



Cite this: *Environ. Sci.: Adv.*, 2025, 4, 2091

## Are you being served? An examination of chemical risks posed by compostable single-use food service items

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The banning of single-use plastic food service items has led to a shift towards single-use, compostable alternatives. Globally, decision-makers are grappling with balancing the largely uncharacterised risk of compostable single-use food service items with the potential benefits of diverting additional food waste from landfills. To help close this gap in regulatory understanding, food service items were collected from semi-closed locations that only allow compostable items and analysed for their physical and chemical properties. Collected items represented the broad range of material and item types associated with 'compostable' single-use food service items, including food service vessels, straws, cutlery, napkins, sandwich bags and wraps. Analysis determined the likely systematic inclusion of perfluoroalkyl and polyfluoroalkyl substances (PFAS) across a range of sugarcane bagasse samples with a maximum detected concentration of 86 200  $\mu\text{g kg}^{-1}$ . Additionally, food service item components such as plastic linings and lids were often found to contain common, non-compostable plastics. Findings indicate that at present, single-use plastic item bans have shifted manufacturers of single-use food service items towards material choices that do not possess viable resource recovery pathways. This paper emphasises the need for policymakers to consider likely market shifts and the risks associated with likely alternative materials prior to taking action on single-use plastic items.

Received 2nd July 2025  
Accepted 29th September 2025

DOI: 10.1039/d5va00195a

rsc.li/esadvances

### Environmental significance

Many single-use food service item manufacturers, businesses and government policies are switching from single-use plastics in favour of compostable items. This paper identified a series of chemical and physical contaminants likely to impact the plastics and organics policy positions of a variety of global jurisdictions. 'Compostable' single use food service items contained contaminants that are detrimental to compost, including high concentrations of PFAS and non-compostable plastics. Additionally, low concentrations of contaminants such as heavy metals, phthalates, and PFAS were found in items made from recycled content. Non-compostable plastics were also detected in supposedly 'non-plastic' items, such as paper straws.

## Introduction

The waste hierarchy is a set of ranked priorities for the preferential use and disposal of waste. Waste avoidance and reusability are considered the most preferable outcomes, while recycling-based outcomes are considered preferable to waste treatment and waste disposal pathways.<sup>1</sup> Recently, governments have shifted towards policy that prioritises circular economy outcomes, aiming to optimise waste hierarchy outcomes while keeping materials in circulation at their highest possible value for as long as possible.<sup>2</sup> Concurrently, many governments have

been banning the supply of various plastic food service items to reduce the consumption of single-use plastic.<sup>3</sup> These policies aimed to stimulate the uptake of reusable food service items and encourage waste avoidance amongst businesses and consumers.<sup>4</sup> However, these bans have instead shifted some food service item manufacturers and businesses towards non-plastic and compostable plastic single-use alternatives.<sup>5</sup> While these alternatives have decreased the total volume of conventional single-use plastic food service items, they have not encouraged higher waste hierarchy or circular economy-based outcomes. In most jurisdictions, all single-use food service items are destined for either incineration or landfilling.<sup>6</sup>

Most jurisdictions, with some exceptions, such as Italy and South Australia, do not permit compostable food service items to be processed within their industrial composting facilities.<sup>7,8</sup> Most compostable food service items are made from refined, low moisture content cellulosic material or from compostable

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plastics such as polylactic acid (PLA).<sup>9</sup> These materials are typically derived from plants such as sugarcane, birch, eucalyptus and pine.<sup>10,11</sup> While compostable food service items can introduce additional carbon to compost mixtures, they pose risks and provide negligible benefits to land upon compost application.<sup>7,12,13</sup> As such, the composting of packaging materials is more akin to a form of waste treatment than genuine resource recovery.<sup>14</sup> Arguments for including food service items into commercial composting streams are often made on the premise of additional food organics recovery and diversion of packaging from landfills.<sup>15</sup> However, in the hospitality industry, consumers typically eat most of their purchased food, leaving minimal plate waste.<sup>16,17</sup>

Recently, many individual businesses, food court operators and catering agencies have opted to shift towards supplying exclusively compostable food service items, irrespective of local legislative requirements.<sup>18</sup> The impetus for this shift often relates to practical considerations regarding the ease of improving and simplifying reporting of broad environmental, social and governance (ESG) metrics such as resource recovery rates.<sup>19</sup> However, material compliance efforts are often complicated by difficulties in the supply chain, such as manufacturers and distributors selling a mixture of 'certified-compostable', uncertified, and often entirely non-compostable products.

Compostable plastic certifications, such as AS 4736:2006, ISO 17088:2021, EN 14995:2006, and EN 13432:2000, are often used to ensure material quality.<sup>20–23</sup> It is often difficult for a consumer to know if an item has been 'certified compostable' by meeting a relevant standard, if it's likely to compost but has not met a standard, or if it's unlikely to compost.<sup>24</sup> For instance, certified compostable bagasse bowls are almost indistinguishable from their uncertified counterparts. Some items, such as paper straws, wooden cutlery, cardboard clamshells, and napkins, are rarely certified, as standards like AS 4736-2006 are primarily designed to examine plastic materials.<sup>21</sup> Vessels labelled as compostable may also be supplied with non-compostable lids, leading to further consumer confusion and potentially incorrect disposal. This issue is further complicated by the fact that compostable standards often do not align with local disposal rules, industrial composting practices, or the chemical risks associated with new materials.<sup>7</sup> For instance, the timeframes and temperatures used in the certifications often do not reflect industrial or home composting practices and conditions, leading to incomplete breakdown of compostable plastic material.<sup>25</sup> Additionally, the proxy methodologies used to assess ecotoxicity, such as short-term worm and plant growth tests, do not reflect the risk of multiple reapplications of compost containing persistent and bioaccumulative chemicals.<sup>26</sup>

The shift towards fibrous items such as paper straws and bagasse bowls has promoted the use of chemical additives to impart plastic qualities such as hydrophobicity and grease resistance to non-plastic items.<sup>27–31</sup> Persistent chemicals used in the manufacturing of these items such as perfluoroalkyl and polyfluoroalkyl substances (PFAS) and non-compostable plastic inks, coatings and adhesives have potential negative impacts on

the environment and human health.<sup>32,33</sup> Once land-applied in compost, persistent contaminants may decrease soil health, function and structure, potentially impacting plant growth and crop quality. Additionally, contaminants may be taken up by crops and then leave the application site *via* human or animal consumption.

This study aims to capture chemical risks present in various 'compostable' food service items used in semi-closed commercial environments within New South Wales (NSW), Australia. At the time of collection, each semi-closed commercial site mandated the supply of compostable food service items (with limited exceptions), intended for co-disposal with residual food organics. Previous studies have primarily examined the chemical risks and user experience of compostable packaging with respect to human consumption.<sup>33–35</sup> In contrast, this study focuses on assessing if the item's constituent materials are fit for the purpose of entering compost and subsequently being applied to land. The study examines the concentrations of potentially pervasive, accumulative, and harmful contaminants such as PFAS, phthalates, metals, and plastics that may pose a risk to the environment and human health if land applied *via* compost. The analytes examined are not exhaustive and do not reflect the total chemical risk of the materials. The findings of this study demonstrate the need for precautionary approaches to contemporary circular economy policy to avoid future legacy issues.

## Methodology

### Collection

A total of 162 food service items were collected for analysis from private vendors within three universities, the back of house of two large semi-closed institutions without private vendors, and a major supplier of compostable food service items. Private vendors participating from the universities were asked to supply specific food service packaging items for the study voluntarily. A range of items was requested from all sampling sites to ensure the diversity of food service item packaging types was captured.

### Characterisation

**Lid examination.** If items collected at universities are traditionally supplied in two pieces, such as a lidded cup, the complete set of items was requested. The lid materials were analysed to ensure all components of a supplied item had the same consumer disposal pathway. Lid to vessel matching was unable to be performed when items were collected from an institutions' back of house or directly from the food service item manufacturer.

**Material thickness examination.** Measurements of material gauge are important in the assessment of food service items, especially when they are proposed to enter composting pathways. The various compostable plastic standards require material to be certified to a maximum thickness.<sup>20</sup> If the material's physical and chemical risk profiles are commensurate, considerations around factors such as material usage may determine which items are preferable. Food service items were



cut to expose the item's flattest possible surface, and the gauge of the material was measured using a Mitutoyo 547-401 digital thickness gauge. Three measurements were taken, and results were averaged for each item. Actual physical measurements and distributions may be useful for future life-cycle assessments.

**Laboratory sample selection.** All collected items were physically examined and categorised into material types and item types (Table 1). A 50-sample subset was then selected for further laboratory analysis. Selected samples were chosen to avoid duplication of identical items and represent the broadest gamut of material types collected across the different food service item formats. Due to a mixture of material requirements, regulatory restrictions, and vendor preferences, certain items, such as straws, were only collected in a single material type.

### Total properties analysis

The concentration of metals, PFAS and phthalates in the food surface items were measured using a mixture of inductive coupled plasma atomic emission spectroscopy (ICP-AES), liquid chromatography mass spectrometry (LC-MS/MS), and gas chromatography mass spectrometry (GC-MS). Data produced from these analytical methods is representative of the average cross-section of the entire item, including material layers, surface coatings, adhesives, inks and dyes. Further details of methods, analytical reagents, analytes examined, and limits of reporting can be found in the SI.

### Metals analysis

Samples were cut into small pieces, with 1.0–1.2 g of each sample being digested in acid. The material was digested in a combination of concentrated nitric acid (3 mL) and hydrochloric acid (3 mL) at 90–98 °C in a hotblock for at least 2 hours. The acid digests were cooled and then diluted to volume (50 mL) with high purity water. Extracts were then analysed for trace metals using Inductively Coupled Argon Absorption Emission Spectrometry (ICP-AES Agilent 5900 or Agilent 5110) and Cold Vapour Atomic Absorption spectrophotometry (CV-AAS CETAC 7600) for mercury. The list of elements, wavelengths of detection, and detection limits of elements analysed can be found in the SI (Table SI 5).

### PFAS analysis

Samples were cut into small pieces, 1.0–1.2 g of each sample was then extracted with 10 mL basified methanol (prepared by adding 2.2 mL of 30% ammonium hydroxide to 1 L methanol) and 10 µL mass labelled surrogate  $^{13}\text{C}_2$  PFOA (1 mg L<sup>-1</sup>). Samples were extracted by 30 minutes of sonication followed by overnight end-over-end tumbling at  $30 \pm 2$  rpm. After adding 20 µL of glacial acetic acid, the extracts were filtered (0.2 µm) and diluted 5-fold in a 1 : 1 mixture of methanol and ultra-high purity (UHP) water. A mix of mass labelled PFAS internal standards were added (10 µL of 200–1000 µg L<sup>-1</sup> concentration range to 1 mL diluted extract, on column at 0.2–1.0 µg L<sup>-1</sup>) and run on LC-MSMS (Shimadzu 8050) (Table SI 2). PFAS analytes were separated *via* gradient elution using two mobile phases: (A) 5 mM ammonium acetate (prepared by adding 10 mL of 500 mM ammonium acetate and 0.5 mL of glacial acetic acid to approximately 900 mL of filtered ultra-high purity water, then made up to 1 L with UHP water), and (B) methanol. Chromatographic separation was performed using an ACE Excel 1.7 SuperC18 column (20 × 2.1 mm, Avantor). The complete list of PFAS analytes examined along with their limits of reporting is provided in Table SI 1 of the SI accompanying this paper.

### TOPA PFAS analysis

A 4 mL aliquot of the methanol PFAS extract detailed above was spiked with 5 µL of 8.75 mg L<sup>-1</sup>  $^{13}\text{C}$  8 FOSA to trace oxidation, then slowly evaporated to dryness. The sample was resuspended in a caustic persulfate solution containing approximately 1 g of potassium persulfate, 1 mL of 10 N sodium hydroxide and 7.5 mL of ultra-high purity (UHP) water. The oxidizing solution was heated to approximately 85 °C for over 6 hours, then cooled, adjusted to a pH of 6–7 with 20% (v/v) hydrochloric acid, before dilution to 10 mL with UHP water. The sample was then stabilised by the addition of 10 mL of methanol (final solvent ratio 1 : 1 methanol : water). A mix of mass labelled PFAS internal standards are added (10 µL of 200–1000 µg L<sup>-1</sup> concentration range to 1 mL diluted extract, on column at 0.2–1.0 µg L<sup>-1</sup>). The sample was then filtered (0.2 µm) prior to LC-MSMS analysis as described above (Shimadzu 8050).

**Table 1** Number of each item and material type analysed for chemical contaminants and plastic content

Item	Paper	Bagasse	Plastic-lined	Cardboard	Wood	Total
Straws	5	0	0	0	0	5
Sandwich wraps & bags	5	0	1	0	0	6
Napkins	5	0	0	0	0	5
Clamshells & trays	0	6	0	5	0	11
Bowls, plates & containers	0	4	5	0	0	9
Cutlery	0	2	0	0	4	6
Cup lids	0	2	1	0	0	3
Plastic-lined paper cups <sup>a</sup>	0	0	5	0	0	5
Total	15	14	12	5	4	50

<sup>a</sup> Cups were only examined for plastics and not for other contaminants of concern.



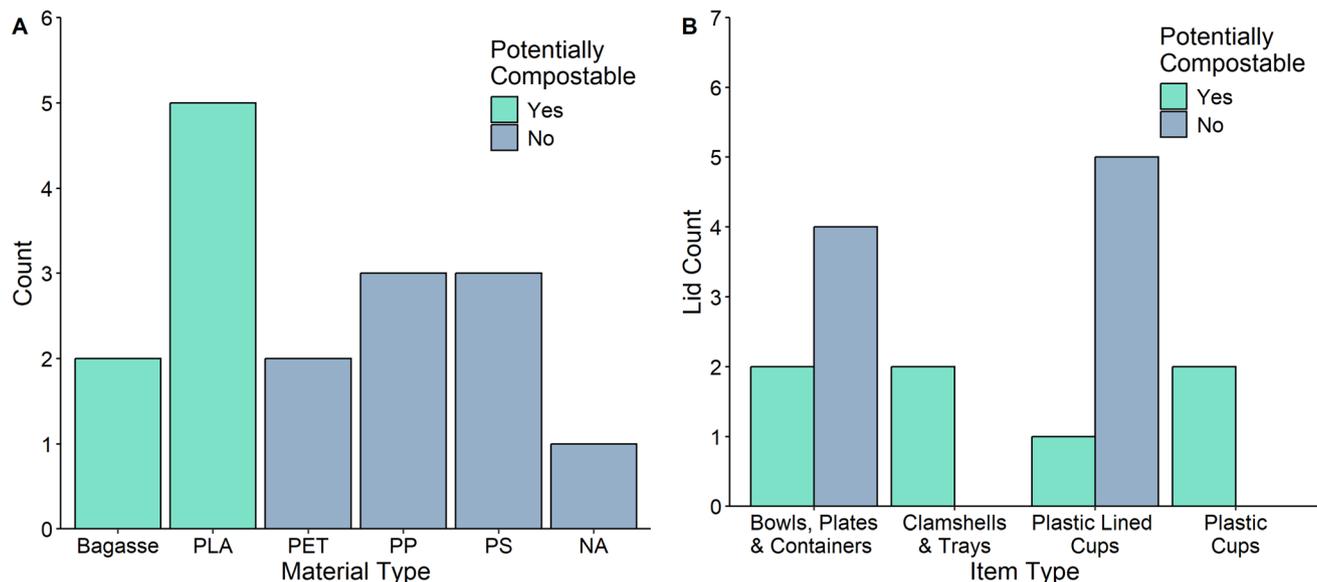


Fig. 1 Materials used as compostable food service packaging lids ( $n = 16$ ) with potentially compostable and non-compostable materials represented by blue and green, respectively. (A) The materials used as lids for compostable food service vessels. The plastic-lined paper sample was a cup lid with a lining that was found to be made of PE. (B) The use of compostable and non-compostable lids across different vessel types.

### Phthalate analysis

Samples were cut into small pieces, and 0.6–1.2 g and extracted with 20 mL of a 1 : 1 methylene chloride : acetone solvent mix. Prior to extraction four surrogates were added to assess recovery: nitrobenzene- $d_5$ , 2-fluorobiphenyl, 2,4,6-tribromophenol, and *p*-terphenyl- $d_{14}$ . The sample and solvent mix were tumbled end-over-end at  $30 \pm 2$  rpm for more than 4 hours, then left overnight. The resulting extracts were vialled and analysed by GC-MS/MS (Shimadzu TQ8050). Instrument internal standards, including deuterated compounds such as acenaphthene- $d_{10}$ , chrysene- $d_{12}$ , naphthalene- $d_8$ , perylene- $d_{12}$ , and phenanthrene- $d_{10}$ , are added online to correct for analytical variations. The GC separation was performed using an Agilent Ultra-Inert DB-5 ms column. The list of phthalate analytes examined along with their limits of reporting is provided in the SI accompanying this paper (Table SI 3).

### Quality assurance and quality control

Duplicate samples were prepared and analysed at a minimum frequency of 1 in 10. A method blank was included with every batch of over 20 samples, undergoing the full preparation and analysis. A laboratory control sample, where analytes or a subset are spiked into a matrix, is also processed to assess recovery, except when analysing TOPA PFAS, where it is not applicable.

### Surface properties analysis

The properties of the food contact surface were characterised using a combination of X-ray photoelectron spectroscopy (XPS), attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR), and Raman spectroscopy. For item types that were not homogenous such as plastic-lined items, unless otherwise stated, the surface in contact with food was

characterised. For items such as straws, the innermost layer of material was characterised.

### XPS analysis

XPS analysis was run on a Kratos Axis Supra + spectrometer and provided elemental composition of the first 10 nm of material thickness. The characterisation was carried out in ultrahigh

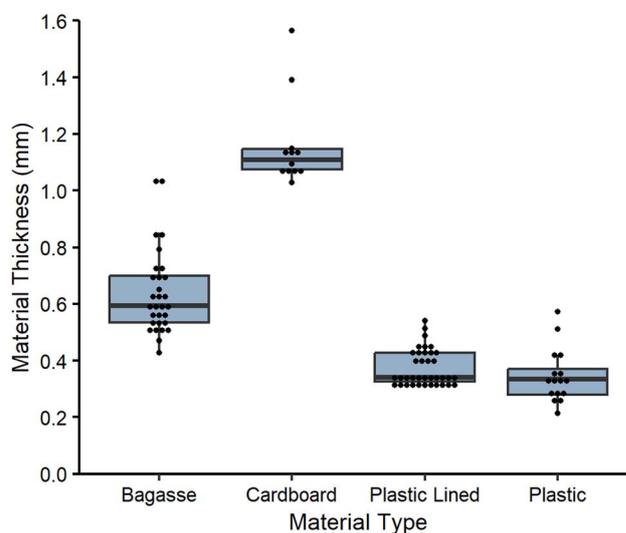


Fig. 2 Material gauge of single-use compostable food service vessels made from different materials ( $n = 95$ ). Cardboard materials exhibited the greatest material thickness followed by bagasse and plastic-lined items, with entirely plastic vessels using the lowest gauge of material. The choice to use a particular material type will be application specific, for instance plastic-lined paper vessels are more suitable than entirely plastic items for high temperature applications such as to contain hot coffee.



vacuum system over a rectangular area of the sample with dimensions  $300 \times 700 \mu\text{m}^2$  using a monochromatic Al K $\alpha$  X-ray source (1486.6 eV photon energy) at power output of 300 W. Survey spectra were collected over a range of 0–1100 eV binding energy with a dwell time of 55 ms using a pass energy of 160 eV and 0.5 eV step size with 3 sweeps. Collected data was charge corrected to adventitious carbon located at 285.0 eV. All spectra were analysed using CasaXPS (version 2.3.24PR1.0).<sup>36</sup>

### FTIR analysis

Infrared spectra were collected between  $4000 \text{ cm}^{-1}$  and  $800 \text{ cm}^{-1}$  under vacuum on a Bruker Vertex 80v FTIR spectrometer equipped with a diamond ATR attachment. FTIR spectrum of each sample comprised an average of 50 scans at a resolution of  $2 \text{ cm}^{-1}$ . Spectral matching was performed with Thermo Fisher OMNIC software using the in-built proprietary library and a user database.

### Raman analysis

Raman spectroscopic analysis was performed on a XploRA Horiba Scientific confocal Raman system using a  $\times 50$  objective

(numerical aperture 0.6). A laser wavelength of 638 nm was used in most cases. If the Raman signal was obscured by fluorescence at 638 nm, a 786 nm laser was selected instead. Laser power, integration time and scan number were selected on a case-by-case basis to achieve suitable signal-to-noise ratios for spectral matching. Elevated baselines due to fluorescence were removed from the spectra using a cubic spline function. Spectral matching was performed using the Wiley KnowItAll software with Wiley proprietary library and a user database.

## Results and discussion

### Lids

Vendors supplied many lidded vessels with lids and bases made of different materials, often compromising the potential compostability of the item set. Of the 19 items collected from university vendors that would typically be supplied with lids, 16 lids were collected. The 3 lids that were not collected were from a typically film lined plastic-lined paper cup (bubble tea), a bagasse bowl and an entirely plastic polyethylene terephthalate (PET) cup. Of the collected lids, 44% were made from potentially compostable materials and 56% were made from

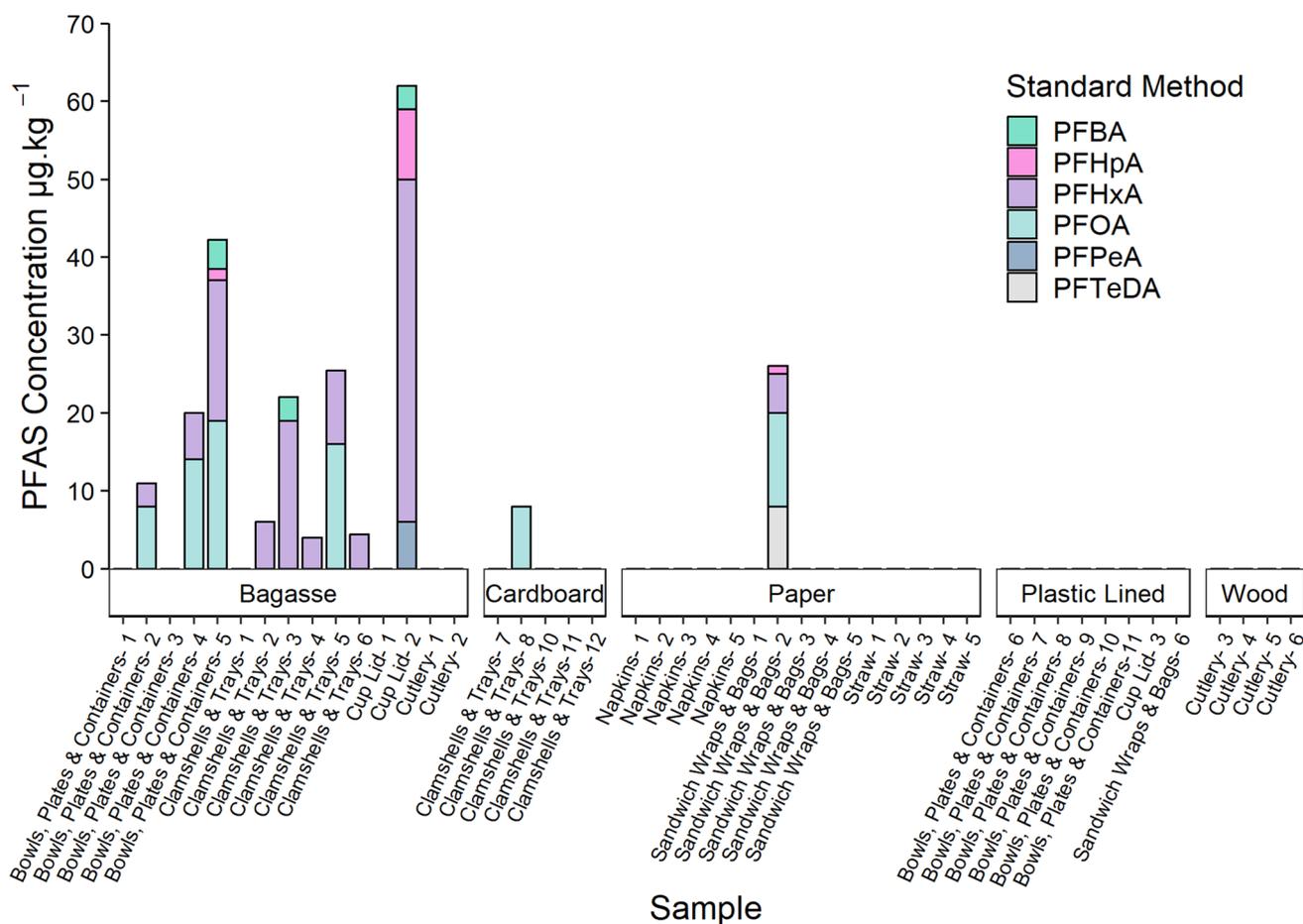


Fig. 3 PFAS concentrations for each sample examined separated by material type and PFAS species. PFAS was found to be present in 8/14 bagasse samples 1/5 cardboard samples and 1/15 paper samples. No PFAS was detected in either plastic-lined, or wooden samples examined. The most common PFAS species detected were PFHxA and PFOA which were detected at maximum concentrations of 44 and  $19 \mu\text{g kg}^{-1}$  respectively.



conventional non-compostable plastics. Most lids were made from plastics such as PLA, polyethylene (PE), polystyrene (PS) and PET, with only two lids being made of cellulosic material (Fig. 1). A single plastic-lined paper lid was collected, which, upon further examination, was found to likely be lined with polyethylene and, as such was considered a non-compostable lid. All entirely plastic vessels were supplied with a lid that matched the accompanying cup's body. Bowls, containers, paper cups, and trays typically used mismatched lid and vessel materials.

Discussions with business operators within the universities revealed two potential reasons for the high prevalence of non-compostable lids used on compostable items:

1. Several businesses claimed compostable lids to be unsuitable for hot beverages, as they experienced material deformation when lids were stored on top of coffee machines; and
2. Businesses often have limited control of their food service supply chain, with their packaging distributor supplying non-compostable plastic lids to fit their compostable vessels.

## Material gauge

Properties such as the gauge of material are often used in life cycle analysis studies to make the comparisons that allow for the efficiency of material use to be determined.<sup>32,37,38</sup> Additionally, many certification bodies such as Standards Australia certify compostable materials to specified tested thicknesses.<sup>20</sup> Cardboard vessels used the highest overall gauge of material with an average thickness of 1.16 mm, while bagasse vessels averaged 0.64 mm and plastic-lined and entirely plastic vessels offered greater efficiency with average gauges of 0.37 and 0.34 mm respectively (Fig. 2). The optimal material choice for a food service application depends on factors such as food temperature, food type, required water and grease resistance, item size, and other more consumer-based preferences such as mouth feel and texture.

## PFAS

Globally, jurisdictions regulate specific PFAS compounds in compost through various mechanisms, including product restrictions, compost input restrictions, concentration limits in

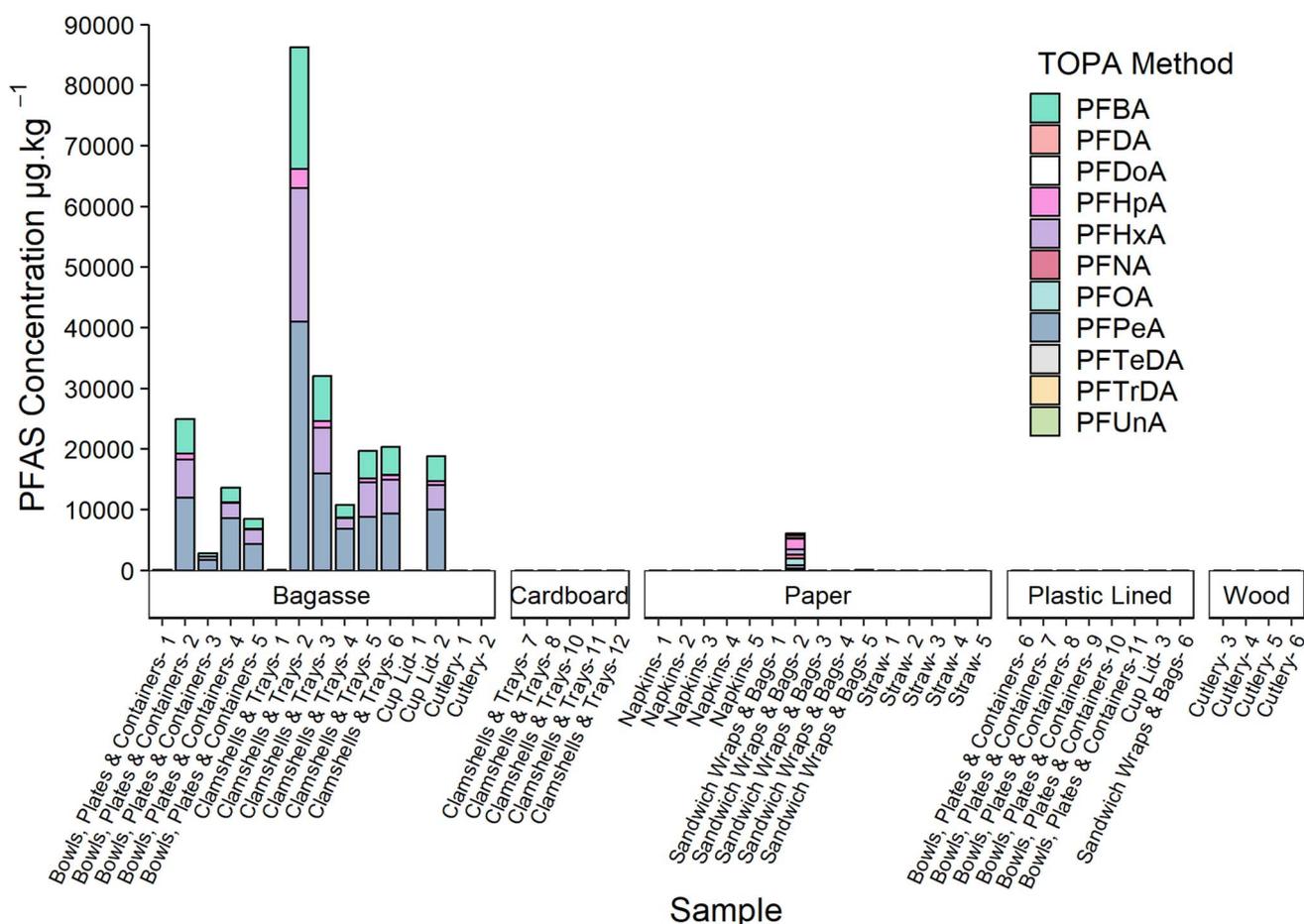


Fig. 4 PFAS concentrations post TOP assay for each sample examined separated by material type and PFAS species. PFAS was found to be present in 11 of 14 bagasse samples examined, with 9 samples possessing total concentrations of over 2500  $\mu\text{g kg}^{-1}$  and two other samples having lower concentrations of 110 and 70  $\mu\text{g kg}^{-1}$ . One sandwich wrap also contained elevated PFAS concentrations with a total detected PFAS concentration of  $\sim 6100 \mu\text{g kg}^{-1}$ . Lower concentrations of PFAS less than 50  $\mu\text{g kg}^{-1}$  were found in 2/5 cardboard samples, 2/15 paper samples and 1/4 wooden samples.



finished compost, and concentration limits at land application sites.<sup>39,40</sup> Permissible PFAS concentrations have generally become more stringent over time, and the scope of regulated compounds has also expanded in response to growing evidence of harm and improved analytical capabilities.<sup>39,41,42</sup> PFAS are not permitted within several compostable food service item certification standards such as AS 4736-2006 and ISO 17088:2021.<sup>21,22</sup> Of the 38 PFAS species analysed, 6 species of perfluoroalkyl carboxylic acids were detected with perfluorohexanoic acid (PFHxA) and perfluorooctanoic acid (PFOA) being the most frequently detected species (Fig. 3). The majority of PFAS detections were in bagasse items with 8 of the 14 samples examined exhibiting PFAS concentrations above the limit of reporting (LOR). The use of PFAS in bagasse items is likely due to manufacturers requiring greater water and grease resistance than the resistance provided by the raw bagasse material itself. No plastic-lined items had any detected PFAS, which is likely due to plastics' inherent hydrophobicity. Items such as straws likely do not require additional PFAS as they pose minimal risk to the consumer upon failure. Additionally, some straws were found to contain external polymeric coatings likely to increase their water resistance (Fig. 11).

The maximum detected total PFAS concentrations increased by several orders of magnitude post oxidation of the material in basified methanol (Fig. 4). In addition to increased detection magnitude, high PFAS concentrations were also detected in samples that had no detections using the standard methodology, indicating that the PFAS species used in the manufacturing of these products is not a species that is examined within our testing suite of 38 species (Table SI 1). The total oxidisable precursor (TOP) assay results are not representative of the total sum of all PFAS species present, they are instead reflective of a portion of present PFAS species that have the potential to be oxidised into products present within the testing suite examined.<sup>43</sup> As such the sum of TOP assay results is likely an underestimation of total PFAS content present within a sample.<sup>43</sup>

Additionally, for the majority of samples (32 of 44) the LOR was an order of magnitude higher post TOP assay than the standard methodology. This led to samples such as the cardboard sample clamshells and trays-8 having detected PFOA at  $8 \mu\text{g kg}^{-1}$  pre-TOP assay but no PFOA detection post TOP assay (TOPA PFOA LOR  $10 \mu\text{g kg}^{-1}$ ). The lower limits of reporting in the 12 samples had detections of less than  $10 \mu\text{g kg}^{-1}$  in 2 of 2 cardboard samples, 2 of 4 paper samples and 1 of 1 wood

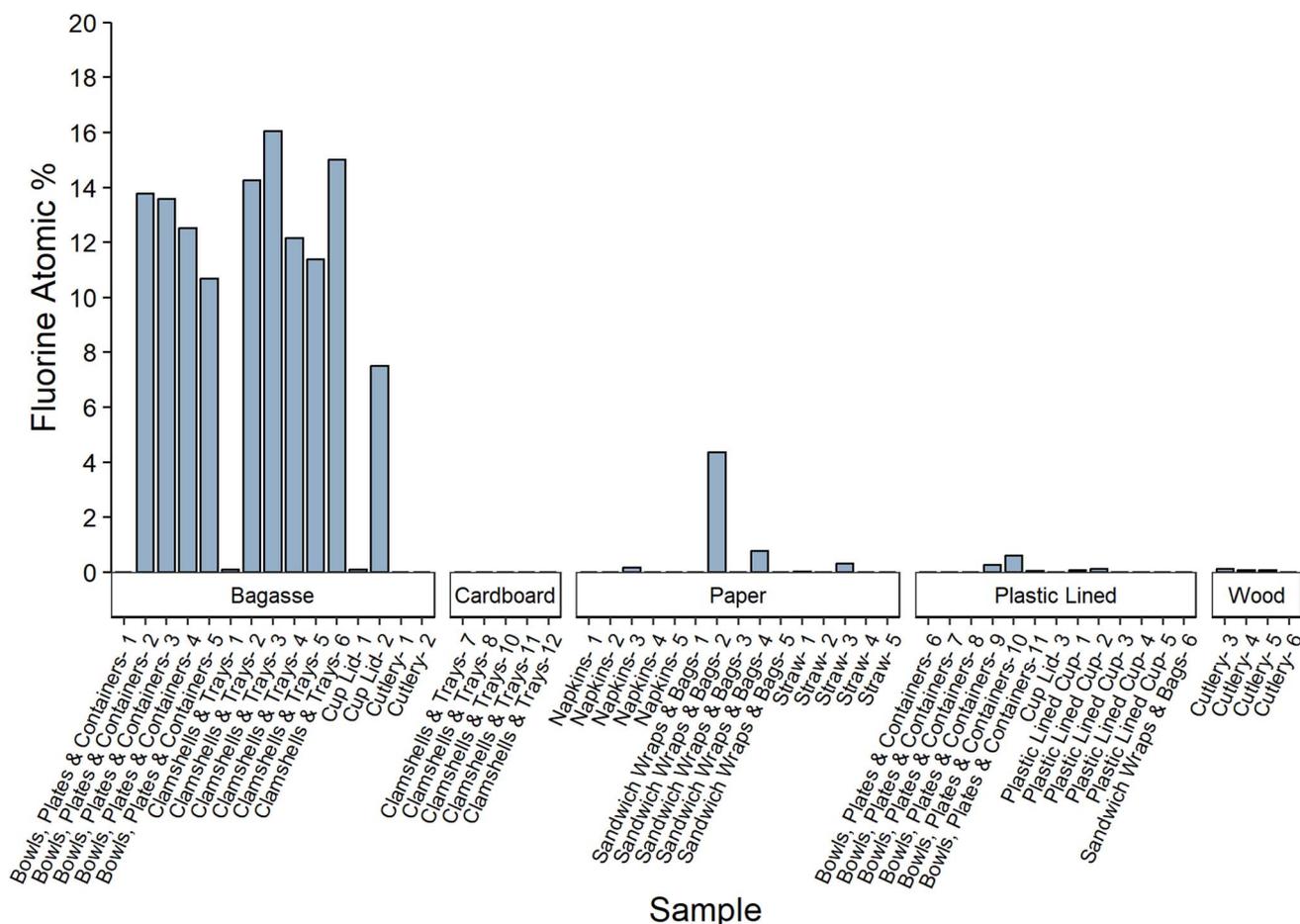


Fig. 5 XPS determination of relative fluorine content at each sample's food contact interface. Bagasse samples had higher total fluorine content at the surface than all other material types. Unsurprisingly, samples that exhibited higher PFAS concentrations in the bulk material also exhibited elevated fluorine concentrations at the food contact surface. Surface fluorine content in samples with no PFAS detection were all below 0.8%.



samples. These low concentrations of PFAS species are likely to be present in more paper and cardboard samples than is reflected in the current analysis due to the higher LORs. The source of the PFAS is likely from the incorporation of PFAS contaminated recycled content into the items. For instance, no PFAS was detected in any of the 10 straws or napkins examined, which are typically made from virgin paper. Whereas PFAS was detected at low concentrations in 3 of 5 cardboard clamshells and trays and 2 of 4 sandwich wraps and bags which are more likely to contain recycled content.

The detected composition of PFAS species was relatively consistent across all bagasse products indicating that the forms of PFAS added during the manufacturing process are likely to be similar across all items. On average  $55.1\% \pm 2.5$  SE of the total PFAS detected was perfluoropentanoic acid (PFPeA),  $22.1\% \pm 1.5$  SE was PFHxA,  $20.7\% \pm 0.8$  SE was perfluorobutanoic acid (PFBA) and  $0.03\% \pm 0.15$  SE was PFOA. The post TOPA PFAS analyte composition was vastly different in the sandwich wraps & bags-2 sample which contained 11 different breakdown products indicating the use of different or additional precursors in the manufacturing of the sandwich wrap. Future work using non-target analysis methods may help determine the exact PFAS compounds used in the manufacturing process of these items.

### Fluorine surface analysis

Analysis of the food contact surface of each item *via* XPS revealed the relative abundance of fluorine at the surface somewhat mirrored the concentration of that detected *via* the TOP assay sample pretreatment protocol (Fig. 5). All bagasse and paper samples with total concentrations of PFAS above  $2500 \mu\text{g kg}^{-1}$  had food surface contact areas with greater than 4% fluorine content. Fluorine surface concentrations within bagasse items did not mirror the relative amplitude of the concentrations of PFAS found within the bulk of the samples. For instance, the sample clamshells and trays-2 had the highest detected total PFAS concentration of  $86\,200 \mu\text{g kg}^{-1}$  however it had a surface fluorine content of 14.3%, whereas bowls plates & containers-3 had a total detected PFAS concentration of  $2850 \mu\text{g kg}^{-1}$ , but a surface fluorine content of 13.6%. This disparity could indicate:

- Different methods of initial PFAS application such as a fine dispersion coating *vs.* mixing with the bulk material;
- Samples possess additional undetected PFAS species which are not detectable using the TOP assay methodology such as ultra-short chain PFAS;
- The presence of other fluorine-based compounds in some samples; and/or.
- Inhomogeneous distribution of the PFAS on the surface, combined with small XPS sampling spot size.

Samples that did not contain high concentrations of fluorine often contained proportionally higher concentrations of silicon, with the sample bowls plates and containers-1 having 2.8% relative silicon content (Fig. SI 10). Silicon-based precursors and polymers could have been used for surface hydrophobisation treatments and may be present in the form of either siloxanes, silicones, or silicates.<sup>44</sup>

### Metals analysis

Plant species used to make paper, cardboard, wooden and bagasse products contain some metal content that is reflective of the plant species capacity to uptake nutrients and the environment in which the plant is grown.<sup>45</sup> Products such as bagasse are typically made from sugar cane, whereas many of the napkins tested are derived from eucalypt, while wooden cutlery items are typically made from birch.<sup>46–48</sup> The source of other paper-based products is typically either softwoods used in the manufacturing of kraft paper in items such as straws, or recycled paper used to construct cardboard and other paper food service items with lower hydrophobicity requirements.<sup>49</sup> Analysis of metal content revealed higher average concentrations of many metals in cardboard items than any other category of item likely due to feedstock contaminants and additives used in the paper recycling process. Contamination within paper recycling feedstock likely incorporates heavy metals such as lead, nickel and tin within recycled paper-based products. Other elements such as boron are likely incorporated into recycled paper products due to the use of paper recycling additives.<sup>50</sup> Paper recyclers use various boron-based compounds used to assist in ink removal and paper bleaching processes.<sup>51,52</sup> Additionally, some cardboard samples contained low concentrations of PFAS species and bis(2-ethylhexyl) phthalate (DEHP), likely indicative of recycled feedstock contamination (Fig. SI 1). No detections above the limit of reporting for silver, arsenic, beryllium, cadmium, mercury, molybdenum or selenium were found in any food service items (Fig. 6).

When examining the average copper concentrations of paper and plastic-lined materials, printed items ( $n = 5$ ) had average

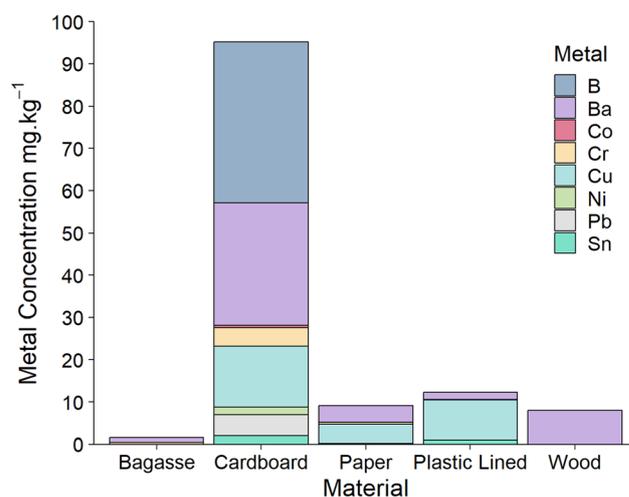


Fig. 6 Detection of average metal content in different material types. Elements including barium, silicon, zinc and manganese were excluded from this graph as they are likely to be present in the different raw feedstock materials. The higher presence of copper in paper and plastic-lined paper materials is likely reflective of the use of copper in the various inks and dyes used in the printing process. The overall increased concentrations of metals found in cardboard is likely due to input impurities and additives used during the paper recycling process. For instance, the elevated average concentration of copper may be due to the processing of printed materials, whereas the elevated concentration of boron is likely due to paper recycling additives.



concentrations of  $22 \text{ mg kg}^{-1}$  whereas unprinted materials ( $n = 18$ ) had average concentrations of  $1.8 \text{ mg kg}^{-1}$  (Fig. SI 2). Metals with high levels of biomass uptake such as zinc and manganese varied likely depending on the plant species the material was derived from. For instance, concentrations of manganese were proportionally higher in wooden cutlery and eucalypt-derived napkins than in other paper and bagasse materials (Fig. SI 15). While bagasse products possessed proportionally more zinc and less manganese than birch-based wooden cutlery (Fig. SI 20).

### Food contact surface material qualities

The examination of the oxygen : carbon (O : C) ratio at the food surface contact interface indicates the kinds of materials used as coatings to impart required material properties. For instance, vessels designed to hold liquids such as coffee cups will often use internal plastic coatings for hydrophobicity and paper outer materials for their insulative properties. Theoretically pure cellulose possesses an O : C ratio of 0.83.<sup>53</sup> However, as paper-based materials are more heterogenous and often functionalised for performance, the O : C ratio of all paper and cardboard samples was lower than the 0.65 (Fig. 7). Plastic-lined materials

possessed different O : C ratios likely due to the different stoichiometries of the various polymer types utilised, with materials such as raw PLA typically possessing O : C ratios of around 0.5, while more conventional hydrocarbons such as PE possess very low O : C ratios as the pure polymer does not contain any oxygen.<sup>54</sup> Different application methods, functionalisation, treatment processes and the incorporation of plasticisers are likely to impact the plastic coatings O : C ratio making them deviate from their theoretical stoichiometry.<sup>55</sup>

The use of PFAS in bagasse samples did not appear to inherently alter the O : C ratio of many samples as the two PFAS-free bagasse cutlery samples exhibited consistent O : C ratios to the majority of high PFAS samples. However, the three bagasse samples with the lowest O : C ratios of 0.08, 0.26 and 0.32 all possessed low PFAS concentrations. These three samples are represented by a clamshell, a bowl and a cup lid which as vessels all have higher water resistance requirements than the previously mentioned cutlery items. The discrepancy in these O : C ratios and intended use requirements of the items suggest the use of either carbon rich additives or a hydrophobisation treatment that reduces surface oxygen content.

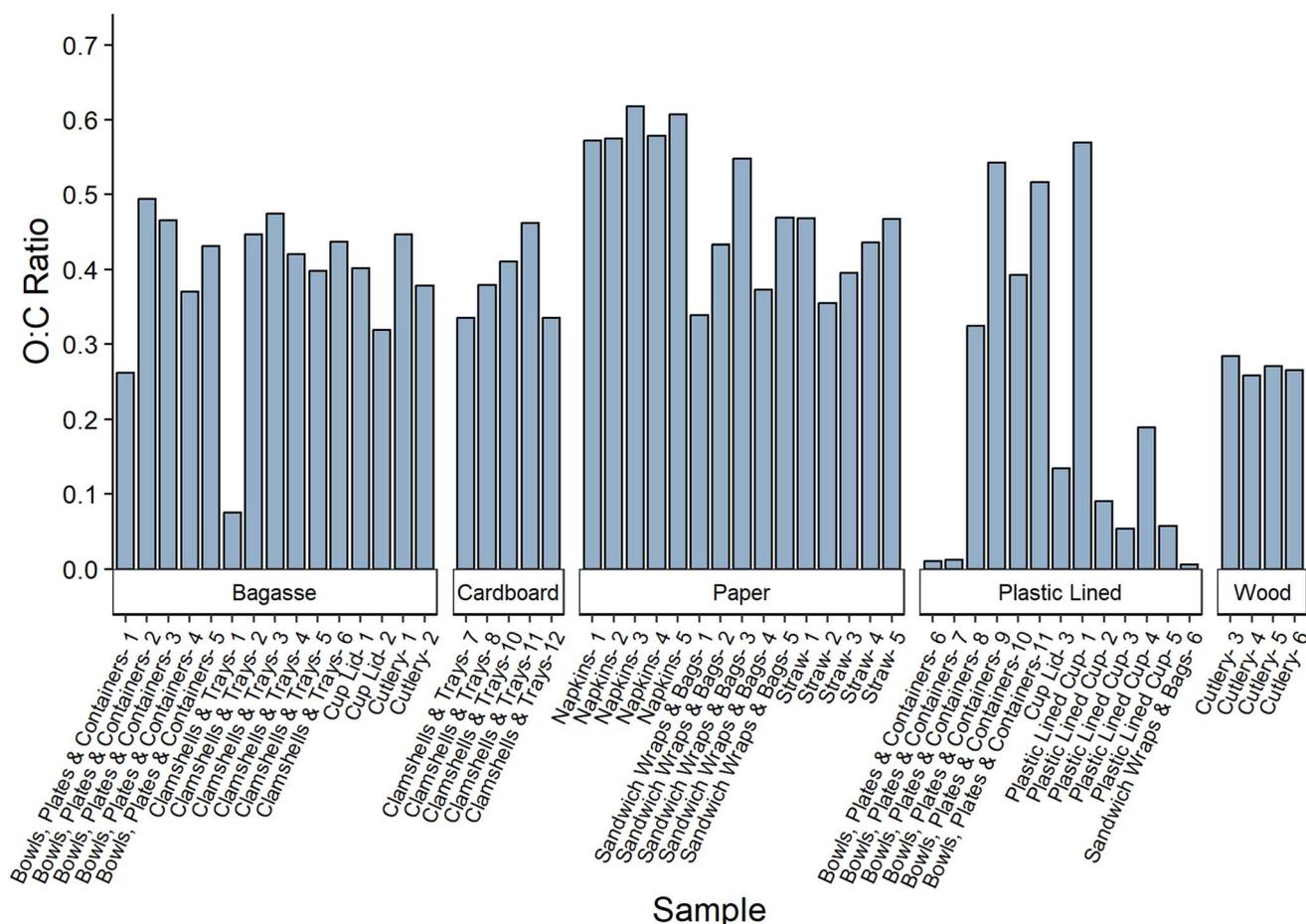


Fig. 7 XPS O : C ratio of food service items measured at the food contact interface. Many item types possessed relatively consistent O : C ratios with all napkins possessing a O : C ratio of between 0.57 and 0.52, while wooden cutlery had O : C ratios averaged an O : C ratio of 0.27. Most bagasse samples had O : C ratios between 0.4 and 0.5, however two samples possessed significantly lower O : C ratios of 0.07 and 0.26 indicating likely different surface chemical characteristics. Plastic-lined materials possessed an almost bi-modal set of O : C ratios indicating the likely use of different plastics to line the materials surface.



### Analysis of plastic coatings

Further examination of the surface material characteristics of plastic-lined items *via* FTIR and Raman spectroscopy supported the XPS O : C ratio data. Most of the 13 plastic-lined items examined were identified as possessing either PLA or PE coatings (Fig. 8). Other plastic types were found in bowls, plates and containers-6 which used a polypropylene coating and in plastic-lined cup-4 which used a styrenated acrylic coating.

Consumers are not able to determine the different plastic materials used in lined vessels which may lead to incorrect disposal. This uncertainty is compounded by marketing materials advertising the compostability of polyethylene-lined vessels. Additionally, prior to characterisation, the plastic-lined sandwich wrap and cup lid samples were thought to be paper and cardboard products respectively. When this perception is compounded by the mismatching of conventional plastic lids to compostable vessels (9/16 samples), it appears highly unlikely that an ordinary consumer will be able to determine the correct disposal pathway for their item.

Raman and FTIR testing was performed on the food contact surface of all samples and determined that paper, cardboard, and bagasse samples were largely derived from materials containing cellulose and lignin (Fig. SI 21–51). Minor variations across item types were observed likely indicative of the use of different additives and treatments applied to the items surface. For instance, clamshells & trays-1 exhibited significant fluorescence using the 638 nm laser and displayed an extra peak at  $506\text{ cm}^{-1}$  with the 786 nm laser (Fig. 9). Combined with the samples disproportionately low O : C ratio of 0.08 it is indicative of a fine additional coating at the samples surface.

Other samples such as cutlery-5 was found to have additional FTIR peaks at  $2917$  and  $2848\text{ cm}^{-1}$  which could indicate the use of an additional oil, wax or polyethylene coating (Fig. 10).<sup>56</sup> Manufacturers of these food service items likely use these

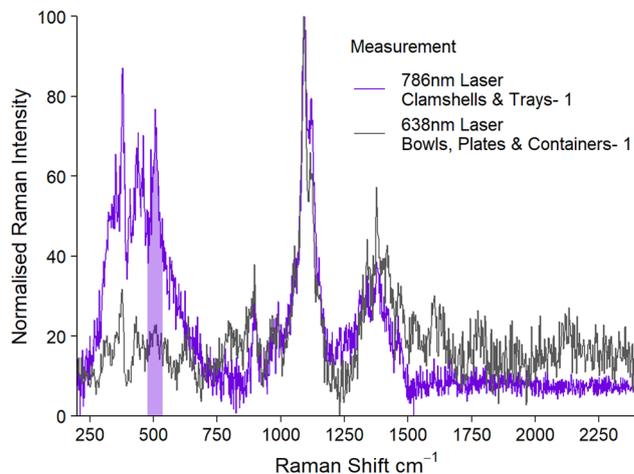


Fig. 9 Normalised Raman spectra of bagasse samples clamshells & trays-1 and bowls, plates & containers-1 exhibited a significant fluorescence using the 638 nm laser and possessed an additional peak at  $506\text{ cm}^{-1}$  when using the 786 nm laser. This combined with the low O : C ratio is indicative of an additional, potentially nanoscale coating on clamshells & trays-1 surface that is not present on other bagasse samples.

coatings to enhance the mouthfeel of otherwise unpleasantly textured products or to increase their hydrophobicity.<sup>57</sup>

### The use of inks dyes and adhesives

Many paper food service items requiring a rigid structure such as paper straws are comprised of several layers of material. Raman analysis of the material between the layers of the straw samples (straw 1, 2, 3 and 4) revealed the likely use of non-uniform adhesive coatings of conventional plastic polymers, including polystyrene and polystyrene/acrylate composites in two of the four samples examined. The sporadic detection of

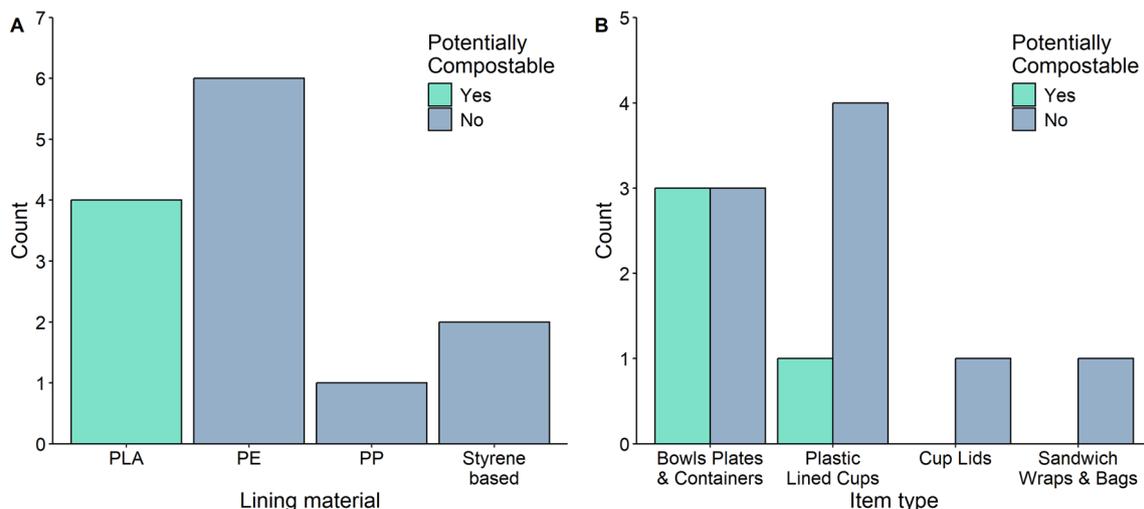


Fig. 8 (A) Plastic polymers used as barrier layers in plastic-lined samples ( $n = 12$ ). The majority of plastic linings and coatings were made from non-compostable materials with only 4/13 of samples utilising plastic types that are potentially compostable. (B) Half the plastic-lined bowls, plates & containers examined used compostable PLA linings (3/6), and only one examined plastic-lined cup used a compostable lining material. In contrast, all other plastic-lined items used non-compostable liners.





## Acknowledgements

Research funding for this project was provided by the NSW EPA. Analytical support was provided by the Nano and Microplastic Research Consortium at Flinders University and Envirolab Services Sydney. The authors acknowledge the financial support of the Australian Research Council (FT200100301), as well as the facilities, instruments, and scientific and technical assistance provided by Australian National Fabrication Facility (ANFF-SA, South Australian node) at Flinders Microscopy and Microanalysis, Flinders University, and Microscopy Australia at the Future Industries Institute, University of South Australia, enabled by NCRIS, and the South Australian state government. Additional support in the collection and analysis of samples was provided by Sofia Prifti, Rhea Lawani, Gil Bloxham, Sam Bannon, Ella Hadzionannou and Alva Lane.

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