



Plastics for dinner: Store-bought seafood, but not wild-caught from the Great Barrier Reef, as a source of microplastics to human consumers

Amanda L. Dawson^{a,b,*}, Joan Y.Q. Li^{b,c}, Frederieke J. Kroon^{a,c}

^a Australian Institute of Marine Science (AIMS), Townsville, Qld, 4810, Australia

^b AIMS@JCU, Division of Research and Innovation, James Cook University, Townsville, Queensland, 4811, Australia

^c College of Science and Engineering, James Cook University Townsville, Bebegu Yumba Campus, Townsville, QLD, 4811, Australia

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ABSTRACT

Seafood accounts for more than 17% of the global consumption of animal protein, with an excess of 335000 t consumed in Australia throughout 2019-2020. Recently, the presence of microplastics (MPs) within commercial seafood and the potential vectorisation of MPs to human consumers has become a significant concern for the public and the scientific community. Here, four commonly harvested wild-caught marine organisms were assessed for MP presence. These species comprise a significant proportion of the Queensland seafood industry, as well as being highly desirable to Australian consumers. The edible muscle tissue and discarded digestive tissue (GIT) of barramundi (*Lates calcifer*), coral trout (*Plectropomus leopardus*), blue leg king prawns (*Melicertus lat-isulcatus*), and Ballot's saucer scallops (*Ylistrum balloti*), were analysed discretely to determine the extent to which these species may be contaminated in the wild (GIT tissue), and the extent to which they themselves may act as a vector for human exposure (edible muscle tissue). Wild-caught seafood was predominantly free of MPs, with digestive tissues from two of ten coral trout containing only two fibres each. All wild-caught muscle tissue samples were free of MPs, as was the GIT of scallops, prawns, and barramundi. On the other hand, fresh, skinless barramundi muscle tissues, purchased from various commercial suppliers, were examined and found to be significantly contaminated with MPs (0.02 - 0.19 MP g⁻¹). Overall, these results highlight the growing consensus that food can become contaminated simply by being prepared in the human environment, and the focus must shift to determining the extent of MP proliferation within the processing and point-of-sale environment.

Introduction

Global seafood consumption currently exceeds 20 kg per capita, with more than 3.3 billion people relying on seafood for the majority of their animal protein intake (FAO, 2020). In Australia, 335000 t of seafood was consumed in 2019-2020, with finfish and crustaceans comprising a significant proportion (Steven et al., 2021). Indeed, the consumption of seafood is recommended by National Guidelines for health benefits (Government, 2013). With seafood playing a vital role as a protein source in Australia and across the world, it is imperative that seafood sold for human consumption does not pose a risk for human health.

However, much of the world's seafood has been reported to be contaminated with environmental pollutants (FRDC, 2018; Marquès et al., 2021) and consequently, can become a source of exposure for human consumers (Bank et al., 2020). These can include legacy and established pollutants, such as metals and methylmercury (e.g. Lavoie

et al., 2018), polychlorinated dibenzo-p-dioxins, furans and polychlorinated biphenyls (e.g. Huang et al., 2020), and polybrominated diphenyl ethers (e.g. Bedi et al., 2020). More recent global contaminants include pharmaceutical care products (e.g. Mello et al., 2022; Miossec et al., 2020), per- and polyfluoroalkyl substances (e.g. Fair et al., 2019) and microplastics (MPs, plastic particles 0.1-5000 µm) (e.g. Ribeiro et al., 2020). Unlike most environmental contaminants, MPs are typically isolated from seafood tissues that are generally discarded, rather than consumed by humans (i.e. the gastrointestinal tract, GIT, and digestive organs) (Dawson et al., 2021). However, there are still concerns for the propensity of seafood to act as a vector for MPs to human consumers and subsequent effects on human health (Vázquez-Rowe et al., 2021).

Numerous recent studies have quantified MPs present in commercial marine seafood organisms, from benthic filter feeders such as oysters (e.g. Zhu et al., 2021) and mussels (e.g. Nalbone et al., 2021), to large

* Corresponding author at: CSIRO Agriculture and Food, 306 Carmody Rd, St Lucia, QLD 4067, Australia

E-mail address: amanda.dawson@alumni.griffithuni.edu.au (A.L. Dawson).

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pelagic roaming predatory fish (e.g. Andreas et al., 2021). However, when determining the risk seafood poses to human consumers, is necessary to analyse the tissue that is actually consumed (Rist et al., 2018). Whereas molluscs, such as oysters and mussels, are shucked and consumed whole, more often the digestive organs of seafood are discarded in preference to the muscle tissue (Dawson et al., 2021). Within Australia, fish, and particularly those at a higher trophic level, are the most consumed seafood, followed by crustaceans and molluscs. The most common method to prepare fish for human consumption is to fillet the skinless muscle tissue into 'plate-size' portions. However, very few Australians catch their own seafood, with 97% purchased from stores, prepared fish fillets being a favourite choice (Intuitive Solutions, 2016, 2019; Steven et al., 2021; Steven et al., 2020). Therefore, to accurately ascertain the risk of seafood to human consumers, wild-caught seafood and prepared commercially available seafood should both be analysed, the latter likely to present a greater risk due to the multiple contamination sources it may be exposed to (Rist et al., 2018). For example, MPs are extremely common within indoor environments (Ageel et al., 2022; Soltani et al., 2021), and are liberated with high movement and friction (De Falco et al., 2020; Gwinnett and Miller, 2021; Rist et al., 2018; Scopetani et al., 2020). Fibres can also be aerosolised in high movement environments (Moore et al., 1986; Sheridan et al., 2020). Thus, there is capacity for food preparation areas to be localised MP hotspots. Despite this, there have been no studies in the literature thus far quantifying the MP contamination present in prepared store purchased skinless muscle fillets.

In this study, MP contamination was assessed in both the edible (muscle) and inedible (GIT) tissues of four commercial seafood organisms commonly caught and consumed in Australia. These were Common Coral Trout (*Plectropomus leopardus*), Barramundi (*Lates calcifer*), Blue Legged King Prawns (*Melicertus latisulcatus*), and Ballots Saucer Scallop (*Ylistrum balloti*), which comprise economically significant commercial fisheries species (Steven et al., 2021). In addition to the wild-caught organisms, filleted barramundi muscle tissues, purchased from local seafood suppliers, were also analysed for MPs, to determine whether MP contamination is introduced through the consumer supply chain.

Methods

Reagents and consumables

The materials used in this study are outlined in the Supplementary material.

Sample description

Wild organisms

Ballot's Saucer Scallop (*Ylistrum balloti*), Blue legged King Prawn (*Melicertus latisulcatus*), and Common Coral Trout (*Plectropomus leopardus*) were obtained from commercial fishers working in the Great Barrier Reef Marine Park, Queensland, (Australia) in March 2019 (Figure S1)(Table 1). Scallops were trawled in 57 m depth in the northwest of Fore and Aft Reef. Blue Legged King Prawns were trawled southeast of John Brewer Reef. Common Coral Trout were caught at 28-83 m depth using a hand line at Yamacutta Reef, Publican Shoals, Duncan Reef, Howie Reef, Pith Reef, and Nathan Reef. Barramundi (*Lates calcifer*) were obtained from commercial fishers and caught using gill nets at 0.5 m depth in Bowling Green Bay in March 2019. Organisms were immediately euthanized and frozen, either on board the vessel for trawled organisms, or immediately upon landing for fished organisms. Fish were euthanised using brain spike by the commercial fishers who supplied the samples. Scallops were sealed shut with an elastic band prior to freezing to ensure they remained closed when frozen. Organisms were frozen in low density polyethylene (LDPE) plastic bags and transferred the laboratory for analysis.

Table 1

Sample description of the analysed seafood organisms.

Organism	Tissue	Sample size	Origin
Ballot's Saucer Scallop	Gastrointestinal tract	10	Wild
	Muscle	10	Wild
Blue legged King Prawn	Gastrointestinal tract	5	Wild
	Muscle	5	Wild
Common Coral Trout	Gastrointestinal tract	10	Wild
	Muscle	10	Wild
Barramundi	Gastrointestinal tract	10	Wild
	Muscle	10	Wild
	Muscle	3	Purchased

Purchased fish

Skinless muscle tissues (i.e. fillets) from wild-caught barramundi were purchased from three seafood shops in Townsville, Queensland (Shops A, B and C). The wild-caught origin of the fillets was verbally confirmed by employees at all three shops. Prior to purchase, fillets at the three shops were stored unpackaged in glass refrigerated display bars, typically used for delicatessen and cold fresh foods in Australia. They include a sealed glass front panel and are accessed from the rear by shop employees using a glass sliding door. Store-bought fillets were placed in a transparent low-density polyethylene (LDPE) plastic bag and wrapped in butcher's (kraft) paper by a store employee. Fillets were frozen inside the LDPE bag and butcher's paper immediately after purchase and transferred to the laboratory for analysis.

QAQC

QAQC methods used to prevent background MP contamination of samples throughout analysis are outlined in the Supplementary material.

Microplastic extraction

Sample description

Whole organisms were defrosted, weighed (wet weight, w.w, 0.1 g), measured (Total length, TL, 0.1 cm) and flushed with reverse osmosis, (RO) water (H₂O) before being transferred to a Class II Biosafety Cabinet for dissection.

Scallop shells (TL 8.7-9.8 cm) (n=10) were flushed with ultrapure H₂O to remove any surface contamination. The shell was opened, and the scallop body also flushed with ultrapure H₂O to remove any exogenous contamination. The edible adductor muscle was separated from the GIT, gills, mantle, and reproductive organs (inedible tissue). Both edible and inedible tissues were flushed with ultrapure H₂O and placed in separate pre-weighed glass vessels for chemical digestion.

Prawns (TL 15.8 – 18.0 cm) (n=5) were flushed with ultrapure H₂O, after which the exoskeleton was carefully dissected, and the abdominal section was removed without rupturing the connection between intestine and digestive gland. The edible tail muscle was halved, flushed with ultrapure H₂O, and the upper half, closest to the cephalothorax (~7-12 g), placed in 1 L Schott bottles for chemical digestion. The digestive tissue was flushed with ultrapure H₂O and placed in pre-weighed glass tubes for chemical digestion.

Whole fish (coral trout (n=10) TL 41.4-58.9 cm; barramundi (n=10) TL 61.5-71.6 cm) were filleted, and the viscera (oesophagus to anus) removed. The edible muscle tissue (i.e. fillets) was skinned, subsampled (~150-200 g), flushed with ultrapure H₂O and placed in pre-weighed glass vessels for chemical digestion. Coral trout and barramundi

stomach (mean: 22.2 g and 16.1 g, respectively) and intestine (49.5 g and 29.2 g, respectively) were excised from the viscera, flushed with ultrapure H₂O and placed in separate glass vessels for digestion.

Purchased barramundi fillets (n=3) were defrosted inside their original LDPE plastic bags, inside a biosafety cabinet. Fillets were cut into ~150 g portions, but as these samples were used to test the hypothesis that seafood is contaminated along the supply chain, unlike wild-caught samples, were not flushed with ultrapure H₂O, before being placed directly into glass vessels for chemical digestion.

Chemical digestion

Fish and scallop muscle and GIT were digested in 10% potassium hydroxide (KOH) w:v for 14 days at room temperature (~22°C) (Dawson et al., 2020). Fillets from shop A, and several fish GIT samples were saponified upon exposure to 10% KOH, thus were treated with 100% ethanol (EtOH) to enable efficient filtration (Dawson et al., 2020).

Prawn muscle and digestive tissues were digested using a 10% hydrogen peroxide (H₂O₂) microwave assisted digestion according to (Li et al., 2022). Tissue was submerged in 10% H₂O₂ at a ratio of 25:1 v:w for 30 min (muscle tissue) and 10 min (digestive tissue) after boiling. Samples were heated to the boiling state inside a commercial domestic microwave oven (LG Microwave Oven MS4042GR, 1100 W) at half power (i.e., 550 W on average). Samples were closely monitored throughout digestion, with the heating briefly paused periodically to avoid H₂O₂ overflow. The glass digestion tubes and Schott bottles were loosely covered with a small glass beaker to prevent build-up of excessive pressure.

Fish muscle, and all scallop and prawn samples were then filtered over pre-cleaned stacked 263 µm, 77 µm and 26 µm stainless steel meshes. Filter cleaning procedure is outlined in Supplementary material. GIT tissues from the wild fish contained significant particulate matter. Thus, these samples were first filtered over a 525 µm stainless steel mesh to remove whole crustaceans, bones, and shells (Figure S2). Retentate was collected on a 26 µm stainless steel mesh, then back-washed into a hypersaline potassium iodide (KI) solution (1.69 g cm⁻¹) (Santana et al., 2022). The solution was density separated in a glass separation funnel overnight (~18 h) at room temperature. The dense settled material was discarded, the supernatant resuspended in KI for 1 h and the density separation process repeated. The resulting supernatant was again filtered over 263 µm and 77 µm stainless steel filters. Filters were then placed in aluminium filter holders and covered with a glass microscope slide, sealed with parafilm and left in a desiccating cabinet to dry.

Microscopic and Fourier transform infrared spectroscopic analysis of putative MPs

Filter retentate was microscopically searched for putative MPs, as outlined in the Supplementary material. All putative MPs were photographed and analysed using attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) or microimaging ATR-FTIR (µFTIR) as outlined in the Supplementary material. Due to the abundance of cotton and cellulose-based clothing worn in the laboratory environment (e.g., laboratory users clothing and cotton laboratory coats), in wild-caught samples, MPs comprising of cotton, cellulose or modified cellulose (e.g., rayon) were assumed to originate from the laboratory analysis (Gwinnett and Miller, 2021; Moore et al., 1986; Rist et al., 2018; Scopetani et al., 2020) and thus were not quantified. For store-bought samples, cellulose-based particles were quantified in samples where the concentration exceeded the background contamination, these were analysed separately to synthetic MPs (see below). Polytetrafluoroethylene (PTFE) and polybutylene terephthalate (PBTP) were also excluded due to their frequent use within the laboratory environment (i.e., wash bottles and Schott bottle lids), respectively. Spectra of all putative MPs were compared against a project specific

contamination library (adapted from Kroon et al., 2018) to check for known laboratory contaminants.

Physical characteristics of each confirmed synthetic particle were described in terms of shape (fibre or fragment), size and colour from the microscopic photographs. Fibres were defined as elongated particles with a relatively homogenous width throughout, and distinctly parallel sides. All other items were defined as fragments. Fibre length and width was determined using FIJI (Schindelin et al., 2012), by measuring the midline of the fibre using the segmented line tool (length), and averaging the width measured at three random locations along the fibre length (width). The length (Ferret's diameter) of fragments was measured using FIJI polygon tool, by tracing the perimeter of each item. Colour was defined by eye. White and transparent particles were grouped due to the high magnification used when taking the photo which obscured the exact colour.

Controls

During dissection, glass containers of ultrapure H₂O were placed in the four corners of the workspace within the biosafety cabinet, to account for airborne fallout of contaminants (blanks). These were processed alongside samples (i.e. filtration, microscopic evaluation and FTIR). Negative controls were prepared in triplicate alongside each sample batch, using a subsample of digestion solution (i.e. 10% KOH, 10% H₂O₂, EtOH). These were processed alongside samples, following the same workflow: digestion, filtration, density separation, microscopic evaluation and FTIR. Positive controls were prepared in triplicate alongside each sample batch using a small sample of muscle tissue (fish, crustacean or scallop), spiked with 5 pieces of polyethylene terephthalate (PET) fibre (1-5 mm), 5 fragments of crystalline polystyrene (PS)(1-2 mm), and 5 fragments of polyvinyl chloride (PVC) (1-2 mm). These were processed alongside samples, following the same workflow: digestion, filtration, density separation, and microscopic evaluation. Putative MPs identified in the blanks and negative controls were analysed using FTIR, as outlined below.

Data analysis

Spike recovery was used to calculate confidence intervals for the data. MP data was blank corrected using the procedural controls and blanks. MPs isolated from the controls and blanks were spectrally characterised and compared to all MPs isolated from the seafood samples. MPs isolated from samples and controls were compared to the contamination library. If a particle matched spectrally with a >80% correlation and matched visually to a potential contaminant, the particle was removed from the dataset (adapted from Kroon et al., 2018). The remaining MPs isolated from the procedural controls and blanks which did not match with any particles from the samples, or the contaminant library, were used to calculate the Limits of Detection (LOD) and Limits of Quantification (LOQ).

LOD and LOQ were calculated separately for fibres and fragments in each sample. These were used to give reliable estimates at which the levels of MPs detected in the samples can be distinguished from laboratory contamination. LOD_{fibre} and LOD_{fragment} were calculated as mean + 3×SD. LOQ_{fibre} and LOQ_{fragment} were calculated as mean + 10×SD. Due to the prolific abundance of cellulose-based particles isolated from the store-bought fillets, as compared to the wild-caught organisms, in addition to the MPs isolated from the purchased fillets, a LOD and LOQ for cellulose-based items was also calculated. To account for the abundance of these items within the laboratory environment, the LOD_{cellulose} and LOQ_{cellulose} values were derived from the concentration of cellulose-based items isolated from the wild barramundi muscle samples. For all samples, for statistical purposes, values ≤LOQ were treated as zero.

MP abundance in seafood was calculated as MP g tissue⁻¹ (w.w) or MP sample⁻¹. Unpaired T-tests were used to explore the concentration of MPs extracted from the store-bought fillets and the wild fish. A general

linear model with Gaussian distribution was used to explore differences amongst particle sizes extracted from the fillets. Length data was log transformed to improve the goodness of fit of the model. Statistical analysis was carried out using GraphPad Prism 8.4.3 or R version 4.1.1 (packages glmmTMB, DHARMA and emmeans).

Results and discussion

Background Contamination and Blank Corrections

Recovery of spiked tissue samples ranged from 80% - 100% (Table 2). Despite the extensive procedures to prevent extraneous contamination of seafood samples, including working inside a clean air device, and using previously established QAQC criteria (Dawson et al., 2021), fibres and fragments were found in almost all procedural controls and blanks. Based on this sample contamination, the LOD, LOQ and mean MP concentrations isolated from scallops, prawns, and fish were estimated (Table 2). The wild coral trout and barramundi LOD was determined to be 0.54 MP sample⁻¹ for fibres and 0.76 MP sample⁻¹ for fragments. As these values were based on counts, and partial values are not possible, these values were rounded to 1 MP sample⁻¹.

The difficulty experienced in many laboratory environments, including this study, in establishing a MP free environment is concerning (Belontz and Corcoran, 2021; Gwinnett and Miller, 2021). This highlights the important of using representative blanks and controls, which accurately encapsulate potential entry points for contaminants. In this study, beakers with ultrapure water (blanks) contained a total of 33 items, whereas negative controls contained more than 3 times this amount (119 items). These results would suggest that carrying out MP analysis in trace level facilities would be beneficial (Flegal and Smith, 1995; Patterson, 1965).

Microplastics in Australian seafood

Microplastic detection frequency in all samples is presented in Table S1. Ballot's saucer scallop, blue legged king prawn, barramundi and common coral trout did not contain detectable MPs within the edible muscle tissue (Table 2). Prawn GIT were also MP free. Several MPs were isolated from scallop and barramundi digestive tissue; however, the concentration of isolated MPs was below the LOD. Thus, these

items were not reliably distinct from the concentration of MPs isolated from the controls and were considered to be laboratory contaminants. The GIT tissue of wild-caught coral trout was predominantly free of MPs, most samples contained 0 MP or were below the LOD (Table 2). However, two individuals did contain MPs above the LOD and LOQ. Both organisms contained 2 PET fibres each, which equated to 0.19 MP g⁻¹ stomach tissue and 0.14 MP g⁻¹ intestinal tissue. The 2 fibres extracted from the stomach were transparent (590.8 µm) and red (773.1 µm), whereas the 2 fibres extracted from the intestine were both blue, with similar lengths (436.6 and 382.9 µm), suggesting these 2 fibres may have both originated from the same source (Fig. 1b; Figure S3).

All three samples of purchased barramundi fillets contained measurable quantities of microplastics. Fibre concentration ranged from 0.02 - 0.17 MP g⁻¹. Fragments were only quantifiable in one fillet sample (0.10 MP g⁻¹). Cellulose-based particles outnumbered synthetic MPs in most samples, with concentrations ranging from 0.13 - 0.25 items g⁻¹. Through the use of LOD and LOQ values, the concentration of MPs isolated from purchased fillets was found to be significantly, and reliably, higher than laboratory background contamination, including background cellulose fibres (Table 2). Fibres were significantly more common than fragments (p<0.05), and the most common polymers included PET, and polyethylene (PE) along with cellulose-based fibres (Fig. 1a). Polymer and colour were reasonably consistent within each sample (Fig. 1a), but varied across the three stores, likely reflecting the variable environments the fillets were exposed to before purchase. The high number of transparent fibres was likely influenced by the bleaching effect KOH has on cellulose-based fibres (Dawson et al., 2020). Notably, a subset of the synthetic fibres extracted from the samples visually matched the colour of the uniform worn by employees working around the fillets. Although this was not confirmed using spectroscopic analysis, it is likely that many of the fibres extracted from the store-bought fillets originated from the clothing worn by employees (Gwinnett and Miller, 2021; Rist et al., 2018; Scopetani et al., 2020). Polyester and cellulose-based fibres are both common clothing materials, prone to shedding (De Falco et al., 2020; Sheridan et al., 2020) and dominate indoor environments (Dris et al., 2017). Fibre length also seemed to be related to the store environment, with length varying across all the three shops (Fig. 2a, Table S2). Fibre length also varied between synthetic and cellulose-based fibres (Table S3). The length of cellulose-based fibres was consistent across all three shop fillets (p>0.05), however, synthetic

Table 2

Microplastics (MPs) isolated from wild-caught and store-bought seafood samples. Spike recovery for individual polymers is presented in Table S3. NA = not applicable, LOD = Limits of Detection, LOQ = Limits of Quantification, ≤LOD = MPs were detected in concentrations less than or equal to the limit of detection in one or more samples, 0 = no samples contain MPs.

Origin	Seafood	Tissue	Average Spike Recovery (95% Confidence Interval)	Sample	Fibre		MP g ⁻¹ Mean	Fragment		MP g ⁻¹ Mean	Total Number of Items Detected
					MP sample ⁻¹ LOD	MP sample ⁻¹ LOQ		MP sample ⁻¹ LOD	MP sample ⁻¹ LOQ		
Wild	Scallop	Digestive Tract	95.56% (88.50-102.60)	1-10	1.28	3.89	≤LOD	2.92	7.97	≤LOD	-
		Muscle		1-10			0			≤LOD	-
Wild	Prawn	Digestive Tract	91.12% (77.76-104.66)	1-5	NA	NA	0	NA	NA	0	-
		Muscle		1-5	NA	NA	0	NA	NA	0	-
Wild	Coral Trout	Stomach	80.00% (59.66-100.34) 92.22% (81.90-102.54)	1-3,5-10	0.54	1.72	≤LOD	0.76	2.42	≤LOD	-
		Intestine		4			0.19			≤LOD	2
				1,3-10			≤LOD			≤LOD	-
				2			0.14			≤LOD	2
Wild	Barramundi	Muscle	96.67% (91.54-101.78)	1-10			≤LOD			≤LOD	-
		Stomach	94.44% (88.74-100.16)	1-10			≤LOD			≤LOD	-
		Intestine		1-10			≤LOD			≤LOD	-
		Muscle	93.33% (82.46-104.2)	1-10			0			0	-
Store	Barramundi	Muscle	100% (100-100)	1	0	0	0.17	1.87	5.30	≤LOQ	32
				2			0.02			≤LOD	3
				3			0.10			0.10	34
		Muscle-Cellulose	-	1	3.72	8.62	0.25	-	-	-	48
				2			0.13			-	26
				3			0.18			-	30

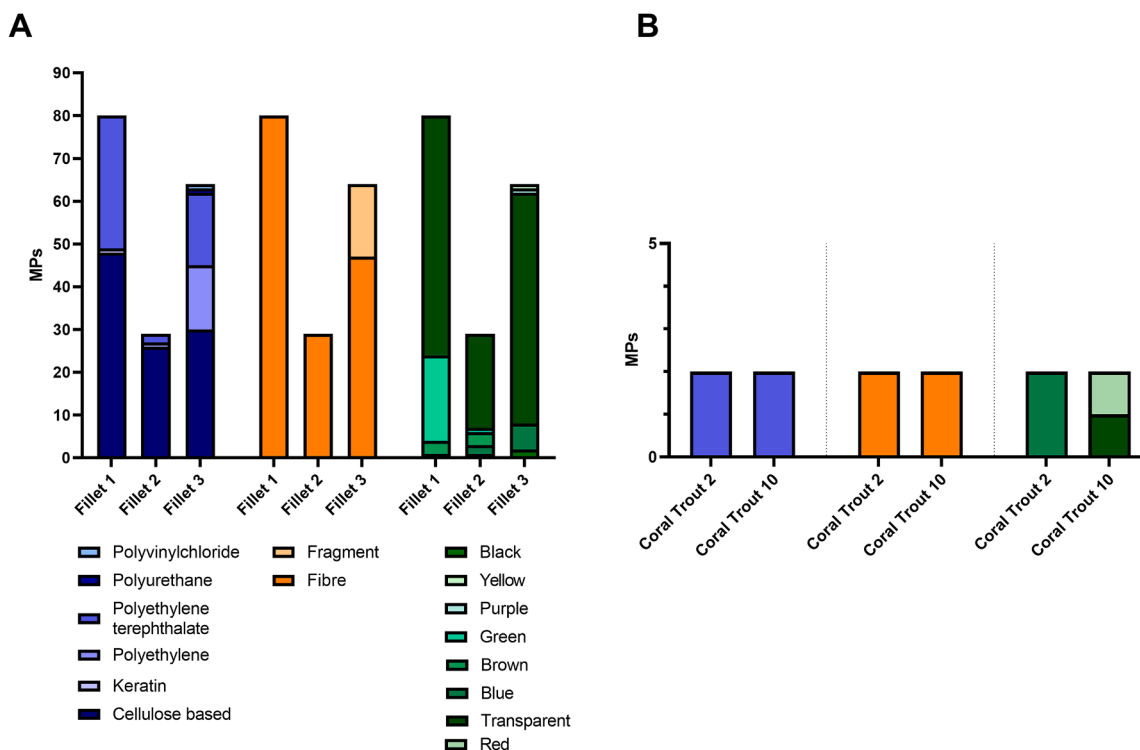


Fig. 1. Total polymer (blue), shape (orange) and colour (green) of microplastics and cellulosic items isolated from A) store-bought barramundi fillets and B) wild coral trout GIT.

fibres isolated from shop A fillets were significantly smaller than both the cellulose-based fibres within the same sample ($p < 0.05$), and synthetic fibres isolated from shop C fillets ($p < 0.01$).

In comparison to the wild-caught fish samples, the shop fillets contained significantly more MPs than both the wild fish muscle ($p < 0.0001$) and, unexpectedly, the wild fish GIT tissue samples ($p < 0.01$) (Fig. 3). MP were detected in 100% of the store-bought fillets, whereas MP were only detected in 2 of the 10 coral trout GIT samples. It is noteworthy that despite having numerous MPs present in the store-bought fillets, the weight-corrected means for store-bought fillets was similar to the MP concentration isolated from the two coral trout GIT samples which contained MP. The 2 fibres isolated from each coral trout GIT sample equated to 0.14 and 0.19 MP fibres g^{-1} . Whereas the store-bought fillets ranged from 32 fibres isolated from barramundi fillet 1, which equated to 0.17 MP fibres g^{-1} , to 3 fibres in barramundi fillet 2, which equated to 0.02 MP fibres g^{-1} . This observation is likely due to the different exposure mechanisms between the two sample types. As the wild barramundi muscle samples were free of MPs, it is logical to propose that the store-bought fillets were unlikely to have become contaminated while the fish was alive, becoming contaminated only after filleting and exposure to the human environment, as is common with other butchered meats (Habib et al., 2022). Thus, MPs were unlikely to be homogeneously distributed throughout the fillet tissue, but rather deposited on the surface from the shop environment, similar to previous studies on meat contaminated with MP originating from packaging (Kedzierski et al., 2020). Therefore, the weight-corrected means of the store-bought fillets are evidence of a dilution effect based on the larger sample weight (barramundi store-bought fillets: 186.15 ± 8.38 g and coral trout stomach and intestine: 22.2 ± 13.2 g and 16.1 ± 6.17 g, respectively). Surface area and exposure time (if known) may be useful metrics to express MP concentration in samples that are similarly exposed through deposition, rather than weight. This pathway is likely to be more relevant to food exposure rather than biological processes, such as ingestion.

A recent study found that cutting boards are a source of plastic

fragments in butchered meat (Habib et al., 2022). Shop C was the only fillet sample that contained fragments above the LOQ. Of these, 88% consisted of transparent/white PE fragments, which were visually and spectrally identical. Therefore, it is highly likely that all PE fragments within this sample originated from the same source, and it is possible they originated from cutting boards used in shop C to prepare the fillets. Habib et al. (2022) found that PE chopping boards contributed between 1.2 – 6.5 MP g^{-1} of meat. Although this value is higher than the abundance of PE found in the current study, the meat in the former study was cut into much smaller pieces, approx. 2.1 cm and 1.44 g. Therefore, these sample had a higher surface area to weight ratio, which may explain the increased abundance of PE fragments found (Habib et al., 2022).

Several recent studies have demonstrated MPs are not detectable in wild-caught fish muscle tissue when analysed in conjunction with blanks and controls. For example Su et al. (2019), Akoueson et al. (2020) and Rasta et al. (2021) all found no statistical difference between the MP concentration isolated from the fish muscle and the blanks. Although only at a single temporal sampling location, the findings here suggest that wild scallops, prawns and predatory fish, such as barramundi and coral trout, do not accumulate environmental microplastics $> 26 \mu m$ in their muscle tissue. It is currently unknown if nanoplastics and very small MPs translocate to muscle tissue in the wild. Although several previous studies have claimed to have isolated microplastics from muscle tissue, many of these have not used reliable or appropriate methods to extract and analyse microplastics (Dawson et al., 2021). In fact, it has been proposed that translocation of such large sized particles (μm to mm) from digestive organs to muscle tissue is biologically implausible (Jovanović et al., 2018; Kim et al., 2020; Schur et al., 2019; Zeytin et al., 2020). The lack of microplastics isolated from muscle tissue in all 4 wild-caught species tested here, adds weight to this hypothesis. Further, many studies do not spectrally identify every putative MP to confirm their synthetic nature (e.g. McIlwraith et al., 2021), and this can lead to inaccuracies when extrapolating from a subsample (Brandt et al., 2021). Some studies also include cellulosic fibres along with synthetic

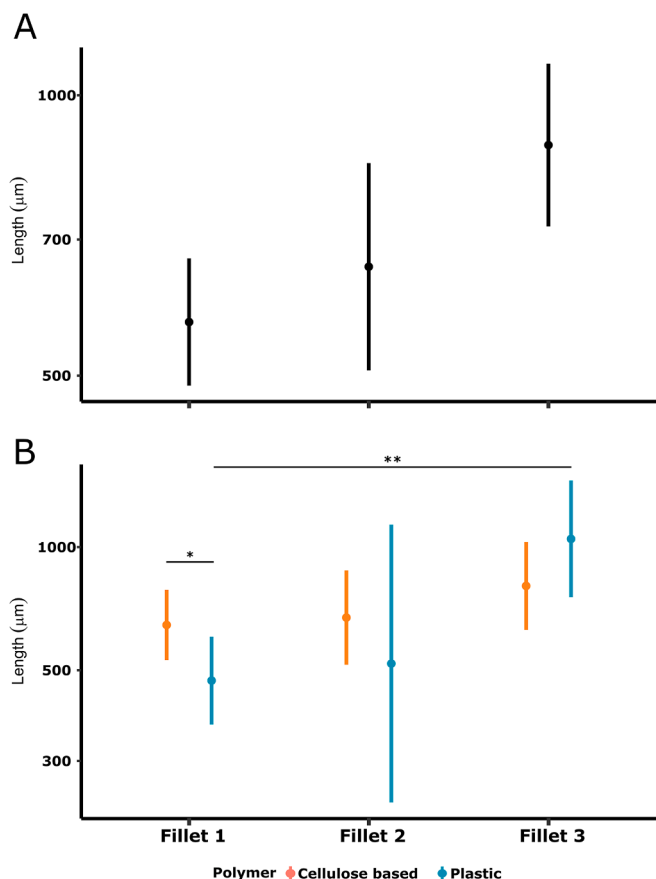


Fig. 2. Particle length of fibres isolated from the three store-bought fillets. A) All fibres combined, B) microplastic and cellulose-based fibres. Symbols indicate mean \pm 95% confidence limits. Lines denote significantly different comparisons using general linear model at the 0.05 level of significance.

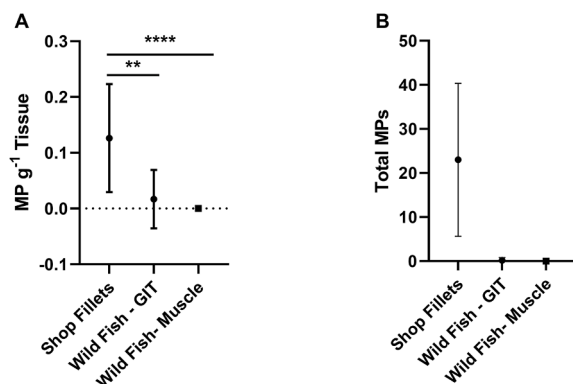


Fig. 3. Microplastic (MP) particles isolated from store-bought fillets and wild-caught fish. A) MP g⁻¹ (SD) tissue analysed B) Total number of MPs (SD) isolated from the samples. Cellulose-based items are not shown. Lines denote significantly different comparisons using unpaired T-tests at the 0.05 level of significance.

fibres when analysing MPs (e.g. McIlwraith et al., 2021). Due to the abundance of cotton and cellulose-based clothing and laboratory coats used in MP laboratories, there is a very high likelihood these textiles are shedding fibres into the workspace and contaminating samples (Athey et al., 2020; Gwinnett and Miller, 2021). This certainly was the case in the current study, where the LOQ_{cellulose} (8.62), for store-bought barramundi fillets was calculated based on the background contamination of cellulose isolated from the wild-caught barramundi muscle.

All four wild-caught species contained few or zero MPs in their GITs. This could be considered surprising for the filter feeding benthic scallops and detritivore benthic prawns. One possible explanation is that MPs are only present in trace levels in the GIT of these species. Sample analysis in this study was not carried out in a trace level laboratory facility, and background contamination may have masked any underlying MP contamination from the environment. This may have been the case for scallops, which had LOD_{fibre} of 1.28 and LOD_{fragment} of 2.9, highlighting a possible limitation of this study. This scenario is unlikely for coral trout and barramundi as they are both high trophic level ambush predators (Davis, 1985; St John, 1999), with recent research on trophic biomagnification finding MPs are significantly less abundant in such organisms (Walkinshaw et al., 2020). Coral trout, in particular, have been noted to rarely consume more than one prey item before evacuating their GIT (St John, 1999). If this is the case, then the probability of MP detection may be low, depending on the gut content of the single prey item consumed before capture. In terms of improving detection limits, conducting sample analysis within purpose-built trace level facilities would be ideal (ANZG, 2018), as has been done for other environmental contaminants which are frequently encountered in the human environment but of lower abundance in the natural environment (e.g. Flegal and Smith, 1995; Patterson, 1965; Patterson, 1976). Furthermore, methods that do not rely on visual assessment such as GC-MS, may allow for smaller MP and nanoplastics to be quantified in these matrices (Ribeiro et al., 2020).

Human consumption of microplastic contaminated seafood

These results add to the growing consciousness that food is frequently contaminated with MPs (Dessi et al., 2021; Fadare et al., 2020; Karami et al., 2018; Kedzierski et al., 2020). Rather than gradually biomagnifying through trophic levels, edible muscle tissue is more likely contaminated by simply being present in our human environment, through airborne fallout from clothes, packaging, processing and household dust (Catarino et al., 2018; Dris et al., 2017; Dris et al., 2016; Gasperi et al., 2018; Habib et al., 2022). This is a troubling hypothesis and requires significant further study to encapsulate this exposure route. A recent study of Australian seafood also proposed that plastics isolated from store-bought fish may have originated from the store environment and packing (Ribeiro et al., 2020). The authors hypothesised packaging may have been the primary source of PE isolated from sardine skin and fillets (Ribeiro et al., 2020). However, in the current study, packaging may have only contributed minor contamination, as the barramundi fillets were packaged in LDPE, whereas the most prevalent polymers across all three samples were cellulose-based and PET. This may indicate that contamination within the human environment is highly site specific, and thus human exposure to MP will vary considerably.

Conclusion

Overall, based on these results it seems that Australian seafood sourced directly from wild-caught organisms, which typically comprises of muscle tissue, presents little exposure risk to human consumers. In fact, the organisms assessed in this study also contained very little MPs within their GITs, suggesting one of three hypotheses a) that plastic pollution within the Great Barrier Reef may not be prolific, b) that these specific organisms are not susceptible to plastic ingestion, or c) the depuration of ingested plastics exceeds ingestion. Given that this study analysed MPs larger than 77µm, the abundance of smaller MPs and nanoplastics within these matrices remains unknown. Conversely, the prolific contamination of the purchased skinless fillets suggests that regardless of the lack of contamination of seafood in the marine environment, it becomes contaminated with MPs after entering the human environment, and prior to human consumption. Whether or not the consumption of contaminated seafood is more or less of a risk when compared to other sources, such as inhalation and food in general,

requires significant further study.

Author contribution

AD designed the study, collected the data, analysed samples, analysed data, and wrote the manuscript draft. JL collected data, and analysed samples, FK obtained funding and contributed to writing the manuscript.

Declaration of Competing Interest

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envadv.2022.100249. Data is available at <https://apps.aims.gov.au/metadata/view/2ea27f68-f65b-47ad-9a84-eb284170cbde>

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