recode The amount of plastic waste produced	-8.364	2463	0.000	0.028
Pair 10	Q6_2_recode Water pollution - Q6_4_recode Plastic in the ocean	-19.682	2468	0.000	0.136
Pair 11	Q6_2_recode Water pollution - Q6_5_recode The amount of general waste going to landfill	-3.576	2471	0.000	0.005
Pair 12	Q6_2_recode Water pollution - Q6_6_recode Climate change (global warming)	13.691	2437	0.000	0.071
Pair 13	Q6_2_recode Water pollution - Q6_7_recode Natural resource depletion (forest, water, energy)	6.363	2443	0.000	0.016
Pair 14	Q6_2_recode Water pollution - Q6_8_recode Endangered species and biodiversity	2.969	2430	0.003	0.004
Pair 15	Q6_2_recode Water pollution - Q6_9_recode Water shortages	12.229	2427	0.000	0.058
Pair 16	Q6_3_recode The amount of plastic waste produced - Q6_4_recode Plastic in the ocean	-14.934	2472	0.000	0.083
Pair 17	Q6_3_recode The amount of plastic waste produced - Q6_5_recode The amount of general waste going to landfill	5.556	2473	0.000	0.012
Pair 18	Q6_3_recode The amount of plastic waste produced - Q6_6_recode Climate change (global warming)	19.712	2434	0.000	0.138
Pair 19	Q6_3_recode The amount of plastic waste produced - Q6_7_recode Natural resource depletion (forest, water, energy)	14.783	2446	0.000	0.082
Pair 20	Q6_3_recode The amount of plastic waste produced - Q6_8_recode Endangered species and biodiversity	11.411	2431	0.000	0.051
Pair 21	Q6_3_recode The amount of plastic waste produced - Q6_9_recode Water shortages	17.762	2422	0.000	0.115
Pair 22	Q6_4_recode Plastic in the ocean - Q6_5_recode The amount of general waste going to landfill	17.393	2479	0.000	0.109
Pair 23	Q6_4_recode Plastic in the ocean - Q6_6_recode Climate change (global warming)	26.454	2439	0.000	0.223
Pair 24	Q6_4_recode Plastic in the ocean - Q6_7_recode Natural resource depletion (forest, water, energy)	24.522	2449	0.000	0.197
Pair 25	Q6_4_recode Plastic in the ocean - Q6_8_recode Endangered species and biodiversity	22.339	2434	0.000	0.170
Pair 26	Q6_4_recode Plastic in the ocean - Q6_9_recode Water shortages	25.881	2429	0.000	0.216
Pair 27	Q6_5_recode The amount of general waste going to landfill - Q6_6_recode Climate change (global warming)	16.972	2441	0.000	0.106
Pair 28	Q6_5_recode The amount of general waste going to landfill - Q6_7_recode Natural resource depletion (forest, water, energy)	10.371	2452	0.000	0.042
Pair 29	Q6_5_recode The amount of general waste going to landfill - Q6_8_recode Endangered species and biodiversity	6.412	2435	0.000	0.017
Pair 30	Q6_5_recode The amount of general waste going to landfill - Q6_9_recode Water shortages	14.215	2432	0.000	0.077
Pair 31	Q6_6_recode Climate change (global warming) - Q6_7_recode Natural resource depletion (forest, water, energy)	-11.220	2424	0.000	0.049
Pair 32	Q6_6_recode Climate change (global warming) - Q6_8_recode Endangered species and biodiversity	-12.923	2410	0.000	0.065
Pair 33	Q6_6_recode Climate change (global warming) - Q6_9_recode Water shortages	-4.365	2410	0.000	0.008
Pair 34	Q6_7_recode Natural resource depletion (forest, water, energy) - Q6_8_recode Endangered species and biodiversity	-3.961	2424	0.000	0.006

Pair 35	Q6_7_recode Natural resource depletion (forest, water, energy) - Q6_9_recode Water shortages	7.240	2418	0.000	0.021
Pair 36	Q6_8_recode Endangered species and biodiversity - Q6_9_recode Water shortages	10.002	2406	0.000	0.040

Q6_4_recode Plastic in the ocean * Gender Crosstabulation

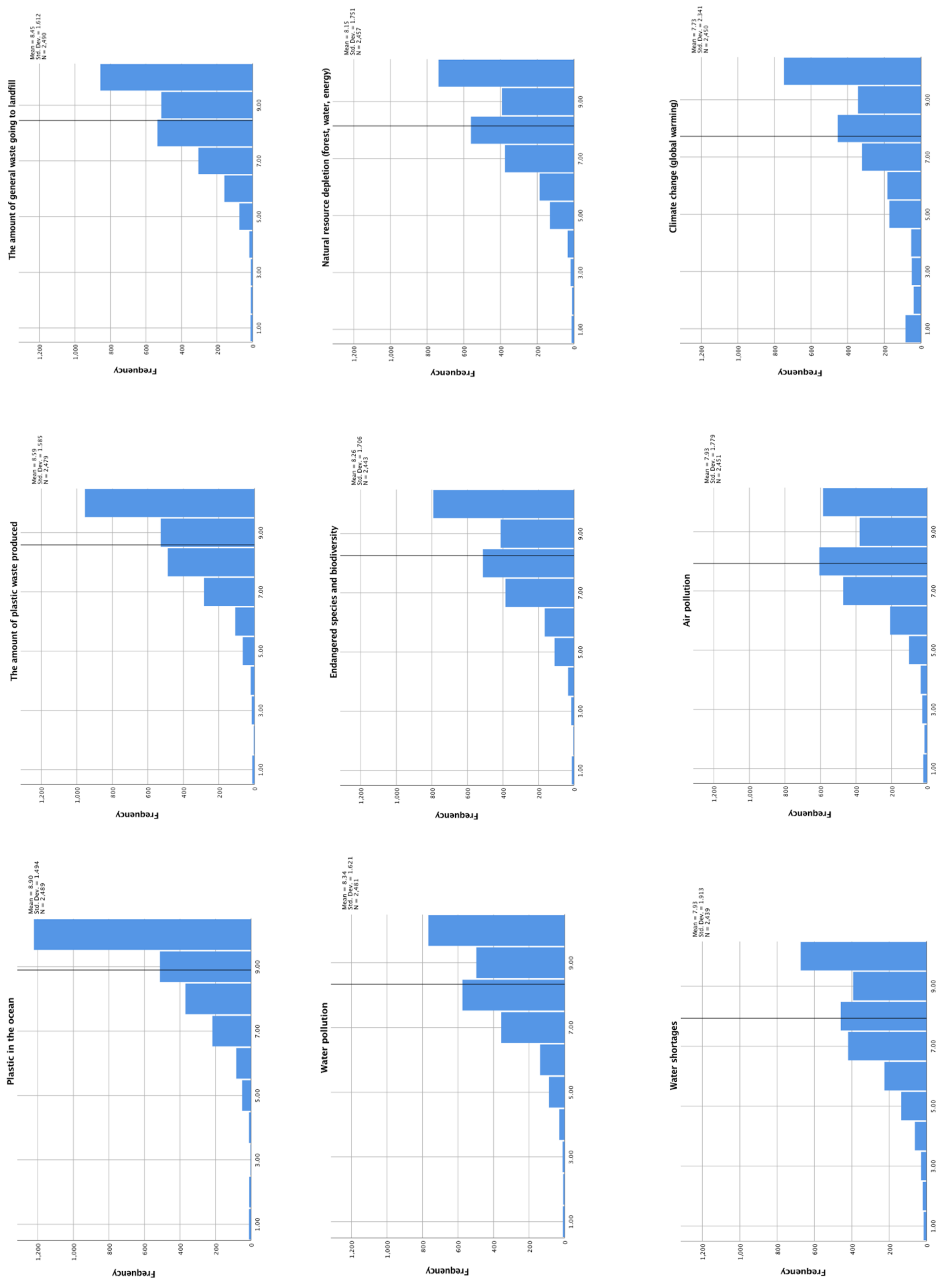
			Gender		
			male	female	Total
Q6_4_recode Plastic in the ocean	5	Count	24	26	50
		% within Q6_4_recode Plastic in the ocean	48.0%	52.0%	100.0%
		% within Gender	2.1%	2.0%	2.0%
	6	Count	45	37	82
		% within Q6_4_recode Plastic in the ocean	54.9%	45.1%	100.0%
		% within Gender	4.0%	2.8%	3.3%
	7	Count	135	82	217
		% within Q6_4_recode Plastic in the ocean	62.2%	37.8%	100.0%
		% within Gender	11.9%	6.2%	8.9%
	8	Count	180	189	369
		% within Q6_4_recode Plastic in the ocean	48.8%	51.2%	100.0%
		% within Gender	15.9%	14.4%	15.1%
	9	Count	235	278	513
		% within Q6_4_recode Plastic in the ocean	45.8%	54.2%	100.0%
		% within Gender	20.7%	21.1%	20.9%
	10	Count	515	704	1219
		% within Q6_4_recode Plastic in the ocean	42.2%	57.8%	100.0%
		% within Gender	45.4%	53.5%	49.8%
Total		Count	1134	1316	2450
		% within Q6_4_recode Plastic in the ocean	46.3%	53.7%	100.0%
		% within Gender	100.0%	100.0%	100.0%

Figure C4: Results for crosstabulation of concern for plastic in the ocean versus gender

Table C4: Results of the factor analysis showing variables included in each factor and their loadings

Factor	Variance explained (%)	Variable (in order of contribution to factor)
Concern regarding plastic waste production and fate	41.0	Q6_4_5cat_recode, Q6_5_5cat_recode, Q6_3_5cat_recode, Q9_2, Q9_4, Q9_3,
Recognised utility of plastic food packaging	14.6	RQ10_3 RQ10_4
Negative perception of plastic food packaging	6.7	RQ10_2 RQ10_1

Figure C5: Raw rankings for each of the environmental issues



Demographic results compared to census statistics

What is your gender?

	Survey (count)	Survey (%)	2016 census data (%)
Male	1180	46.9	49.3
Female	1334	53.0	50.7
Other	4	0.2	

What is your age?

	Survey (count)	Survey (%)	2016 census data (only considering 18+)
18-24	282	11.2	12.4
25-34	497	19.7	18.3
35-44	535	21.2	17.2
45-54	486	19.3	16.9
55-64	448	17.8	15.0
65+	270	10.7	15.7

Census results are approximate given that there is no 18-24 age bracket in the census which means it is not clear what proportion of the population is over 18.

Which state/territory do you live in?

	Survey (count)	Survey (%)	2018 census data (%)
Australian Capital Territory	60	2.4	1.7
New South Wales	741	29.4	31.9
Northern Territory	23	0.9	1.0
Queensland	511	20.3	20.0
South Australia	221	8.8	7.0
Tasmania	98	3.9	2.1
Victoria	614	24.4	25.8
Western Australia	250	9.9	10.4

How best would you describe the area in which you live?

	Survey (count)	Survey (%)	2016 census data (%)
Major City	685	27.2	N/A
Suburban edges of a small town	1100	43.7	
Major town	313	12.4	
Small town	349	13.9	
Remote	56	2.2	
Other	15	0.6	

In which type of dwelling do you live?

	Survey (count)	Survey (%)	2016 census data (%)
House – detached	1790	71.1	72.9
Townhouse (or other semi-detached dwelling)	245	9.7	12.7
Unit/Flat	455	18.1	13.1
Boarding house/College	12	0.5	
Other	16	0.6	

Which of the following best describes your household?

	Survey (count)	Survey (%)	2016 census data (%)
Living full time with dependent child(ren) as parent or carer	851	33.8	N/A
Living part time with dependent child(ren) as parent or carer	68	2.7	
Living with a partner without child(ren)	665	26.4	
Living with other family members (e.g. parents, extended family or non-dependent children)	307	12.2	
Living in shared household with people other than family	152	6.0	
Living in a single person household	432	17.2	
Other (please specify)	43	1.7	

How many dependent children live in your household?

	Survey (count)	Survey (%)	2016 census data (%)
0	56	N/A	N/A
1	375		
2	365		
3	115		
4	33		
5 or more	18		

Which best describes the highest level of education you have achieved or are studying for?

	Survey (count)	Survey (%)	2016 census data (%)
Year 10 or below	264	10.5	10.8
Year 11 or equivalent	105	4.2	4.9
Year 12 or equivalent	442	17.6	15.7
Trade certificate or apprenticeship	675	26.8	24.7
Bachelor or honours degree	721	28.6	Bachelor's degree level and above = 22
Postgraduate degree (e.g. Masters, PhD)	255	10.1	
Other (please specify)	56	2.2	

Which of the following categories best describes your occupational status?

	Survey (count)	Survey (%)	2016 census data (%)
Student	161	6.4	
Employed - full time	1025	40.7	57.5
Employed - part time	472	18.7	30.4
Unemployed - not looking for work	107	4.2	Unemployed = 6.9
Unemployed - looking for work	188	7.5	
Retired	324	12.9	
Unable to work	148	5.9	
Other (please specify)	93	3.7	

How much is your estimated household income before tax?

	Survey (count)	Survey (%)	2016 census data (%)
Less than $25,000	259	10.3	
$25,000 - $49,999	471	18.7	Households with yearly income less than $34,000 = 20.0
$50,000 - $74,999	426	16.9	
$75,000 - $99,999	393	15.6	
$100,000 - $124,999	254	10.1	
$125,000 - $174,999	237	9.4	Households with yearly income more than $150,000 = 16.4
$175,000 - $199,999	79	3.1	
More than $200,000	103	4.1	
Prefer not to say	296	11.8	

In politics, people sometimes talk about the 'left' and the 'right'. On a scale where '0' means left and '10' means right, where would you place yourself on the scale?

	Survey (count)	Survey (%)	2016 census data (%)
0	76	3.0	N/A
1	47	1.9	
2	125	5.0	
3	173	6.9	
4	167	6.6	
5	1154	45.8	
6	223	8.9	
7	224	8.9	
8	170	6.8	
9	60	2.4	
10	99	3.9	

Census references

Australian Bureau of Statistics, 2018. Australian Demographic Statistics [WWW Document]. URL http://www.abs.gov.au/ausstats/abs@.nsf/0/D56C4A3E41586764CA2581A70015893E?Opendocument (accessed 22.11.18).

Australian Bureau of Statistics, 2016. 2016 Census Quick stats [WWW Document]. URL http://quickstats.censusdata.abs.gov.au/census_services/getproduct/census/2016/quickstat/036 (accessed 22.11.18).

APPENDIX D Supplementary information for Chapter 8

Table D1: Results of the paired t-test for determining if rating of plastic food packaging, durable plastic products and biodegradable plastics was significantly different for a variety of traits (results deemed negligible or not significantly different highlighted in grey)

		t	df	Sig. (2-tailed)	Eta-squared
Pair 1	Q10 Please rate plastic food packaging against the following traits: Harmful/Beneficial - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Harmful/Beneficial	-32.504	2517	0.000	0.296
Pair 2	Q10 Please rate plastic food packaging against the following traits: Harmful/Beneficial - Q17A Please rate bioplastic food packaging against the following traits: Harmful/Beneficial	-36.595	2517	0.000	0.347
Pair 3	Q10 Please rate plastic food packaging against the following traits: Bad/Good - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad/Good	-32.009	2517	0.000	0.289
Pair 4	Q10 Please rate plastic food packaging against the following traits: Bad/Good - Q17A Please rate bioplastic food packaging against the following traits: Bad/Good	-41.057	2517	0.000	0.401
Pair 5	Q10 Please rate plastic food packaging against the following traits: Inconvenient/Convenient - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Inconvenient/Convenient	1.891	2517	0.059	0.001
Pair 6	Q10 Please rate plastic food packaging against the following traits: Inconvenient/Convenient - Q17A Please rate bioplastic food packaging against the following traits: Inconvenient/Convenient	18.401	2517	0.000	0.119
Pair 7	Q10 Please rate plastic food packaging against the following traits: Not useful/Useful - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Not useful/Useful	-12.085	2517	0.000	0.055
Pair 8	Q10 Please rate plastic food packaging against the following traits: Not useful/Useful - Q17A Please rate bioplastic food packaging against the following traits: Not useful/Useful	2.284	2517	0.022	0.002
Pair 9	Q10 Please rate plastic food packaging against the following traits: Reduces food waste/Increases food waste - Q17A Please rate bioplastic food packaging against the following traits: Reduces food waste/Increases food waste	12.250	2517	0.000	0.056
Pair 10	Q10 Please rate plastic food packaging against the following traits: Bad for the environment/Good for the environment - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad for the environment/Good for the environment	-30.354	2517	0.000	0.268
Pair 11	Q10 Please rate plastic food packaging against the following traits: Bad for the environment/Good for the environment - Q17A Please rate bioplastic food packaging against the following traits: Bad for the environment/Good for the environment	-58.414	2517	0.000	0.576
Pair 12	Q10 Please rate plastic food packaging against the following traits: Bad for my health/Good for my health - Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad for my health/Good for my health	-16.792	2517	0.000	0.101
Pair 13	Q10 Please rate plastic food packaging against the following traits: Bad for my health/Good for my health - Q17A Please rate bioplastic food packaging against the following traits: Bad for my health/Good for my health	-27.425	2517	0.000	0.230
Pair 14	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Harmful/Beneficial - Q17A Please rate bioplastic food packaging against the following traits: Harmful/Beneficial	-6.362	2517	0.000	0.016
Pair 15	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad/Good - Q17A Please rate bioplastic food packaging against the following traits: Bad/Good	-12.598	2517	0.000	0.059
Pair 16	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Inconvenient/Convenient - Q17A Please rate bioplastic food packaging against the following traits: Inconvenient/Convenient	17.992	2517	0.000	0.114
Pair 17	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Not useful/Useful - Q17A Please rate bioplastic food packaging against the following traits: Not useful/Useful	14.391	2517	0.000	0.076

Pair 18	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad for the environment/Good for the environment - Q17A Please rate bioplastic food packaging against the following traits: Bad for the environment/Good for the environment	-32.222	2517	0.000	0.292
Pair 19	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: Bad for my health/Good for my health - Q17A Please rate bioplastic food packaging against the following traits: Bad for my health/Good for my health	-15.144	2517	0.000	0.084

Table D2: Results of the paired t-test for determining if the rating of the environmental performance of alternative packaging materials compared to plastic were significantly different to each other (results deemed negligible or not significantly different highlighted in grey)

		t	df	Sig. (2-tailed)	Eta-squared
Pair 1	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biodegradable plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biobased plastic	1.293	2517	0.196	0.001
Pair 2	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biodegradable plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Paper	-4.167	2517	0.000	0.007
Pair 3	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biodegradable plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Degradable plastic	10.672	2517	0.000	0.043
Pair 4	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biodegradable plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Glass	2.062	2517	0.039	0.002
Pair 5	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biobased plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Paper	-5.300	2517	0.000	0.011
Pair 6	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biobased plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Degradable plastic	9.635	2517	0.000	0.036
Pair 7	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Biobased plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Glass	1.276	2517	0.202	0.001
Pair 8	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Paper - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Degradable plastic	13.835	2517	0.000	0.071
Pair 9	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Paper	6.715	2517	0.000	0.018

	- Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Glass				
Pair 10	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Degradable plastic - Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? Glass	-6.584	2517	0.000	0.017

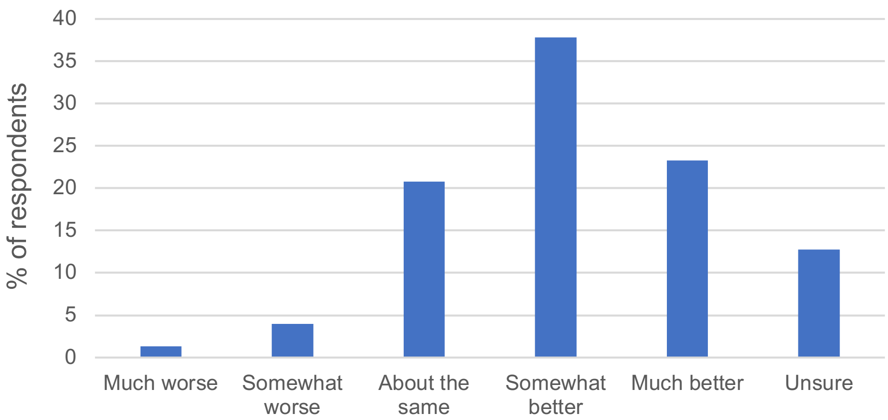


Figure D1: On a scale from 1 = Much worse to 5 = Much better; Do you think biodegradable plastics are better for the environment or worse for the environment when compared to easily recyclable plastics?

Crosstab

			Q33 In general, are you concerned about environmental problems? No, not concerned	Yes, a little	Yes, a fair amount	Yes, a great deal	Total
Q21 Please indicate whether you agree or disagree with the following statements: I would like more of the plastic items I use to be biodegradable	Disagree	Count	5	30	25	34	94
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would like more of the plastic items I use to be biodegradable	5.3%	31.9%	26.6%	36.2%	100.0%
		% within Q33 In general, are you concerned about environmental problems?	5.6%	4.0%	2.6%	4.8%	3.7%
		% of Total	0.2%	1.2%	1.0%	1.4%	3.7%
	Unsure	Count	49	294	233	132	708
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would like more of the plastic items I use to be biodegradable	6.9%	41.5%	32.9%	18.6%	100.0%
		% within Q33 In general, are you concerned about environmental problems?	54.4%	39.7%	24.0%	18.5%	28.1%
		% of Total	1.9%	11.7%	9.3%	5.2%	28.1%
	Agree	Count	36	417	714	549	1716
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would like more of the plastic items I use to be biodegradable	2.1%	24.3%	41.6%	32.0%	100.0%
		% within Q33 In general, are you concerned about environmental problems?	40.0%	56.3%	73.5%	76.8%	68.1%
		% of Total	1.4%	16.6%	28.4%	21.8%	68.1%
Total		Count	90	741	972	715	2518
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would like more of the plastic items I use to be biodegradable	3.6%	29.4%	38.6%	28.4%	100.0%
		% within Q33 In general, are you concerned about environmental problems?	100.0%	100.0%	100.0%	100.0%	100.0%
		% of Total	3.6%	29.4%	38.6%	28.4%	100.0%

Chi-Square Tests

	Value	df	Asymptotic Significance (2-sided)
Pearson Chi-Square	131.190[a]	6	.000
Likelihood Ratio	128.538	6	.000
Linear-by-Linear Association	72.184	1	.000
N of Valid Cases	2518		

a. 1 cells (8.3%) have expected count less than 5. The minimum expected count is 3.36.

Symmetric Measures

		Value	Approximate Significance
Nominal by Nominal	Phi	.228	.000
	Cramer's V	.161	.000
N of Valid Cases		2518	

Figure D2: Results for crosstabulation of concern about environmental problems versus desire for greater use of biodegradable plastics.

Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable * Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts Crosstabulation

			Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts			
			Disagree	Unsure	Agree	Total
Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable	Disagree	Count	136	724	613	1473
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable	9.2%	49.2%	41.6%	100.0%
		% within Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts	60.2%	49.5%	73.9%	58.5%
		% of Total	5.4%	28.8%	24.3%	58.5%
	Unsure	Count	47	609	93	749
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable	6.3%	81.3%	12.4%	100.0%
		% within Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts	20.8%	41.6%	11.2%	29.7%
		% of Total	1.9%	24.2%	3.7%	29.7%
	Agree	Count	43	130	123	296
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable	14.5%	43.9%	41.6%	100.0%
		% within Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts	19.0%	8.9%	14.8%	11.8%
		% of Total	1.7%	5.2%	4.9%	11.8%
Total		Count	226	1463	829	2518
		% within Q21 Please indicate whether you agree or disagree with the following statements: I would not be worried about plastic entering the ocean if it was biodegradable	9.0%	58.1%	32.9%	100.0%
		% within Q21 Please indicate whether you agree or disagree with the following statements: Biodegradable plastics can have negative environmental impacts	100.0%	100.0%	100.0%	100.0%
		% of Total	9.0%	58.1%	32.9%	100.0%

Chi-Square Tests

	Value	df	Asymptotic Significance (2-sided)
Pearson Chi-Square	252.514[a]	4	.000
Likelihood Ratio	272.942	4	.000
Linear-by-Linear Association	30.064	1	.000
N of Valid Cases	2518		

a. 0 cells (0.0%) have expected count less than 5. The minimum expected count is 26.57.

Symmetric Measures

		Value	Approximate Significance
Nominal by Nominal	Phi	.317	.000
	Cramer's V	.224	.000
N of Valid Cases		2518	

Figure D3: Results for crosstabulation of knowledge about the environmental impact of biodegradable plastics versus concern about biodegradable plastics entering the ocean.

APPENDIX E Full copy of the survey questions

Original question numbering has been retained. All original questions have been included as well as any recoding that was performed. Recoded questions are highlighted in red and the recoding formula is provided. Questions used in **Chapter 7** are highlighted in green, questions used in **Chapter 8** are highlighted in blue.

All 'reverse_recodes' were performed to put data into the format:

Code	1	2	3	4	5
Plastic	Like plastics/think positively about plastics (or don't think they are an issue)				Hate plastics/think negatively about plastics (think they are a major issue)
Environment	Don't care about the environment				Care a lot about the environment

Recoding in this format does not change the meaning of the data or artificially impose any relationship between thinking negatively about plastics and having strong concern for the environment. Recoding was performed as it was believed that most people who felt strongly about the environment would be more likely to have negative feelings towards plastics, so having them coded in the same direction would make interpretation of the results easier.

Q2

Please record the first two words/phrases that come to mind when you hear the word 'plastic'

Q2_1	Please record the first two words/phrases that come to mind when you hear the word 'plastic'. 1
Q2_2	Please record the first two words/phrases that come to mind when you hear the word 'plastic'. 2

Q3

Please record the first word/phrase that comes to mind when you think about the positive impacts of plastic

Q3_1	Please record the first word/phrase that comes to mind when you think about the positive impacts of plastic. Response 1 - required
Q3_2	Please record the first word/phrase that comes to mind when you think about the positive impacts of plastic. Response 2 - optional

Q4

Please record the first word/phrase that comes to mind when you think about the negative impacts of plastic

Q4_1	Please record the first word/phrase that comes to mind when you think about the negative impacts of plastic. Response 1 - required
Q4_2	Please record the first word/phrase that comes to mind when you think about the negative impacts of plastic. Response 2 - optional

Q5

Please choose the three product categories you most immediately associate plastic materials with

Coding		0	1
		Not selected	Selected
Q5_1	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Automotives**		
Q5_2	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Agriculture**		
Q5_3	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Building and construction products**		
Q5_4	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Clothing**		
Q5_5	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Electronics**		

Q5_6	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Food related packaging**		
Q5_7	Q5 Please choose the three product categories you most immediately associate plastic materials with. **All other non-food packaging**		
Q5_8	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Furniture**		
Q5_9	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Household goods (not packaging)**		
Q5_10	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Medical products**		
Q5_11	Q5 Please choose the three product categories you most immediately associate plastic materials with. **Single use carrier bags**		

Q6

You will now be presented with nine different environmental issues, please indicate how serious you think each of the following environmental issues are

Coding		1 , 2, 3, 4, 5, 6, 7, 8, 9, 10	99
		1 – not serious, 2, 3, 4, 5, 6, 7, 8, 9, 10- extremely serious	Don't know
Q6_1	Air pollution		
Q6_2	Water pollution		
Q6_3	The amount of plastic waste produced		
Q6_4	Plastic in the ocean		
Q6_5	The amount of general waste going to landfill		
Q6_6	Climate change (global warming)		
Q6_7	Natural resource depletion (forest, water, energy)		
Q6_8	Endangered species and biodiversity		
Q6_9	Water shortages		

Q6_5cat_recode

You will now be presented with nine different environmental issues, please indicate how serious you think each of the following environmental issues are

Coding		1	2	3	4	5
Recode		IF Q6 = 1 OR 2	IF Q6 = 3 OR 4	IF Q6 = 5 OR 6	IF Q6 = 7 OR 8	IF Q6 = 9 OR 10
		Not serious	2	3	4	Extremely serious
Q6_1	Air pollution					
Q6_2	Water pollution					
Q6_3	The amount of plastic waste produced					
Q6_4	Plastic in the ocean					
Q6_5	The amount of general waste going to landfill					
Q6_6	Climate change (global warming)					
Q6_7	Natural resource depletion (forest, water, energy)					
Q6_8	Endangered species and biodiversity					
Q6_9	Water shortages					

Q7

In general, would you say that the overall impact from the use of plastic is negative or positive?

Coding	1	2	3	4	5
	1 - Negative	2	3	4	5 - Positive
In general, would you say that the overall impact from the use of plastic is negative or positive?					

Q8

Please indicate how often you think about the following:

Coding		1	2	3	4	5
		Never	Rarely	Occasionally	Very Frequently	Everyday
Q8_1	Q8 Please indicate how often you think about the following: **Marine plastic pollution**					
Q8_2	Q8 Please indicate how often you think about the following: **The impact of plastic on your health**					
Q8_3	Q8 Please indicate how often you think about the following: **How much waste you produce**					

Q9

Please indicate your level of concern for each of the following:

Coding		1	2	3	4	5
		1 - Not at all concerned	2	3 - Somewhat concerned	4	5 - Extremely concerned
Q9_1	Q9 Please indicate your level of concern for each of the following: **Plastic pollution on land**					
Q9_2	Q9 Please indicate your level of concern for each of the following: **Plastic pollution in the oceans**					
Q9_3	Q9 Please indicate your level of concern for each of the following: **The amount of plastic waste produced daily in Australia**					
Q9_4	Q9 Please indicate your level of concern for each of the following: **Plastic being disposed of in landfill**					
Q9_5	Q9 Please indicate your level of concern for each of the following: **The impact of plastic on your health**					

Q10, Q10

Please rate plastic food packaging against the following traits:

Coding		1	2	3	4	5
		2	1	0	1	2
Q10_1	Q10 Please rate plastic food packaging against the following traits: **Harmful/Beneficial**					
Q10_2	Q10 Please rate plastic food packaging against the following traits: **Bad/Good**					
Q10_3	Q10 Please rate plastic food packaging against the following traits: **Inconvenient/Convenient**					
Q10_4	Q10 Please rate plastic food packaging against the following traits: **Not useful/Useful**					
Q10_5	Q10 Please rate plastic food packaging against the following traits: **Reduces food waste/Increases food waste**					
Q10_6	Q10 Please rate plastic food packaging against the following traits: **Makes my life easier/Makes my life harder**					
Q10_7	Q10 Please rate plastic food packaging against the following traits: **Reduces food hygiene/Increases food hygiene**					
Q10_8	Q10 Please rate plastic food packaging against the following traits: **Bad for the environment/Good for the environment**					
Q10_9	Q10 Please rate plastic food packaging against the following traits: **Bad for my health/Good for my health**					

RQ10_reverse_recode

Please rate plastic food packaging against the following traits:

Coding		1	2	3	4	5
Recode formula		IF Q10 = 5	IF Q10 = 4	If Q10 = 3	If Q10 = 2	IF Q10 = 1
		2	1	0	1	2
Q10_1	Q10 Please rate plastic food packaging against the following traits: **Beneficial/Harmful**					
Q10_2	Q10 Please rate plastic food packaging against the following traits: **Good/ Bad**					
Q10_3	Q10 Please rate plastic food packaging against the following traits: **Convenient/ Inconvenient**					
Q10_4	Q10 Please rate plastic food packaging against the following traits: **Useful/ Not useful**					
Q10_5	Q10 Please rate plastic food packaging against the following traits: **Reduces food waste/Increases food waste**					
Q10_6	Q10 Please rate plastic food packaging against the following traits: **Makes my life easier/Makes my life harder**					

Q10_7	Q10 Please rate plastic food packaging against the following traits: **Increases food hygiene/ Reduces food hygiene/**					
Q10_8	Q10 Please rate plastic food packaging against the following traits: **Good for the environment/ Bad for the environment/**					
Q10_9	Q10 Please rate plastic food packaging against the following traits: **Good for my health/ Bad for my health**					

Q11

Please indicate how often do you do the following:

		1	2	3	4	5	6	7
		Never	Rarely	Roughly 30% of the time	Roughly 50% of the time	Roughly 70% of the time	Roughly 90% of the time	Always
Q11_1	Q11 Please indicate how often do you do the following: **Reduce your use of 'on-the-go' plastic (e.g. bring your own take-away coffee cup, bring your own take-away container)**							
Q11_2	Q11 Please indicate how often do you do the following: **Reduce your use of packaging (e.g. buy at a 'packaging-free, zero-waste' store; avoid packaged personal care products)**							
Q11_3	Q11 Please indicate how often do you do the following: **Reduce your use of non-disposable plastic (e.g. replace plastic containers with glass, buy wooden household goods as opposed to plastic goods)**							

Q11_recode

Please indicate how often do you do the following:

Coding		1	2	3	4	5
Recode		IF Q11 = 1 OR 2	IF Q11 = 3	IF Q11 = 4	IF Q11 = 5	IF Q11 = 6 OR 7
		Not often	Roughly 30% of the time	Roughly 50% of the time	Roughly 70% of the time	Very often
Q11_1_recode	Q11 Please indicate how often do you do the following: **Reduce your use of 'on-the-go' plastic (e.g. bring your own take-away coffee cup, bring your own take-away container)**					
Q11_2_recode	Q11 Please indicate how often do you do the following: **Reduce your use of packaging (e.g. buy at a 'packaging-free, zero-waste' store; avoid packaged personal care products)**					
Q11_3_recode	Q11 Please indicate how often do you do the following: **Reduce your use of non-disposable plastic (e.g. replace plastic containers with glass, buy wooden household goods as opposed to plastic goods)**					

Q12

Please rate durable plastic products (containers, furniture, stationary) against the following traits:

Coding		1	2	3	4	5
		2	1	0	1	2
Q12_1	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Harmful/Beneficial**					
Q12_2	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Bad/Good**					
Q12_3	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Inconvenient/Convenient**					
Q12_4	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Not useful/Useful**					
Q12_5	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Makes my life easier/Makes my life harder**					
Q12_6	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Bad for the environment/Good for the environment**					
Q12_7	Q12 Please rate durable plastic products (containers, furniture, stationary) against the following traits: **Bad for my health/Good for my health**					

Q13

Please indicate your level of agreement with each of the following statements:

Coding		1	2	3	4	5
		Strongly Disagree	Disagree	Neither Agree or Disagree	Agree	Strongly Agree
Q13_1	Q13 Please indicate your level of agreement with each of the following statements: **I would like to reduce my use of disposable plastic**					
Q13_2	Q13 Please indicate your level of agreement with each of the following statements: **I would like to reduce my use of plastic used in longer-term applications (buy items made from alternative materials)**					
Q13_3	Q13 Please indicate your level of agreement with each of the following statements: **It is important to those close to me that I reduce my use of disposable plastic**					
Q13_4	Q13 Please indicate your level of agreement with each of the following statements: **I feel pressure from society to reduce my use of disposable plastic**					
Q13_5	Q13 Please indicate your level of agreement with each of the following statements: **I have no control over how much disposable plastic I use**					

RQ13_reverse_recode

Please indicate your level of agreement with each of the following statements:

Coding		1	2	3	4	5
Recode formula		IF Q13 = 5	IF Q13 = 4	IF Q13 = 3	IF Q13 = 2	IF Q13 = 1
		Strongly agree	agree	Neither Agree or Disagree	Disagree	Strongly Disagree
Q13_5	Q13 Please indicate your level of agreement with each of the following statements: **I have no control over how much disposable plastic I use**					

Q14
Please indicate your level of agreement with each of the following statements:

Coding		1	2	3	4	5
		Strongly Disagree	Disagree	Neither Agree or Disagree	Agree	Strongly Agree
Q14_1	Q14 Please indicate your level of agreement with each of the following statements: **These days, too many items are made out of plastic**					
Q14_2	Q14 Please indicate your level of agreement with each of the following statements: **Most people in my community reduce their use of disposable plastics**					
Q14_3	Q14 Please indicate your level of agreement with each of the following statements: **If all plastic is recycled, there is no need to reduce my use of it**					
Q14_4	Q14 Please indicate your level of agreement with each of the following statements: **If plastic food packaging reduces food wastage, that justifies its increased use**					

Q15
Are you familiar with the term 'microplastics'? Would you say:

Coding	1	2	3
Q15 Are you familiar with the term 'microplastics'? Would you say:	I've heard of it and know what it means	I've heard of it but I'm not sure what it means	I've never heard of it

Q16
Do you think of microplastics negatively or positively?

Coding	1	2	3
Do you think of microplastics negatively or positively?	Positively	Negatively	Don't know

Q17
Please record the first two words/phrases that come to mind when you hear the word 'bioplastic'

Q17_1	Q17 Please record the first two words/phrases that come to mind when you hear the word 'bioplastic': 1
Q17_2	Q17 Please record the first two words/phrases that come to mind when you hear the word 'bioplastic': 2

Q17A
Please rate bioplastic food packaging against the following traits:

Coding		1	2	3	4	5
		2	1	0	1	2
Q17A_1	Q17A Please rate bioplastic food packaging against the following traits: **Harmful/Beneficial**					
Q17A_2	Q17A Please rate bioplastic food packaging against the following traits: **Bad/Good**					
Q17A_3	Q17A Please rate bioplastic food packaging against the following traits: **Inconvenient/Convenient**					
Q17A_4	Q17A Please rate bioplastic food packaging against the following traits: **Not useful/Useful**					
Q17A_5	Q17A Please rate bioplastic food packaging against the following traits: **Reduces food waste/Increases food waste**					
Q17A_6	Q17A Please rate bioplastic food packaging against the following traits: **Bad for the environment/Good for the environment**					
Q17A_7	Q17A Please rate bioplastic food packaging against the following traits: **Bad for my health/Good for my health**					

Q18
Please indicate whether you agree or disagree with the following statements:

Coding		1	2	3
		Disagree	Unsure	Agree
Q18_1	Q18 Please indicate whether you agree or disagree with the following statements: **All bioplastics are biodegradable**			
Q18_2	Q18 Please indicate whether you agree or disagree with the following statements: **I have used an item made from a bioplastic before**			
Q18_3	Q18 Please indicate whether you agree or disagree with the following statements: **Some bioplastics are indistinguishable from regular plastics**			

Q19, Q19
Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment?

Coding		1	2	3	4	5	6
		Much Worse	Somewhat Worse	About the same	Somewhat Better	Much Better	Other (elaborate in the text box below)
Q19_1	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? **Biodegradable plastic**						
Q19_2	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? **Biobased plastic**						
Q19_3	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? **Paper**						
Q19_4	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? **Degradable plastic**						
Q19_5	Q19 Considering food packaging applications and bags: compared to normal plastics, do you think the following materials are better for the environment or worse for the environment? **Glass**						
Q19OE	Q19OE If you have selected 'other' please elaborate on your answer.						

Q20
Do you think biodegradable plastics are better for the environment or worse for the environment when compared to easily recyclable plastics?

Coding	1	2	3	4	5	6
	Much worse	Somewhat worse	About the same	Somewhat better	Much better	Unsure
Q20 Do you think biodegradable plastics are better for the environment or worse for the environment when compared to easily recyclable plastics? Biodegradable plastics are:						

Q21
Please indicate whether you agree or disagree with the following statements:

Coding		1	2	3
		Disagree	Unsure	Agree
Q21_1	Q21 Please indicate whether you agree or disagree with the following statements: **All plastics made from plants are biodegradable**			
Q21_2	Q21 Please indicate whether you agree or disagree with the following statements: **Leaving a biodegradable plastic food package at the beach shouldn't be considered as littering because the material is biodegradable**			
Q21_3	Q21 Please indicate whether you agree or disagree with the following statements: **I would be less worried about plastic entering the ocean if it was biodegradable**			
Q21_4	Q21 Please indicate whether you agree or disagree with the following statements: **I would not be worried about plastic entering the ocean if it was biodegradable**			
Q21_5	Q21 Please indicate whether you agree or disagree with the following statements: **I would like more of the plastic items I use to be biodegradable**			
Q21_6	Q21 Please indicate whether you agree or disagree with the following statements: **Biodegradable plastics can have negative environmental impacts**			
Q21_7	Q21 Please indicate whether you agree or disagree with the following statements: **I would be happy to pay up to 10% more if a product was made from a biodegradable or biobased plastic rather than a regular plastic.**			
Q21_8	Q21 Please indicate whether you agree or disagree with the following statements: **I would be happy to pay up to 10% more if a product was made from recycled plastic rather than new plastic.**			

No Q22

Q23

Please select how you would currently dispose of a biodegradable plastic material (e.g. a food package or a take away container)

Coding		1	2	3	4	5
		Recycling bin	Regular bin	Home compost bin	Don't know	Other (please specify)
Q23	Q23 Please select how you would currently dispose of a biodegradable plastic material (e.g. a food package or a take away container) :					
Q23_5_OTHER	Q23 Please select how you would currently dispose of a biodegradable plastic material (e.g. a food package or a take away container): Other					

Q24

For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year.

Coding		1	2	3	4	5	6	7
		0% (None)	25%	50%	75%	100% (As much as possible)	I don't know	Can't be recycled in my area
Q24_1	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Glass bottles/containers**							
Q24_2	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Plastic bottles/containers**							
Q24_3	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Aluminum, steel and tin cans**							
Q24_4	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Paper/cardboard**							
Q24_5	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Food waste (eligible portion such as vegetable scraps)**							
Q24_6	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Garden waste**							

Q24_recode

For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year.

Coding		1	2	3	4	5	System missing
Recode		IF Q21=1	IF Q21=2	IF Q21=3	IF Q21=4	IF Q21=5	IF Q24 = 6 OR 7
		0% (None)	25%	50%	75%	100% (As much as possible)	I don't know/can't be recycled in my area
Q24_2	Q24 For the following waste items produced by your household, please indicate what percentage you recycled/composted in the last year. **Plastic bottles/containers**						

Q25

How important are the following factors in motivating your household to recycle?

Coding		0	1	2	3	4	5
		0 - Not Important	1	2	3	4	5 - Very important
Q25_1	Q25 How important are the following factors in motivating your household to recycle? **It is beneficial for the environment**						
Q25_2	Q25 How important are the following factors in motivating your household to recycle? **I think it is my civic duty**						
Q25_3	Q25 How important are the following factors in motivating your household to recycle? **I want to be seen by others as a responsible citizen**						

Q26

Are you familiar with this symbol for plastics? (image of a mobius strip plastic identification code)

Coding	1	2
	Yes	No
Q26 Are you familiar with this symbol for plastics?		

Q27

What does the symbol indicate?

Coding	1	2	3	4	5
	How many times the plastic item has been recycled	How many times the plastic item can be recycled	The type of plastic the item is made from	The category of plastic the item belongs to (e.g. film, bottle, multilayer)	I don't know
Q27 What does the symbol indicate?					

Q28

Do you look for instructions on an item you want to dispose of (e.g. look for a recycling symbol) to decide which items to put in the recycling bin and which items to put in the regular bin?

Coding	1	2
	Yes	No
Q28 Do you look for instructions on an item you want to dispose of (e.g. look for a recycling symbol) to decide which items to put in the recycling bin and which items to put in the regular bin?		

Q29

Please indicate the level of responsibility each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic.

Coding		1	2	3	4
		Not at all responsible	Somewhat responsible	Moderately responsible	Mostly responsible
Q29_1	Q29 Please indicate the level of responsibility each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic. **The government (through legislation)**				
Q29_2	Q29 Please indicate the level of responsibility each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic. **Companies/industry**				
Q29_3	Q29 Please indicate the level of responsibility each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic. **Individuals (through their consumer choices)**				

Q29_ranking

Please indicate the level of responsibility each of the following parties (Government, Industry and Individuals) for reducing the use of disposable plastic.

Coding		1	2	3	4	5	6	7
Recode formula		IF Q29_3 =29_2 & Q29_3 =29_1	IF Q29_3>29_2 & Q29_3>29_1	IF Q29_1>29_2 & Q29_1>29_3	IF Q29_2>29_1 & Q29_2>29_3	IF Q29_3=29_1 & Q29_3>29_2	IF Q29_2=29_1 & Q29_2>29_3	IF Q29_2=29_3 & Q29_2>29_1
		All equal	Individual greater than govt AND industry	Govt greater than individual AND industry	Industry greater than individual AND govt	Individual = govt and greater than industry	Industry = govt and greater than individual	Industry = individual and greater than govt
Q29_1	**The government (through legislation)**							
Q29_2	**Companies/industry**							
Q29_3	**Individuals (through their consumer choices)**							

Q30

When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally

Coding		1	2	3	4	5	6
		Not at all important	Low Importance	Slightly Important	Moderately Important	Very Important	Extremely Important
Q30_1	Q30 When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally - **No extra cost to the consumer**						
Q30_2	Q30 When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally - **No extra cost to the producer**						
Q30_3	Q30 When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally - **No extra inconvenience to the consumer**						
Q30_4	Q30 When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally - **All plastic recycling collected through roadside collection**						
Q30_5	Q30 When considering legislation aimed at reducing plastic waste, please indicate how important each of the following factors would be to you personally - **All recyclables go in the one collection bin**						

Q31

Please indicate the extent to which you agree or disagree with each of the following statements

Coding		1	2	3	4	5	6
		Strongly Disagree	Disagree	Neither Agree or Disagree	Agree	Strongly Agree	Don't know
Q31_1	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **Measures should be taken to reduce the use of single-use plastic items (e.g. shopping bags, straws...)**						
Q31_2	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **Disposing of plastic waste in landfill sites should be banned**						
Q31_3	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **The use of microplastics in consumer cosmetics and similar products should be banned**						
Q31_4	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **I would buy a water bottle made from recycled plastic even if the colour was more yellow/hazy than other bottled water (but I was assured it did not impact the quality of the bottle)**						

Q31_recode

Please indicate the extent to which you agree or disagree with each of the following statements

Coding		1	2	3	4	5	System missing
Recode formula		IF Q31 = 1	IF Q31 = 2	IF Q31 = 3	IF Q31 = 4	IF Q31 = 5	IF Q31 = 6
		Strongly Disagree	Disagree	Neither Agree or Disagree	Agree	Strongly Agree	Don't know
Q31_1	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **Measures should be taken to reduce the use of single-use plastic items (e.g. shopping bags, straws...)**						
Q31_2	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **Disposing of plastic waste in landfill sites should be banned**						
Q31_3	Q31 Please indicate the extent to which you agree or disagree with each of the following statements - **The use of microplastics in consumer cosmetics and similar products should be banned**						

Q32

Please indicate your level of support for each of the following waste management options:

Coding		1	2	3	4	5
		Strongly oppose	Somewhat oppose	Neutral	Somewhat favour	Strongly favour
Q32_1	Q32 Please indicate your level of support for each of the following waste management options: **Closed-loop recycling (e.g. making a new milk bottle from a recycled milk bottle)**					
Q32_2	Q32 Please indicate your level of support for each of the following waste management options: **Open-loop recycling (e.g. recycling plastic food packaging to make park benches)**					
Q32_3	Q32 Please indicate your level of support for each of the following waste management options: **Waste-to-energy (incineration to produce energy)**					
Q32_4	Q32 Please indicate your level of support for each of the following waste management options: **Landfill**					
Q32_5	Q32 Please indicate your level of support for each of the following waste management options: **Composting (when applicable)**					

Q32_recode

Please indicate your level of support for each of the following waste management options:

Coding		1	2	3
		IF Q32 =1,2	IF Q32 = 3	IF Q32 = 4,5
		Oppose	Neutral	Support
Q32_1	Q32 Please indicate your level of support for each of the following waste management options: **Closed-loop recycling (e.g. making a new milk bottle from a recycled milk bottle)**			
Q32_2	Q32 Please indicate your level of support for each of the following waste management options: **Open-loop recycling (e.g. recycling plastic food packaging to make park benches)**			
Q32_3	Q32 Please indicate your level of support for each of the following waste management options: **Waste-to-energy (incineration to produce energy)**			
Q32_4	Q32 Please indicate your level of support for each of the following waste management options: **Landfill**			
Q32_5	Q32 Please indicate your level of support for each of the following waste management options: **Composting (when applicable)**			

Q33

In general, are you concerned about environmental problems?

Coding	1	2	3	4
	No, not concerned	Yes, a little	Yes, a fair amount	Yes, a great deal
Q33 In general, are you concerned about environmental problems?				

Q34

Please indicate to what extent you agree or disagree with each of the following statements:

Coding		1	2	3	4	5
		Strongly disagree	Disagree	Neither agree nor disagree	Agree	Strongly agree
Q34_1	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **I am not willing to do anything about the environment if others don't do the same**					
Q34_2	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **Environmental impacts are frequently overstated**					
Q34_3	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **I am willing to make compromises in my lifestyle for the benefit of the environment**					
Q34_4	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **Policies introduced by the government to address environmental issues should not cost me extra money**					
Q34_5	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **I try to bear the environment and nature in mind in my daily behaviour**					

RQ34_reverse_recode

Please indicate to what extent you agree or disagree with each of the following statements:

Coding		1	2	3	4	5
recode formula		IF Q34=5	IF Q34=4	IF Q34=3	IF Q34=2	IF Q34=1
		Strongly agree	agree	Neither agree nor disagree	Disagree	Strongly disagree
Q34_1	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **I am not willing to do anything about the environment if others don't do the same**					
Q34_2	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **Environmental impacts are frequently overstated**					
Q34_4	Q34 Please indicate to what extent you agree or disagree with each of the following statements: **Policies introduced by the government to address environmental issues should not cost me extra money**					

Q35

Please indicate how similar the person described below is to yourself ‘Looking after the environment is important to this person; to care for nature and save life resources’

Coding	1	2	3	4	5	1
	Not at all like me	Not like me	A little like me	Somewhat like me	Very much like me	Not at all like me
Q35 Please indicate how similar the person described below is to yourself ‘Looking after the environment is important to this person; to care for nature and save life resources’						

Q36

Please indicate to what extent you agree or disagree with each of the following statements:

Coding		1	2	3	4	5
		Strongly disagree	Disagree	Neither Agree or Disagree	Agree	Strongly agree
Q36_1	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Individuals can not contribute to the reduction of plastic pollution.**					
Q36_2	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **I feel partly responsible for plastic pollution.**					
Q36_3	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Using less plastic will not significantly reduce humans' environmental impact.**					

Q36_3_recode

Please indicate to what extent you agree or disagree with each of the following statements:

Coding		1	2	3
Recode formula		IF Q36_3 = 1 OR 2	IF Q36_3 = 3	IF Q36_3 = 4 OR 5
		Disagree	Neither Agree or Disagree	Agree
Q36_1	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Individuals can not contribute to the reduction of plastic pollution.**			
Q36_2	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **I feel partly responsible for plastic pollution.**			
Q36_3_recode	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Using less plastic will not significantly reduce humans' environmental impact.**			

RQ36_reverse_recode

Please indicate to what extent you agree or disagree with each of the following statements:

Coding		1	2	3	4	5
Recode formula		IF Q36 = 5	IF Q36 = 4	IF Q36 = 3	IF Q36 = 2	IF Q36 = 1
		Strongly agree	agree	Neither Agree or Disagree	Disagree	Strongly disagree
Q36_1	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Individuals can not contribute to the reduction of plastic pollution.**					
Q36_3	Q36 Please indicate to what extent you agree or disagree with each of the following statements: **Using less plastic will not significantly reduce humans' environmental impact.**					

Demographics

Q37
What is your gender

Coding		1	2	3	4
		Male	Female	Prefer not to say	Other (please specify)
Q37	Q37 What is your gender?				
Q37_4_OTHER	Q37 What is your gender? Other				

Gender

Coding		1	2	System missing
Recode		If Q37 = 1	IF Q37 = 2	IF Q31 = 3 OR 4
		Male	Female	System missing
Q37	Q37 What is your gender?			

Q38
What is your year of birth (YYYY)?
Converted by market research company into age brackets

Coding	1	2	3	4	5	6
	18-24	25-34	35-44	45-54	55-64	65+
What is your age?						

Q39
In which Australian State/Territory do you live?

Coding	1	2	3	4	5	6	7	8
	Australian Capital Territory	New South Wales	Northern Territory	Queensland	South Australia	Tasmania	Victoria	Western Australia
Q39 In which Australian State/Territory do you live?								

Q40
How would you best describe the area in which you live?

Coding		1	2	3	4	5	6
		Major city	Suburban edges of a major city	Major town	Small town	Remote	Other (please specify)
Q40	Q40 How would you best describe the area in which you live?						
Q40_6_OTHER	Q40 How would you best describe the area in which you live? Other						

Q41
In which type of dwelling do you live?

Coding		1	2	3	4	5
		House - detached	Townhouse (or other semi-detached dwelling)	Unit/Flat	Boarding house/College	Other (please specify)
Q41	Q41 In which type of dwelling do you live?					
Q41_5_OTHER	Q41 In which type of dwelling do you live? Other					

Q42
Which of the following best describes your household?

Coding		1	2	3	4	5	6	7
		Living full time with dependent child(ren) as parent or carer	Living part time with dependent child(ren) as parent or carer	Living with a partner without child(ren)	Living with other family members (e.g. parents, extended family or non-dependent children)	Living in shared household with people other than family	Living in a single person household	Other (please specify)
Q42	Q42 Which of the following best describes your household?							
Q42_7_ OTHER	Q42 Which of the following best describes your household? Other							

Q42_recode
Children vs no children

Coding	1	2
Recoding formula	IF Q42 = 1 OR 2	IF Q42 = 3,4,5,6 OR 7
	Has children	Doesn't have children
Does the respondent have children that live with them?		

Q43
How many dependent children live in your household?

Coding	1	2	3	4	5	6
	0	1	2	3	4	5 or more
Q43 How many dependent children live in your household?						

Q44
How many children under 5 live in your household?

Coding	1	2	3	4	5
	0	1	2	3	4 or more
Q44 How many children under 5 live in your household?					

Q45
Which best describes the highest level of education you have achieved or are studying for?

Coding		1	2	3	4	5	6	7
		Year 10 or below	Year 11 or equivalent	Year 12 or equivalent	Trade certificate or apprenticeship	Bachelor or honours degree	Postgraduate degree (e.g. Masters, PhD)	Other (please specify)
Q45	Q45 Which best describes the highest level of education you have achieved or are studying for?							
Q45_7_OT HER	Q45 Which best describes the highest level of education you have achieved or are studying for? Other							

Q46
Which of the following categories best describes your occupational status?

Coding		1	2	3	4	5	6	7	8
		Student	Employed - full time	Employed - part time	Unemployed - not looking for work	Unemployed - looking for work	Retired	Unable to work	Other (please specify)
Q46	Q46 Which of the following categories best describes your occupational status?								
Q46_8_OTHER	Q46 Which of the following categories best describes your occupational status? Other								

Q47
How much is your estimated household income before tax? (survey responses are NOT linked to any personally identifying information)

Coding	1	2	3	4	5	6	7	8	9
	Less than $25,000	$25,000 - $49,999	$50,000 - $74,999	$75,000 - $99,999	$100,000 - $124,999	$125,000 - $174,999	$175,000 - $199,999	More than $200,000	Prefer not to say
Q47 How much is your estimated household income before tax? (survey responses are NOT linked to any personally identifying information)									

Q48
In politics, people sometimes talk about the 'left' and the 'right'. On a scale where '0' means left and '10' means right, where would you place yourself on the scale?

Coding	0	1	2	3	4	5	6	7	8	9	10
	0 - LEFT	1	2	3	4	5	6	7	8	9	10 - RIGHT
Q48 In politics, people sometimes talk about the 'left' and the 'right'. On a scale where '0' means left and '10' means right, where would you place yourself on the scale?											

Q48_recode
In politics, people sometimes talk about the 'left' and the 'right'. On a scale where '0' means left and '10' means right, where would you place yourself on the scale?

Coding	1	2	3
Recoding formula	IF Q48 = 0,1,2,3 OR 4	IF Q48 = 5	IF Q48 = 6,7,8,9 OR 10
	Left of centre	Centre	Right of centre
Q48 In politics, people sometimes talk about the 'left' and the 'right'. On a scale where '0' means left and '10' means right, where would you place yourself on the scale?			

APPENDIX F Copy of ethics approval letters

THE UNIVERSITY OF QUEENSLAND

Sub-Committee Human Research Ethics Approval

Project Title:	Understanding Australians' Attitudes Towards Plastics
Chief Investigator:	Dr Bronwyn Laycock, Dr Peta Ashworth, Ms Leela Dilkes-Hoffman
Supervisor:	Dr Bronwyn Laycock, A/Prof Steven Pratt, Prof Paul Lant
Co-Investigator(s):	A/Prof Steven Pratt, Prof Paul Lant
School(s):	School of Chemical Engineering
Approval Number:	2018000492
Granting Agency/Degree:	PhD
Duration:	1st March 2019

Comments/Conditions:

- HREA Form, 08/03/2018
- Participant Information Sheet (Focus Groups), 08/03/2018
- Participant Consent Form (Focus Groups), 08/03/2018
- Project Description, 08/03/2018
- Online survey, 08/03/2018
- Focus group questions, 08/03/2018
- A quick guide to bioplastics, 08/03/2018

Note: If this approval is for amendments to an already approved protocol for which a UQ Clinical Trials Protection/Insurance Form was originally submitted, then the researchers must directly notify the UQ Insurance Office of any changes to that Form and Participant Information Sheets & Consent Forms as a result of the amendments, before action.

Name of responsible Sub-Committee:
University of Queensland Engineering, Architecture and Information Technology, Low & Negligible Risk Ethics Sub-Committee
This project complies with the provisions contained in the *National Statement on Ethical Conduct in Human Research* and complies with the regulations governing experimentation on humans.

Name of Ethics Sub-Committee representative:
Professor Deanna Kemp
Acting Chairperson
University of Queensland Engineering, Architecture and Information Technology, Low & Negligible Risk Ethics Sub-Committee

Signature ____________ Date **27/04/2018**

THE UNIVERSITY OF QUEENSLAND

Sub-Committee Human Research Ethics Approval

Project Title:	Understanding Australians' Attitudes Towards Plastics – 10/05/2018 AMENDMENT
Chief Investigator:	Dr Bronwyn Laycock, Dr Peta Ashworth, Ms Leela Dilkes-Hoffman
Supervisor:	Dr Bronwyn Laycock, A/Prof Steven Pratt, Prof Paul Lant
Co-Investigator(s):	A/Prof Steven Pratt, Prof Paul Lant
School(s):	School of Chemical Engineering
Approval Number:	2018000492
Granting Agency/Degree:	PhD
Duration:	1st March 2019

Comments/Conditions:

Amendment 10/05/2018:

- Perform national survey before focus groups;
- Re-worded survey questions;
- Voluntary collection of participants emails during a survey;
- 13th March 2018 Draft Online Survey Questions, 10/05/2018;
- Questionnaire Final, 10/05/2018;
- Project Description Final V2, 10/05/2018.

Note: If this approval is for amendments to an already approved protocol for which a UQ Clinical Trials Protection/Insurance Form was originally submitted, then the researchers must directly notify the UQ Insurance Office of any changes to that Form and Participant Information Sheets & Consent Forms as a result of the amendments, before action.

Name of responsible Sub-Committee:
University of Queensland Engineering, Architecture and Information Technology, Low & Negligible Risk Ethics Sub-Committee
This project complies with the provisions contained in the *National Statement on Ethical Conduct in Human Research* and complies with the regulations governing experimentation on humans.

Name of Ethics Sub-Committee representative:
Professor Deanna Kemp
Acting Chairperson
University of Queensland Engineering, Architecture and Information Technology, Low & Negligible Risk Ethics Sub-Committee

Signature ____________ Date **15/05/2018**

APPENDIX G Mass flows for Sankey Diagram

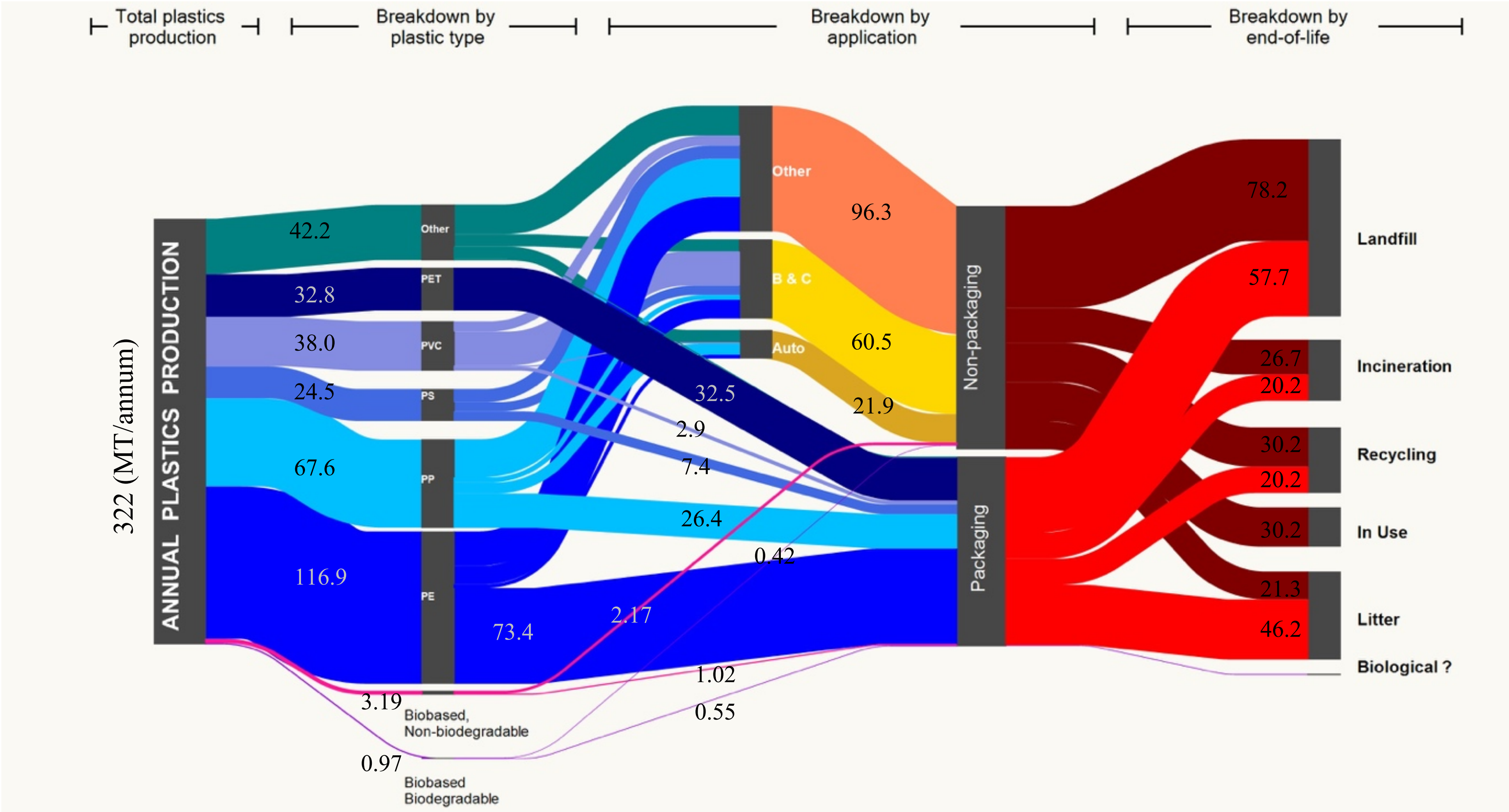


Figure G1: Mass flows for Sankey Diagram (all numbers in million tonnes per annum)