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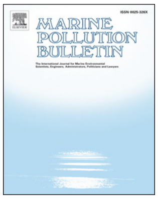

Diverse land uses and high coastal urbanisation do not always result in harmful environmental pollutants in fisheries species

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ABSTRACT

Human activities in coastal catchments can cause the accumulation of pollutants in seafood. We quantified the concentration of heavy metals, pesticides and PFASs in the flesh of the fisheries species yellowfin bream *Acanthopagrus australis* (n = 57) and mud crab *Scylla serrata* (n = 65) from 13 estuaries in southeast Queensland, Australia; a region with a variety of human land uses. Pollutants in yellowfin bream were best explained by the extent of intensive uses in the catchment. Pollutants in mud crabs were best explained by the extent of irrigated agriculture and water bodies. No samples contained detectable levels of pesticides, and only six samples contained low levels of PFASs. Metals were common in fish and crab flesh, but only mercury in yellowfin bream from the Mooloolah River breached Australian food safety standards. High pollutant presence and concentration is not the norm in seafood collected during routine surveys, even in estuaries with highly modified catchments.

1. Introduction

Ecosystems are increasingly impacted by human activities globally (Halpern et al., 2019). These impacts have consequences for the condition and functioning of coastal ecosystems, and the abundance and diversity of animals in coastal seascapes (Heery et al., 2017; Mouillot et al., 2013). Some impacts, like runoff from catchments that are modified by agricultural, urban or industrial developments can affect how safe seafood captured from coastal ecosystems is for human consumption due to potential exposure to harmful environmental pollutants (Bosch et al., 2016; Jian et al., 2017; Landos, 2013; Murray et al., 2010). For example, pesticides can runoff from agricultural lands, and harmful industrial compounds can runoff from urbanised and industrial lands. Heavy metals are naturally occurring elements that have broad importance to people through a variety of applications, but can accumulate to dangerous levels in ecosystems due to human activities (Castro-Gonzalez and Mendez-Armenta, 2008). This is of increasing concern to managers because catchment modifications and potential pollutant sources continue to increase and expand in prevalence and geographic scope globally (Halpern et al., 2019), as does the demand for wild-caught seafood (FAO, 2018). Consequently, quantifying the prevalence and concentrations of pollutants in the flesh of key seafood species, and identifying potential sources and drivers of these pollutants is a key focus for coastal managers (Budtz-Jorgensen et al., 2007; Carbery et al., 2018; Waltham et al., 2011).

Estuaries are an ideal study system to test for pollutants in seafood because they are subject to substantial runoff from catchments that vary significantly in land use and potential pollutant sources (Crain et al., 2008; Halpern et al., 2008). Pollutants that runoff from degraded and modified catchments concentrate in waters near the mouths of estuaries before being released into the ocean (Barletta et al., 2019; Warwick et al., 2018; Wen et al., 2017). These can bioaccumulate in seafood through a variety of pathways. Therefore, the exposure of seafood species to pollutants is likely contingent on their biology, ecology and behaviour (Liu et al., 2019; Russell et al., 1999; Suedel et al., 1994). For example, species that regularly feed in or on the benthos are potentially at a greater risk of consuming harmful concentrations of sediment-associated heavy metals (Chen and Chen, 1999; Schlacher et al., 2007). People can make assumptions about the health consequences of consuming some seafood based on species feeding behaviour or the extent

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of anthropogenic impacts around capture sites. Testing these assumptions using broad scans of the full suite of potential pollutants within individual systems is therefore important to ensure that economic, social and health benefits of catching and eating seafoods are not missed. Accurately quantifying the sources of pollutants of human health concern, and determining whether particular features of catchments, the marine environment and/or species biology and ecology increase the risk of pollutant bioaccumulation is an increasing focus for coastal managers.

There are numerous pollutants that can accumulate in seafood (Hellberg et al., 2012; Thomsen et al., 2018; Ye et al., 2018). Perfluorinated alkylated substances (PFASs) are emerging contaminants of international concern that are used for numerous domestic, industrial and agricultural purposes (Murray et al., 2010; Xiao, 2017). They have recently been subject to significant media attention globally due to their use in firefighting foams at airports, and their being released into nearby waterways and water tables (Food Standards Australia and New Zealand, 2018; Xiao, 2017). PFASs are highly resistant to environmental degradation and bioaccumulate in food chains (Lindstrom et al., 2011; Taylor and Johnson, 2016). Consequently, there are now several examples globally of PFAS accumulation in the flesh of coastal seafood species (e.g. Taylor and Johnson, 2016). There is increasing evidence of potential carcinogenic and immunological effects of PFASs in people, meaning that they are of increasing concern for human health (Grandjean and Clapp, 2015; Lau, 2015).

There is significant and increasing concern about the health implications of high concentrations of anthropogenically derived heavy metals for people (Tchounwou et al., 2012). For example, lead, cadmium, mercury and arsenic, have significant health consequences for people, and occur in high concentrations in many fish species (Jarup, 2003; Verdouw et al., 2010; Wang et al., 2005; Yi et al., 2011). Whilst heavy metals often occur naturally within the biosphere, their broad industrial, domestic, agricultural and technological applications mean that their concentrations and distribution in the biosphere can be modified by anthropogenic activities (Bosch et al., 2016; Tchounwou et al., 2012). Despite ongoing warnings regarding the health consequences of heavy metal consumption by people, exposure rates remain high in many human populations, especially those that regularly consume seafood (Bosch et al., 2016; Castro-Gonzalez and Mendez-Armenta, 2008).

Pesticides are persistent environmental pollutants of significant community concern. Pesticides are used broadly across landscapes as control agents for weeds, pests, and diseases, such that there are now no groups of people that remain unexposed to pesticides (Kim et al., 2017). Whilst the most widely reported use of these compounds in coastal catchments is in agricultural areas, they are also used in urban parklands and sporting fields for controlling weeds and pests, and are also present in common household items such as shampoo and building materials (Kim et al., 2017; Nicolopoulou-Stamati et al., 2016). There is evidence that exposure to certain pesticides causes both short term (e.g. skin irritation, dizziness; Fareed et al., 2012; Kim et al., 2017) and long term (e.g. cancer, diabetes; Bassil et al., 2007; Colette Sylvie et al., 2013; Kim et al., 2017) health effects in people. Combined, these three groups of environmental pollutants (PFASs, heavy metals, and pesticides) represent a variety of human health risks and effects (from short-term, non-lethal effects, to long term and potentially lethal effects), sources in coastal areas (from both point and non-point source releases), and potential bioaccumulation pathways, and so are the focal pollutants for this study.

Quantifying the extent and likely drivers of pollutant contamination in seafood is vital in understanding the effects of human impacts on coastal ecosystems, how these impacts might reduce the value of fisheries assets, and whether these sorts of impacts have potential implications for human health. In this study, we quantify the concentrations of a suite of PFASs, pesticides and heavy metals in seafood captured from estuaries in southeast Queensland, Australia, and seek to determine which attributes of land use in estuarine catchments correlate most with the suite of pollutants present in coastal seafood species. Estuaries in southeast Queensland, Australia are ideal to test for these effects because the region has a broad suite of human impacts, land uses and potential sources of pollutants in the catchment, and these catchments release into estuaries with a diversity of sizes and extent of marine vegetation (Gilby et al., 2017a; Olds et al., 2018; Schlacher et al., 2007). Despite earlier results showing compliant pollutant levels in fish in southeast Queensland estuaries (Waltham et al., 2011), there has been increasing concern among the public in this region about the health risks associated with consuming some coastal seafood species, especially after some recent high-profile spills of pollutants into estuaries. In this sense, there is a tendency of local people to assume that certain seafood species are ‘inedible’ based on assumed effects of surrounding land use. We chose two species from this study region that are important commercial and recreational targets for local fisheries, but also represent two fundamentally different lifecycles of coastal organisms that might result in different exposure to pollutants. Yellowfin bream *Acanthopagrus australis* are one of the most commonly targeted recreational finfish species in Queensland (Webley et al., 2015), and are a generalist consumer in estuarine ecosystems (Froese and Pauly, 2019), meaning that they are exposed to multiple pollutant accumulation pathways. Giant mud crabs *Scylla serrata* are a prized recreational and commercial species in Queensland due to their size (often > 25 cm carapace width) and ample consumable flesh (Alberts-Hubatsch et al., 2016; Webley et al., 2015). Giant mud crabs are top benthic predators and consume a variety of vertebrate and invertebrate fauna. They also create large burrows in sediments in coastal ecosystems (Alberts-Hubatsch et al., 2016) and so are likely to have greater exposure to sediment-bound pollutants. We hypothesised that the concentration of different pollutants would correlate with land uses in surrounding catchments and that this would likely result in hotspots for the accumulation of certain pollutants, but that these effects would differ between the two species we sampled.

2. Methods

2.1. Study region

We collected yellowfin bream and giant mud crabs from 13 estuaries in southeast Queensland, Australia (Fig. 1). These estuaries have previously been sampled for both broad, and habitat-specific patterns in fish assemblages. It has been established that the abundance and diversity of fish assemblages are shaped in this region by the extent and connectivity of both natural and anthropogenic habitats (Gilby et al., 2018; Gilby et al., 2017a; Gilby et al., 2017b; Olds et al., 2018), and that these effects can extend to key functional traits of key fisheries species, including giant mud crabs (Gilby et al., 2020). Our sampling extent within each estuary therefore followed the extent of these previously completed surveys; from the estuary mouth, to the point in the estuary where long-term winter salinity values (from the previous 10 years of monitoring) averaged 30 ppt (EHMP, 2019), which standardises the salinity range in which we sample in estuaries that vary widely in hydrology and size. Some species targeted by fisheries in the region are not adversely affected by either instream or catchment coastal urbanisation (Brook et al., 2018; Olds et al., 2018), and so it is plausible that these species have long-term exposure to pollutants in some estuaries.

We sourced the most recent land use information from the Queensland State Government (Queensland Government, 2015b), and clipped the land use layer for the catchment of each estuary sampled (Queensland Government, 2015a) in QGIS (QGIS Development Team, 2019). We then calculated the cover of primary and secondary land use types within the catchment of each estuary.

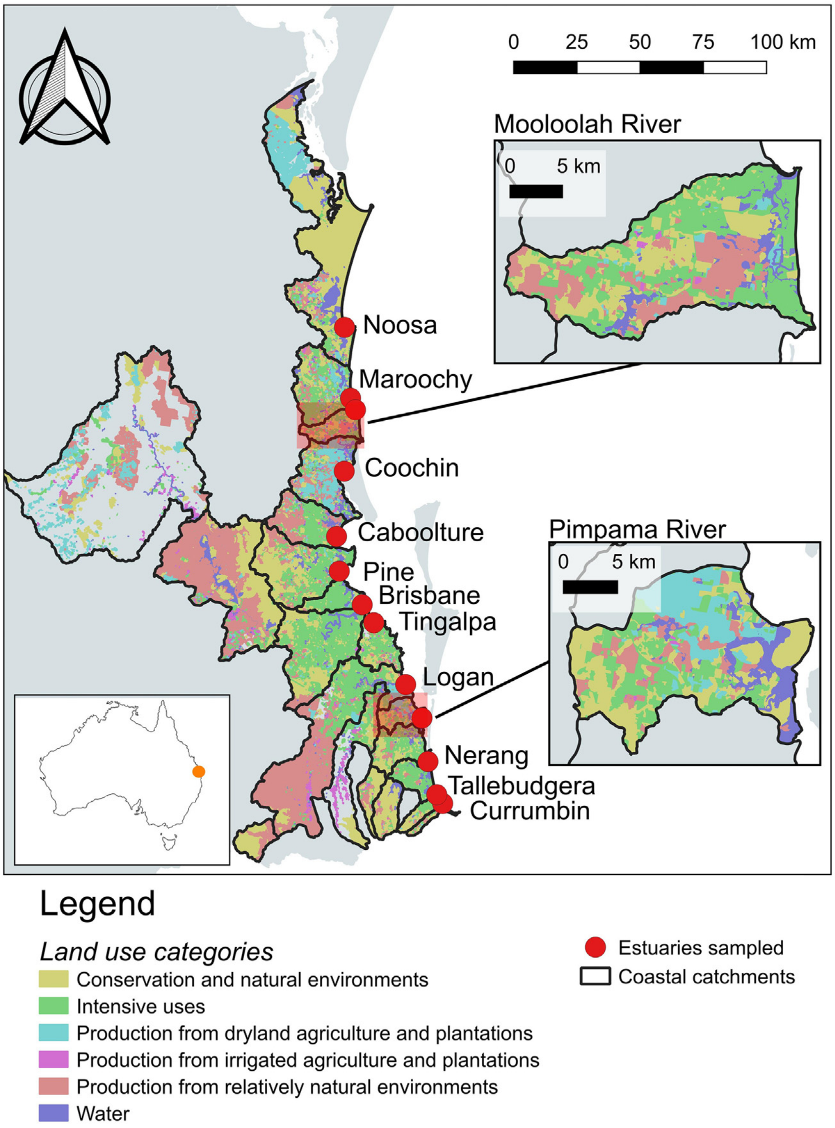

Fig. 1. Map of sampled estuaries and their land uses with the surrounding catchment.

2.2. *Specimen collections*

We collected between three and five legal sized (> 25 cm total length) yellowfin bream from each estuary (for a total of 57 yellowfin bream) using gill nets, cast nets, and angling between June and September 2018. We collected between 2 and 9 legal sized (> 15 cm carapace width, males only) giant mud crabs from each estuary (for a total of 65 giant mud crabs) between September 2018 and February 2019 using crab pots baited with sea mullet *Mugil cephalus*. Upon capture, all specimens were euthanized via blunt trauma to the cranium (according to USC animal ethics protocol ANA18126), placed into food grade Ziploc bags on ice, and frozen at −20 °C upon return to the laboratory. Rainfall in the region in the months leading up to and during the sampling period was average to below average (Australian Government Bureau of Meteorology, 2019).

2.3. *Pollutant analysis*

We quantified the concentration of pollutants (mg/kg wet weight) in the flesh of edible sized portions from each individual; flesh from shelled, male giant mud crabs and descaled whole yellowfin bream fillets (skin on, as this is how it is often consumed in the region). We quantified the concentration of 70 compounds: 14 heavy metals, including 4 arsenic species, 37 pesticides, and 19 PFASs (Table 1). All collections and sample preparation were conducted according to established protocols for the handling and processing of seafood samples for pollutant analysis to avoid any sample contamination (e.g. absence of field jackets and knives with coatings containing PFAS; Queensland Department of Environment and Science, 2018). PFAS and metal concentrations were quantified from all individuals captured, and pesticide concentrations were quantified from composite samples from each estuary (so equal weight of each subsample was taken to give a final sample weight of 10 g). All methods used for the analysis were accredited to ISO17025 by the National Association of Testing Authorities, Australia (NATA). The quality controls analysed with each batch of samples include blanks, duplicate samples, blank and matrix spikes and surrogates.

2.3.1. *PFASs*

Whole edible samples were homogenised and then extracted using 10 mL of 1% acetic acid (Sigma Aldrich Pharma grade) in acetonitrile (LiChrosolv Merck) using a Geno/Grinder 2010 SPEX. QuEChERS salts (Agilent) were added to the samples, the samples cooled and centrifuged. 2 mL of the supernatant was concentrated to 200 μL and PFASs

Table 1
List of pollutants sampled and their limit of analytical detection. Pollutants which had samples of either yellowfin bream or mud crabs detected above the limit of analytical detection given are shown in bold. Detection limits are never higher than food safety standards.

Pollutant group Limit of analytical detection (mg/kg)	Pollutant
Metals	
0.005	**Arsenic**, cadmium, **mercury**, **lead**
0.01	**Cobalt**, **chromium**, **manganese**, **nickel**, **antimony**
0.04	**Titanium**
0.05	**Copper**, **selenium**, tin, **zinc**
Arsenic speciation	
0.1	**Arsenobetaine**, dimethylarsinic acid, monomethylarsonic acid, inorganic arsenic
Pesticides[a]	
0.01	Chlordane cis, chlordane trans, DDD (o,p), DDD (p,p), DDE (o,p), DDT (p,p), endrin, HCB, HCH-a, HCH-B, lindane (HCH-y), methoxychlor, nonachlor cis, nonachlor trans
0.02	Aldrin, dieldrin, chlordene, chlordene epoxide, DDE (pp), DDT (o,p), endosulfan alpha, endosulfan beta, endosulfan ether, endosulfan sulfate, endrin aldehyde, HCH-gamma, heptachlor, heptachlor epoxide, oxychlordane
0.04	Total aldrin and dieldrin, total chlordane, total HCH isomers, total heptachlor
0.05	Chlordene (1-hydroxy), chlordene (1-OH-2,3-epoxy)
0.06	Total DDT, total endosulfan
PFAS	
0.001	**Perflurobutanoic acid**, perfluropentanoic acid, **perflurohexanoic acid**, **perfluroheptanoic acid**, perfluorooctanoic acid, perflurononanoic acid, **perflurodecanoic acid**, perflurobutanesulfunic acid, perflurohexanesulfunic acid, **perflurooctanesulfunic acid**, **perflurodecanesulfunic acid**
0.002	Perfluroundecanoic acid, perflurotridecanoic acid
0.004	Total C4–C10 sulfonic acids
0.005	Perflurododecanoic acid, perflurotetradecanoic acid, 4:2 fluorotelomer sulfonic acid, 8:2 fluorotelomer sulfonic acid
0.02	Total C4–C14 carboxylic acids

[a] Samples tested on compound samples of all individuals (separated by species) from each estuary only.

concentrations determined using Orbitrap LC-MS (Thermo Fisher Scientific).

2.3.2. Pesticides

Samples (due to cost, composite samples from each estuary, so equal weight of each subsample was taken to give a final sample weight of 10 g) were extracted with 2 × 100 mL portions of Acetone:Hexane (Merck Suprasolv) (10% v/v) using an IKA Ultra Turrex. Extracts were dried with sodium sulfate followed by concentration by rotary evaporation, transfer to pre-weighed glass tubes and final solvent was removed using dry block heating with nitrogen. The extracts were dissolved in dichloromethane (Fisher Optima), filtered and cleaned up using a gel permeation chromatography (J2Scientific using Waters Envirogel column). Post GPC the extracts were solvent exchanged to hexane and further cleaned up using Florisil macro columns (Labchem 60/100mesh pest. residue grade). Following rotary evaporation and nitrogen blow down the final 1 mL extracts were analysed using GCMS (Shimadzu QP2010 Plus).

2.3.3. Heavy metals

Samples were macerated and homogenised and the 1.0 g subsamples were taken. Subsamples were microwave digested (MarXpress, CEM) with concentrated nitric acid (4 mL) at gradual increase in temperature. The levels of trace elements in the solution were determined by the inductively coupled plasma mass spectrometry (ICP-MS, Agilent 8800). The analysis of arsenic speciation was carried out using HPLC-ICP-MS after nitric acid extraction.

2.4. Statistical analyses

We used distance-based linear models (DistLMs) in PrimerE to quantify correlations between the concentration of the suite of pollutants identified above the limit of analytical detection for each species, and the extent (in km^2) of primary land use categories in the catchment in which the individual was captured. Best-fit DistLMs were identified using corrected Akaike information criterion (AICc) on a Euclidean dissimilarity matrix of pollutants and normalised matrix of land use extents in each catchment. Differences in the suite of pollutants and the set of land use categories between catchments were visualised using non-metric multi-dimensional scaling (nMDS) ordinations with Pearson vector overlays.

3. Results

3.1. Land use

Overall, 26% of total land use in the region was protected in conservation areas and natural environments, which represents the lowest impact land use category, and likely lowest sources of pollutants. The extent of conservation areas and natural environments ranged between 18.6 and 1210 km^2, or 12.7 to 51.0% of land use within catchments (Figs. 1, 2, Table S1). Areas of production from relatively natural environments incorporate agricultural production from relatively unmodified ecosystems, including grazing of natural vegetation, and comprised 35% of the region's land use. The area of production from relatively natural environments ranged between 9.9 and 1607.6 km^2, or 6.7 to 46.4% of land use within catchments (Figs. 1, 2, Table S1). Intensive uses encompass highly modified landscapes for residential, mining, manufacturing, waste treatment and disposal and other services, and comprised 23% of the total regional land use. The area of intensive uses ranged between 0.7 and 360.0 km^2, or 12.3 to 90.1% of land use within catchments (Figs. 1, 2, Table S1). The area of dryland agriculture such as sugar, plantation forestry and other dryland cropping comprised 9% overall, and ranged between 38.0 and 844.6 km^2, or 0.49 to 89.1% of land use within catchments (Figs. 1, 2, Table S1). The area of irrigated agriculture and plantations (e.g. production from irrigated cropping and pastures; 2%) and waterbodies (e.g. rivers, wetlands and reservoirs; 5%) had the area of lowest land uses throughout the region (Figs. 1, 2, Table S1). The estuaries surveyed therefore represent the full range of conditions present in estuaries throughout south-east Queensland (Gilby et al., 2017b), ranging from estuaries that have catchments heavily urbanised by humans, to estuaries with up to

Fig. 2. Krona Plot of catchment land use types for (A) all 13 catchments in the study and (B) Mooloolah and (C) Pimpama Rivers. See supplementary material for all individual catchment plots. Colours match with colours for land use categories in Fig. 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

50% of the catchment area protected in conservation areas (Figs. 1, 2, Table S1). This selection represents a range from very highly impacted, with many potential sources of pollutants, to estuaries that have much less urbanisation, and so have fewer potential sources of pollutants.

3.2. Pollutants

We identified low occurrence and concentrations of most pollutants in the consumable flesh of yellowfin bream and giant mud crabs in southeast Queensland. Fifty-two (74%) of pollutants analysed returned no samples with concentrations above the limit of analytical detection (Table 1). In total, 8.96% of pollutant/sample combinations registered concentrations above the limit of analytical detection (Table 2). No samples contained concentrations of any pesticides above the limit of analytical detection (Tables 1, 2).

We found concentrations of 12 heavy metals above the limit of analytical detection in our samples (Table 1). Metals were detected (at or above the limit of analytical detection) in about half of all analyses, with more detections in fish (52%) than in giant mud crabs (40%) (Table 2). There is, however, considerable variation among metal species with some being detected in all individuals (arsenic, copper, mercury, selenium and zinc), whilst others were at concentrations below the limit of analytical detection in all samples (Tables 1, S2, S3). Between 80 and 100% of arsenic in samples was arsenobetaine, and we had no samples that recorded concentrations over the limit of analytical detection for dimethylarsinic acid, monomethylarsonic acid or inorganic arsenic. Detectable levels of cobalt (63% of yellowfin bream samples), chromium (14% of yellowfin bream samples), nickel (14% of

Table 2
Summary of pollutant groups, and the number and proportion of samples for yellowfin bream *Acanthopagrus australis*, giant mud crabs *Scylla serrata* and for all samples that registered concentrations above the limit of analytical detection (LoD).

Pollutant group	Species	Samples (n)	> LoD n	> LoD prop
Metals	Yellowfin bream	798	411	51.50%
	Giant mud crab	910	361	39.67%
	Total	1708	772	45.20%
Pesticides	Yellowfin bream	2109	0	0%
	Giant mud crab	2405	0	0%
	Total	454	0	0%
PFAS	Yellowfin bream	1197	3	0.25%
	Giant mud crab	1365	12	0.88%
	Total	2562	15	0.59%
All pollutants	Yellowfin bream	4104	414	10.08%
	Giant mud crab	4680	373	7.97%
	Total	8784	787	8.96%

yellowfin bream samples), lead (7% of yellowfin bream samples and 34% giant mud crab samples), antimony (2% of yellowfin bream samples), titanium (21% of yellowfin bream samples) and cadmium (22% of giant mud crab samples) were also identified (Tables S2, S3).

We found concentrations of six PFAS compounds above the limit of analytical detection in our samples (Table 1). PFAS compounds were detected (at the limit of analytical detection or above) in 0.59% of all analyses, with more detections in giant mud crabs (0.88%) than in yellowfin bream (0.25%) (Table 2). Six PFAS compounds were identified in mud crab samples (perfluoro-butanoic acid, perfluoro-decanoic acid, perfluoro-heptanoic acid, perfluoro-hexanesulfonic acid, perfluoro-octanesulfonic acid and perfluoro-octanoic acid), in up to 33% of samples from individual estuaries (Table S3). Only one PFAS compound was detected in yellowfin bream samples, with 50% (n = 2) of samples from Coochin Creek containing trace amounts perfluoro-octanesulfonic acid (PFOS) (0.002 mg/kg) (Table S2).

We identified spatial patterns in the suite of pollutants identified above the limit of analytical detection for both yellowfin bream and giant mud crabs. The suite of pollutants detected above the limit of analytical detection in yellowfin bream was best explained by the extent of intensive use in the surrounding catchment (Table 3A). Here, greater extent of intensive uses in the surrounding catchment correlated with greater concentrations of lead, titanium and nickel in the flesh of yellowfin bream (Fig. 3). Conversely, lower extent of intensive uses in the surrounding catchment correlated with greater concentrations of zinc, antimony and mercury (Fig. 3). Estuaries with the greatest extent of intensive usages in the catchment were the Brisbane, Pine, Logan and Caboolture Rivers (Fig. 1, Table S1). The suite of pollutants detected above the limit of analytical detection in giant mud crabs was best explained by the extent of water bodies and production from irrigated agriculture and plantations in the surrounding catchment (Table 3B). Here, greater extent of water bodies in the surrounding catchment correlated with greater concentrations of mercury and zinc, and greater extent of production from irrigated agriculture and plantations in the surrounding catchment correlated with lead and a suite of PFAS compounds (Fig. 4).

Detecting metals and organics with sensitive analytical equipment (i.e. have values above the limit of analytical detection) does not imply that fish or crustaceans are unfit for human consumptions. Most major pollutants of concern in the region were recorded substantially below the minimum levels that trigger reporting and further investigation by local authorities for either estuary-wide averages, or for individual samples (Fig. 5). Yellowfin bream samples from the Mooloolah River contained average levels of mercury (0.51 mg/kg) slightly above the report limit Australian food safety standards for average concentrations within individual locations (0.5 mg/kg), and were the only pollutant to do so, but had no individual samples above the maximum allowable concentration for individual fish (1.5 mg/kg) (Australian Government, 2017).

4. Discussion

Expansion and intensification of human land use in coastal catchments (Halpern et al., 2019) results in a greater diversity and volume of pollutants being released into waterways (Bosch et al., 2016; Jian et al., 2017; Murray et al., 2010). Because some pollutants can bioaccumulate into the consumable flesh of key seafood species, it is possible that human exposure to pollutants might be high in some heavily modified coastal areas (Barletta et al., 2019; Landos, 2013; Warwick et al., 2018; Wen et al., 2017). Despite this, we found relatively low occurrences and concentrations of all pollutants surveyed in giant mud crabs and yellowfin bream in southeast Queensland, Australia; a region that we hypothesised would have hotspots for the accumulation of certain pollutants given the diverse land uses present. We found that the extent of intensive uses, irrigated agriculture and water bodies in catchments surrounding estuaries correlated with the suite of pollutants in seafood species in southeast Queensland but that only the average concentration of mercury in one estuary was above Australian national guidelines for food safety (Australian Government, 2017; Food Standards Australia and New Zealand, 2018). In this sense, we found that very few attributes of land use in coastal catchments correlate with key pollutants in seafood species in this region. There may, therefore, be some natural sources for metals within individual systems contributing to the patterns found here that require further investigation. Such surveys might, however, be considered less important by local authorities as all levels (except for mercury in the Mooloolah River) identified in this study were below (usually significantly below) trigger levels in local food safety standards. This means that assuming the health consequences of eating seafood from these locations from presumed influences of the surrounding catchment might be incorrect in some scenarios. Whilst there are potentially risks for people that consume a significant amount of some seafood, these risk factors are not covered in local food safety

Table 3
Distance-based linear model (DistLM) marginal tests for correlations between the suite of pollutants identified above the limit of analytical detection with extents of land use types for A) yellowfin bream *Acanthopagrus australis* and B) giant mud crabs *Scylla serrata* in estuaries southeast Queensland, Australia. Values in bold are significant at α = 0.05.

Land use type	A. Yellowfin bream			B. Giant mud crabs		
	Pseudo-F	P	Prop. var.	Pseudo-F	P	Prop. var.
Conservation and natural environments	1.249	0.288	0.102	1.73	0.144	0.126
Intensive uses	2.01	**0.042**	0.154[a]	1.68	0.155	0.132
Production from dryland agriculture and plantations	1.256	0.214	0.102	1.36	0.267	0.11
Production from irrigated agriculture and plantations	0.953	0.496	0.079	3.72	**0.025**	0.252[a]
Production from relatively natural environments	1.413	0.199	0.113	1.77	0.116	0.139
Water	1.27	0.263	0.103	1.67	0.168	0.131[a]

[a] Included in best model.

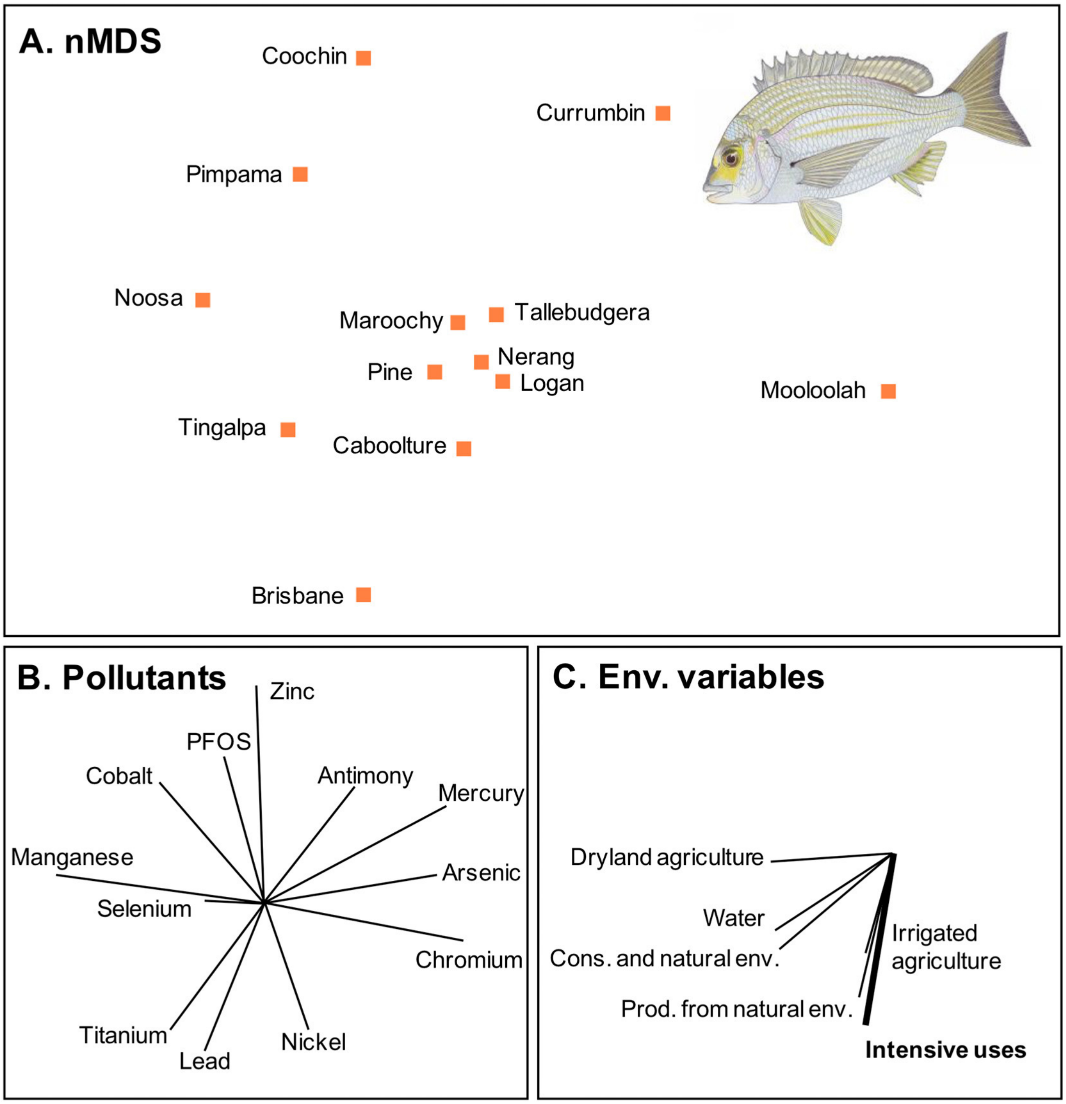

Fig. 3. Non-metric multi-dimensional scaling (nMDS) ordination of pollutants in the flesh of yellowfin bream *Acanthopagrus australis* in southeast Queensland, Australia (A) with Pearson vector overlays of pollutants (B) and environmental variables (C). Thicker environmental vectors with bold text indicate variables included in best-fit DistLM models (see Table 3A).

guidelines, and so require further research.

We found some differences in the number and concentrations of pollutants in the flesh of the two seafood species surveyed. Giant mud crabs had fewer heavy metals detected above the limit of analytical detection, but a higher prevalence of PFASs than yellowfin bream. These levels were, however, consistently lower than trigger levels in local seafood safety guidelines (Australian Government, 2017; Food Standards Australia and New Zealand, 2018). This supports the notion of different exposure pathways between these two species. Our results highlight the risks of generalising either land use or perceived impact levels to likely pollutant levels in coastal seafood species and suggest context-specific risk of pollutant accumulation. This is despite us analysing yellowfin bream fillets with the skin on, which can often significantly increase the concentration of some pollutants. Therefore, these results have consequences for the value of some coastal fisheries that might be perceived as potentially harmful to people in urbanised systems and stress the importance of surveying broadly across regions to quantify potential risks and pollutant levels in coastal seafood species. Our results also suggest, however, that intensification of some land use practices (principally irrigated agriculture and intensive land uses) may increase pollutant exposure risks in the future in this region.

We identified distinct correlations between the concentration of pollutants and heavy metals in the flesh of yellowfin bream and giant mud crabs, with land use in the surrounding catchments. Areas of intensive use in catchments correlated with higher concentrations of some pollutants, and lower concentrations of others in yellowfin bream. Heavy metals are naturally occurring, but are increasingly common in urban estuaries given their diverse usages in areas of extensive human modifications (Bosch et al., 2016; Landos, 2013; Tchounwou et al., 2012), and the centralisation of human populations along coastlines (Halpern et al., 2008). We found higher concentrations of the heavy metals lead, titanium and nickel in yellowfin bream captured in more urbanised estuaries. There are clear links between these pollutants and fuel combustion (lead), paints (lead, titanium) glass (lead), manufacturing (all) and as a component of other chemicals and wastewater (all) (Bosch et al., 2016; Cempel and Nikel, 2006; Tchounwou et al., 2012), and potentially strong effects of these pollutants being centralised towards the mouths of estuaries (Oosthuizen and Ehrlich, 2001). Conversely, yellowfin bream from estuaries with a narrower extent of intensive areas in the catchment have concentrations of heavy metals that are higher for several metal species. For example, there were clear high values of mercury and chromium in the Mooloolah,

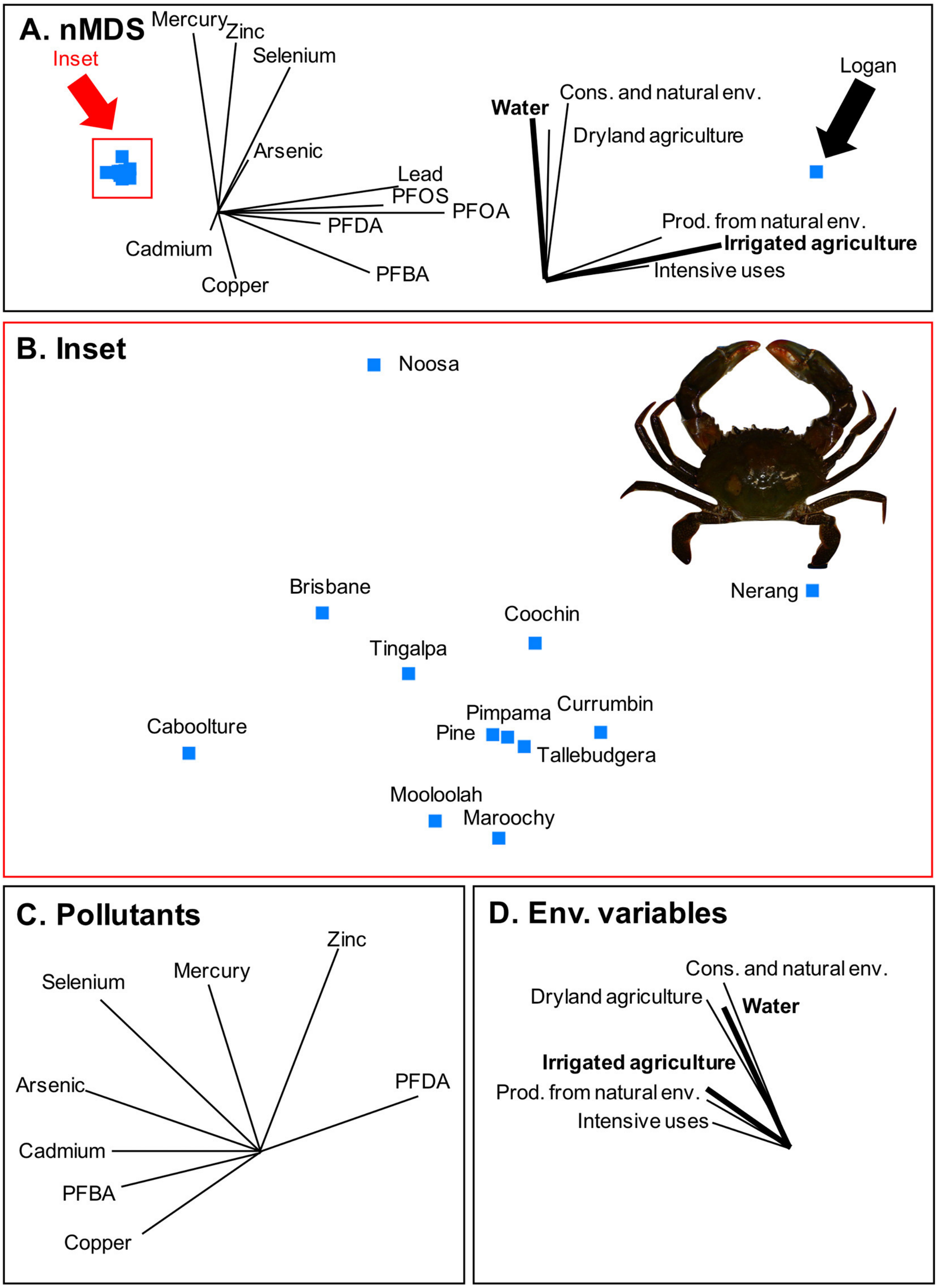

Fig. 4. Non-metric multi-dimensional scaling (nMDS) ordination of pollutants in the flesh of mud crabs *Scylla serrata* in southeast Queensland, Australia with Pearson vector overlays of pollutants and environmental variables. Panel A is the main nMDS plot, with panels B, C and D showing the details of the clustered sites to the left of the main panel. Thicker environmental vectors with bold text indicate variables included in best-fit DistLM models (see Table 3B).

Tallebudgera and Currumbin estuaries; some of the smaller estuaries and catchments we sampled. It is also, possible, however, that given the low pollutant concentrations found in this study, that were predominantly below the trigger levels of local food safety guidelines, that the correlations found are simply cause by some random variation of very low concentrations of pollutants. This hypothesis, however, requires further testing.

The Logan River sits significantly apart from all other estuaries for giant mud crabs primarily due to detectable (albeit low) levels of several PFAS compounds in the crabs from this system. Patterns in pollutants in giant mud crabs were best explained by the extent of water bodies and irrigated agriculture in the catchment. There may be an effect of some forms of irrigation on higher likelihood of lead being washed into the estuary (O'Sullivan et al., 2012), but there are no clear sources of PFAS in the Logan River catchment (e.g. very large airports), thereby potentially indicating that it is simply a co-incidental

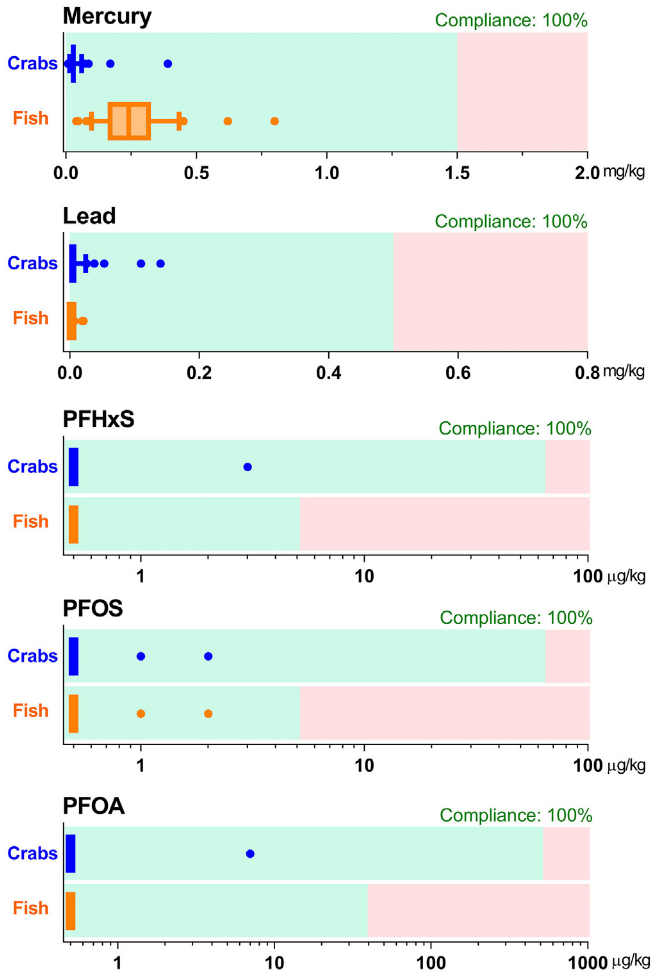

Fig. 5. Box plots of distribution of pollutants of major concern in the flesh of giant mud crabs *Scylla serrata* (crabs) and yellowfin bream *Acanthopagrus australis* (fish) in southeast Queensland, Australia, relative to Australian Food Safety Standards (compliance levels in green for individual fish/samples, levels above compliance and requiring reporting to authorities in red) (Australian Government, 2017; Food Standards Australia and New Zealand, 2018). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

correlation with un-identified PFAS sources in the catchment in this instance. Surprisingly, therefore, we found no sensible correlations between concentrations of some pollutants like PFAS (linked to airports and high urbanisation), mercury (some industrial activities) and pesticides (agricultural activities, principally) with land use variables for either seafood species in our study. There may also be an effect of larger catchments having greater volumes of water and tidal flushing which in turn dilutes some pollutants as there was no consistent effect of simply larger land uses or catchments having greater pollutant loads. Previous studies have shown larger concentrations of PFASs in the flesh of very similar estuarine species along the Australian east coast (Taylor and Johnson, 2016). PFASs depurate quickly from the flesh of seafood species (Taylor et al., 2017), which may explain the lack of PFAS in our samples collected broadly across estuaries. This also suggests, however, that capturing animals close to PFAS sources (and likely also sources of other contaminants), especially immediately following large rainfall events or spills, may yield significantly different results. Combined, our results indicate that regional spatial patterns in pollutant loads in seafood are much more complex than simply following a natural to urban gradient with respect to catchment land use types and size.

The concentrations of pollutants and heavy metals we identified in this study were essentially all below Australian standards for seafood, some significantly so (Australian Government, 2017). The majority of arsenic found in our samples was arsenobetaine, which is considered to be non-toxic (Sloth et al., 2005), and we found no samples with detectable levels of the more dangerous arsenic species. The key exception was that we found average concentrations of mercury slightly higher than local food safety standards (set at 0.5 mg/kg) in the Mooloolah River (0.51 mg/kg) (Australian Government, 2017). The Mooloolah River occurs in the northern part of our study region, with 39% of its catchment subject to intensive human use, 21% conserved or in relatively natural condition, and 30% subject to agricultural production on relatively natural ecosystems like grasslands. Consequently, there are no attributes of land use in the Mooloolah River that set it significantly apart from land use in other estuaries. The Mooloolah River does, however, have a large harbor towards the estuary mouth that is a major port for local fisheries; the only estuary in our study with this spatial arrangement around a relatively narrow estuary mouth. However, these attributes are unlikely to be contributing to the patterns found here because mercury accumulation in fish is more influenced by concentrations in sediments and water column, and there is unlikely to be major sources of mercury in the harbor (Calta and Canpolat, 2006; Verdouw et al., 2010). The potential causes of the spike in mercury levels could be investigated further in this estuary, especially if further surveys show broader effects across higher trophic level species.

In addition to these low concentrations of heavy metals in our samples, we found low occurrences and low concentrations of both pesticides and PFASs. Concentrations of PFASs were significantly lower than levels that trigger broader investigations in Australian national food safety standard guidelines (Food Standards Australia and New Zealand, 2018). No samples had detectable levels of pesticides. This somewhat surprising result has several potential explanations in southeast Queensland. Many of the estuaries we sampled are large estuaries with wide inlets that open either to the ocean or large bays (Gilby et al., 2017a). Consequently, it could be hypothesised that any pollutant releases are quickly diluted and released into the open ocean via tidal flushing and river flows, thereby reducing the risk of bioaccumulation in fish. These effects might, however, be considered unlikely because we also surveyed relatively small estuaries, with lower flow, narrower estuary inlets, longer water residence times, and a diversity of potential pollutant sources in this study (Gilby et al., 2017a). There is also good evidence to suggest that these compounds can depurate from seafood relatively quickly (at time scales of days) once exposure is removed (Taylor et al., 2017). Rainfall in the months leading up to, and during sampling was average to below average (but not considered drought conditions) according to local authorities (Australian Government Bureau of Meteorology, 2019), so this may also have served to reduce pollutant loads in some systems.

We surveyed the concentrations of pollutants in two key coastal fisheries species that have fundamentally different ecological and biological traits. These species were chosen to represent two particular groups of coastal fisheries targets (yellowfin bream- mobile generalist benthic consumer, and giant mud crab- benthic predator). However, there may be attributes of the species chosen that reduce their likelihood of accumulating pollutants. For example, giant mud crabs are top benthic predators, that live in close association with the benthos (Alberts-Hubatsch et al., 2016); we hypothesised that this would increase their exposure to a diversity of pollutants. Whilst this may have been the case for PFASs, where we identified a higher diversity and occurrence of detectable pollutants in giant mud crabs than in yellowfin bream, the giant mud crabs relatively low trophic level may not expose them to significant bioaccumulation risk that other, higher level predators might be exposed to. Therefore, surveying other, higher level consumers may yield different results. For example, surveying large, mobile piscivorous fish that inhabit the mouths of estuaries over long periods (months to years; like platycephalids, carangids or lutjanids) might yield different results as the capacity for pollutants to bioaccumulate in these longer-lived, large predatory fishes might be greater

(El-Moselhy et al., 2014; Pourang, 1995; Power et al., 2002). Therefore, future studies could survey more broadly across both land use intensity and the trophic spectra to ensure the consistence of our results in the region. Previous studies have, however, found reportable levels of PFASs and other pollutants in the species that we surveyed in this study (e.g. Kamaruzzaman et al., 2012; Taylor and Johnson, 2016).

We hypothesised that the diverse land use and extent of urbanisation present in southeast Queensland would result in high concentrations and diversity of pollutants in seafood. An alternative explanation for the lack of significant concentrations of pollutants in our samples (certainly concentrations above recommended or limit of analytical detection in food safety standards) might be that the region either is simply not modified enough, or contain enough potential sources of pollutants relative to other regions where these effects have been established (Gu et al., 2015; Jian et al., 2017; Tepe et al., 2017), or that local regulations around pollutant releases are stringent and well enforced enough to limit these effects. Combined, these potential effects may result in the low occurrences and concentration of pollutants detected here. Consequently, it is vital for existing management that limits the spread of harmful substances to continue, and for these not to be weakened within the region to ensure the ongoing safety of seafood. Reported occurrences of large releases of pollutants are rare in southeast Queensland and are therefore covered extensively by local media. Public concerns regarding potentially harmful concentrations of pollutants often increase around these events, potentially leading to poorer perceptions of the health and condition of coastal fisheries in the long run. Unfortunately, there are no existing data in the region that could be used as a baseline for this study. Whilst the results of this study could be used as this baseline, it should be considered important to continue to monitor the concentrations of potentially harmful substances in seafood within the region over years and under different rainfall conditions. Given the diversity of land use across the study region (see Figs. 1 and 2), and the type of geology present in the region (Queensland Government, 2020), there is unlikely to be any correlation between land use and geological formation that might explain the patterns we found here. We acknowledge that this study did not analyse the effects of point source releases on pollutant levels in seafood species. For example, the study region contains many (approximately 40) wastewater treatment plants of varying size and positioning in estuaries (i.e. some releasing close to estuary mouths where the influence on this study would be low, and others releasing further upstream). Quantifying the effects of wastewater on the results found here should be the focus of subsequent studies. In any case, the results of this study highlight the need quantify the concentration and prevalence of pollutants in individual species and regions to establish potential exposure risk of pollutants to people.

In this study, we quantified the concentration of a suite of environmental pollutants that are considered potentially harmful to people in two coastal fisheries species within southeast Queensland. Given the diversity of potential environmental pollutants present in coastal ecosystems globally and the diversity of species harvested for seafood (FAO, 2018; Webley et al., 2015), the results of this study do not indicate a complete lack of potentially harmful pollutants in all seafood in the region. For example, global studies of flathead mullet *Mugil cephalus* indicate context specific effects of human impacts on pollutant levels in consumable flesh (Waltham et al., 2013). Our results do suggest, however, that some land use practices may contribute more significantly towards pollutant loads than others, and this could be used as a predictor of potential threats into the future. Despite this, the results of this study are positive indication of potentially lower risks of many environmental pollutants that are of strong concern to people within southeast Queensland. They also support the notion that perceived risk and broad land use information might be poor indicators of human exposure to pollutants through seafood in some regions. We stress however, the importance of thorough surveys across a wider variety of seafood species and a greater number and diversity of pollutants in order to more thoroughly quantify these affects both within this region and beyond.

CRediT authorship contribution statement

Ben L. Gilby: Conceptualization, Methodology, Investigation, Resources, Formal analysis, Data curation, Writing - original draft, Writing - review & editing, Project administration, Funding acquisition. **Andrew D. Olds:** Conceptualization, Methodology, Writing - review & editing, Funding acquisition. **Felicity E. Hardcastle:** Investigation, Resources, Data curation, Writing - review & editing, Project administration. **Christopher J. Henderson:** Conceptualization, Formal analysis, Writing - review & editing. **Rod M. Connolly:** Conceptualization, Writing - review & editing. **Tyson S.H. Martin:** Investigation, Resources, Data curation, Writing - review & editing, Project administration. **Paul S. Maxwell:** Conceptualization, Methodology, Writing - review & editing, Funding acquisition. **Lucy A. Goodridge Gaines:** Investigation, Writing - review & editing. **Tyson R. Jones:** Investigation, Data curation, Writing - review & editing, Project administration. **Ariel Underwood:** Investigation, Writing - review & editing. **Thomas A. Schlacher:** Conceptualization, Resources, Writing - review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2020.111487.

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