



Review

Sources, presence and potential effects of contaminants of emerging concern in the marine environments of the Great Barrier Reef and Torres Strait, Australia

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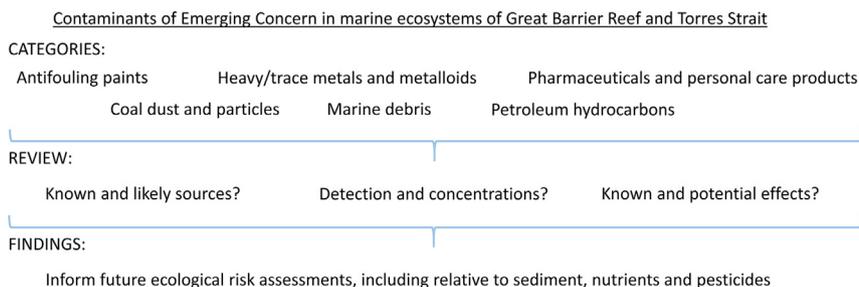
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HIGHLIGHTS

- We review the impacts of contaminants of emerging concern (CECs) on coral reefs.
- The sources, presence and potential effects of six CEC categories are discussed.
- Known and likely sources are relatively well known and spatially represented.
- Monitoring data and effect information for potential high risk CECs are scarce.
- The findings are relevant to tropical marine ecosystems around the world.

GRAPHICAL ABSTRACT



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ABSTRACT

Current policy and management for marine water quality in the Great Barrier Reef (GBR) in north-eastern Australia primarily focusses on sediment, nutrients and pesticides derived from diffuse source pollution related to agricultural land uses. In addition, contaminants of emerging concern (CECs) are known to be present in the marine environments of the GBR and the adjacent Torres Strait (TS). Current and projected agricultural, urban and industrial developments are likely to increase the sources and diversity of CECs being released into these marine ecosystems. In this review, we evaluate the sources, presence and potential effects of six different categories of CECs known to be present, or likely to be present, in the GBR and TS marine ecosystems. Specifically, we summarize available monitoring, source and effect information for antifouling paints; coal dust and particles; heavy/trace metals and metalloids; marine debris and microplastics; pharmaceuticals and personal care products (PPCPs); and petroleum hydrocarbons. Our study highlights the lack of (available) monitoring data for most of these CECs, and recommends: (i) the inclusion of all relevant environmental data into integrated databases for building marine baselines for the GBR and TS regions, and (ii) the implementation of local, targeted monitoring programs informed by predictive methods for risk prioritization. Further, our spatial representation of the known

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and likely sources of these CECs will contribute to future ecological risk assessments of CECs to the GBR and TS marine environments, including risks relative to those identified for sediment, nutrients and pesticides.

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1. Introduction

Contaminants of emerging concern (CECs) have been defined as *'naturally occurring, manufactured or manmade chemicals or materials which have now been discovered or are suspected [to be] present in various environmental compartments and whose toxicity or persistence are likely to significantly alter the metabolism of a living being'* (Sauvé and Desrosiers, 2014). The potential to pose risk to the environment means that CECs include contaminants that have been detected in the environment only recently (i.e. emerging contaminants) (Gavrilescu et al., 2015; Geissen et al., 2015), contaminants that are present in the environment and have become of recent concern (i.e. contaminants of emerging interest), and more well-recognized contaminants for which new information on environmental risk has become available (i.e. traditional contaminants) (Sauvé and Desrosiers, 2014). Consequently, CECs comprise a large variety of chemicals belonging to diverse classes and can be categorized in multiple ways depending on their chemical structure and/or mode of action (Borgert et al., 2004). For example, the US EPA lists persistent organic pollutants (POPs), pharmaceuticals and personal care products (PPCPs), veterinary medicines, endocrine-disrupting chemicals (EDCs) and nanomaterials as CECs (U.S. Environmental Protection Agency Emerging Contaminants Workgroup, 2008). Other studies also consider lifestyle compounds, such as caffeine and nicotine, illicit (recreational) drugs (Robles-Molina et al., 2014) and, more recently, microplastics (Sedlak, 2017) to be CECs. The sources of CECs are as diverse as the chemicals themselves (U.S. Environmental Protection Agency Emerging Contaminants Workgroup, 2008), and include point sources such as urban and industrial wastewater treatment plants (WWTPs), and diffuse sources such as agricultural land uses for animal and crop production (Geissen et al., 2015). Because CECs can potentially cause deleterious effects in aquatic organisms at environmentally relevant concentrations (U.S. Environmental Protection Agency Emerging Contaminants Workgroup, 2008), understanding their transport, fate and effects has received wide-

spread attention in receiving freshwater, coastal and marine environments (Gavrilescu et al., 2015; Islam and Tanaka, 2004; Pal et al., 2010).

The Great Barrier Reef (GBR) extends over 2,000 km along the northeast coast of Australia (Fig. 1a), covering an area of 348,000 km² that includes 20,000 km² of coral reefs, 43,000 km² of seagrass meadows and extensive mangrove forests (GBRMPA, 2014). The GBR is managed as a multiple-use marine protected area, allowing for commercial and non-commercial uses such as commercial marine tourism; commercial, recreational and indigenous fishing; ports and shipping; and defence activities (GBRMPA, 2014). Over the past decades, the condition of GBR ecosystems has shown severe decline (De'ath et al., 2012; Hughes et al., 2017) and continues to be at risk from climate change, poor water quality, coastal development and fishing pressure (GBRMPA, 2014). Although not part of the GBR Marine Park or World Heritage Area (WHA), the reef's ecosystems extend into the Torres Strait (TS) region bordering Papua New Guinea (Lawrey and Stewart, 2016) (Fig. 1a), and support locally important commercial and indigenous fisheries (Plaganyi et al., 2013). The TS marine ecosystems are exposed to immense freshwater and sediment input from nearby coastal rivers in Papua New Guinea (Wolanski et al., 2013) and to intense international shipping activities transiting between the Indian and Pacific Oceans (Det Norske Veritas Ltd, 2013).

Current policy and management for marine water quality in the GBR Marine Park and WHA focusses primarily on sediment, nutrients and pesticides derived from diffuse source pollution related to agricultural land uses (Brodie et al., 2012; Kroon et al., 2016). Other contaminants, including ones that could be considered CECs, have been detected in the marine ecosystems of the GBR and TS such as antifouling paints (Haynes et al., 2002; Haynes and Loong, 2002), coal dust and particles (GHD Pty Ltd, 2012a), marine debris (Hardesty et al., 2016; Haynes, 1997) and microplastics (Hall et al., 2015; Kroon et al., 2018; Reisser et al., 2013), heavy/trace metals and metalloids (Haynes and Johnson, 2000; Haynes and Kwan, 2002; Negri et al., 2009) and petroleum hydrocarbons

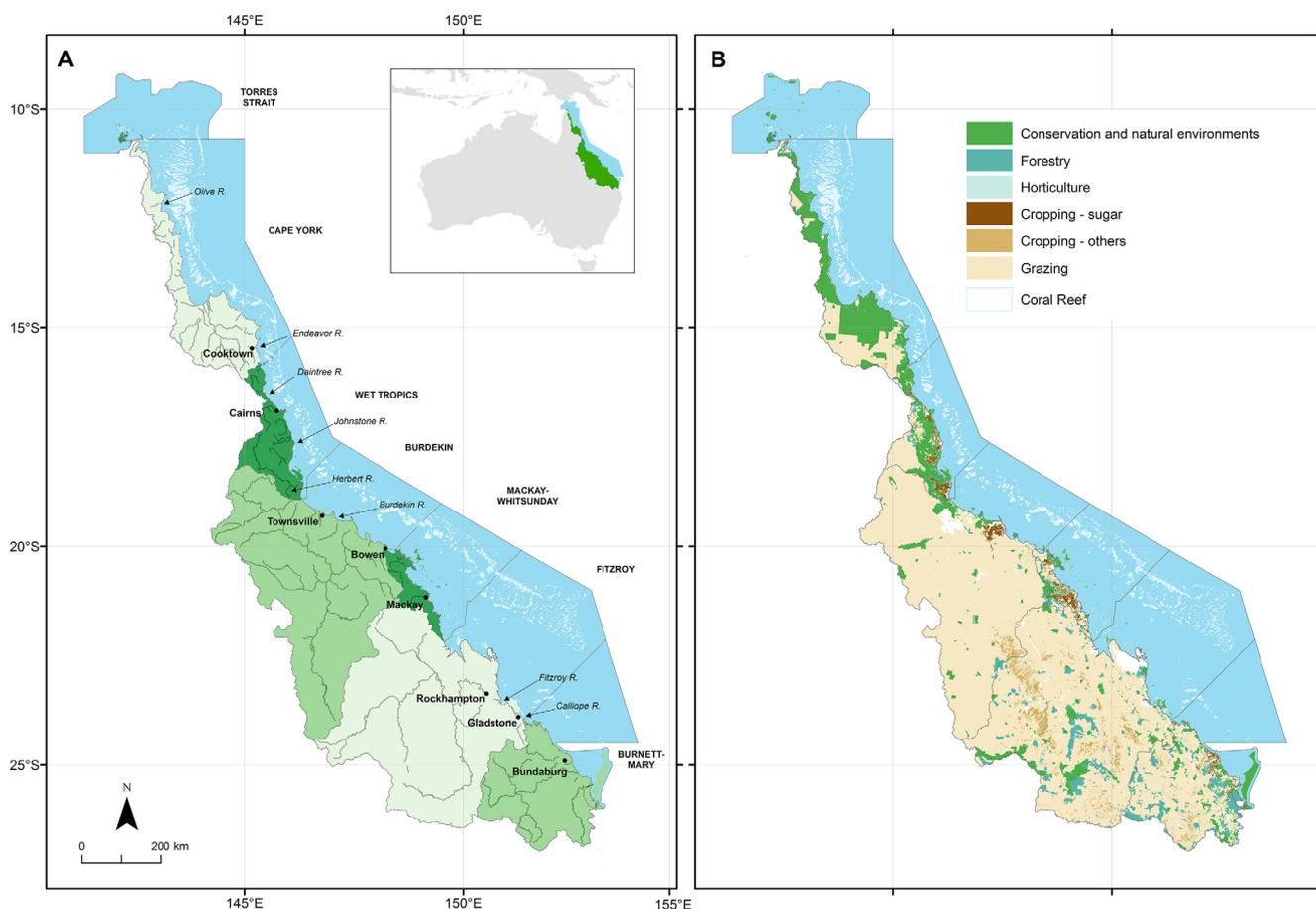


Fig. 1. The location of the Great Barrier Reef (GBR) and Torres Strait marine ecosystems along the northeast coast of Australia, including (A) major population centres, rivers, and seven natural resource management regions that overlap with these ecosystems, and (B) main agricultural production land uses, as well as conservation and natural environment land uses in the adjacent catchment area. Spatial data were sourced from the GBR Marine Park Authority, the Queensland Department of Natural Resources and Mines, and the Queensland Land Use Mapping Program (accessed April 2017) (Queensland Government, 2017b). Land use data were classified according to the Australian Land Use and Management Classification Version 8 (ABARES, 2016) (see Supplementary Material Text S1 for more detail).

(Haynes and Johnson, 2000). In addition, pharmaceuticals and personal care products (PPCPs) have been detected in treated domestic wastewater discharged into coastal waters adjacent to the GBR (O'Brien et al., 2014). Although the sources of these six categories of CECs are relatively well understood, little is known about their transport, fate and effects on the GBR and TS marine ecosystems. Importantly, current and projected agricultural, urban and industrial developments are likely to increase the sources and diversity of contaminants being released into these marine ecosystems (Det Norske Veritas Ltd, 2013; GBRMPA, 2014).

In this review, we evaluated the sources, presence and potential effects of six different categories of CECs known to be present, or likely to be present in GBR and TS marine ecosystems. The six categories of CECs to review were prioritized based on outcomes of a one-day workshop with the project team, end users and key stakeholders in 2015 (Kroon et al., 2015). Specifically, we (i) determined the presence and locations of these CECs based on available monitoring data and the types of human activities present, and (ii) examined the potential effects of these CECs on the marine ecosystems based on general mode of action and organisms likely to be exposed. As many of the known or likely sources for these six different categories of CECs are the same, we first described and spatially represented these sources in and adjacent to the GBR and TS marine ecosystems. Then, for each of the six different categories of CECs, we defined and described the individual CEC category, and summarized available source, monitoring and effect information from the primary scientific literature and from available grey liter-

ature and databases. Our evaluation was conducted at the scale of the GBR and TS marine ecosystems, as well as at the scale of seven individual natural resource management (NRM) regions that overlap with these ecosystems and are managed by regional NRM bodies responsible for protecting and managing Australia's natural resources (Fig. 1a). Based on our findings, we made recommendations to improve future ecological risk assessments of CECs to the GBR and TS marine ecosystems.

2. Sources of contaminants to the Great Barrier reef and Torres Strait

The decline in GBR water quality is associated with substantial increases in terrestrial pollutant fluxes (Bartley et al., 2017; Kroon et al., 2012), following the conversion of most of the 423,000 km² catchment (~80%) into agricultural production since the 1850 s (Brodie et al., 2012; Waters et al., 2014) (Fig. 1b). In addition to rangeland cattle grazing, forestry, horticulture and cropping (Fig. 1b), at least 500 intensive animal farming facilities are present in the GBR catchment including beef cattle feedlots, pig and poultry farms, dairy sheds and yards, and aquaculture facilities (Department of the Environment and Energy, 2019; Queensland Government, 2017b) (Fig. 2d). Other known and likely sources of contaminants to the GBR and TS marine environments include urban residential areas (via stormwater runoff), waste treatment and disposal, mining, industrial areas, ports and shipping, and

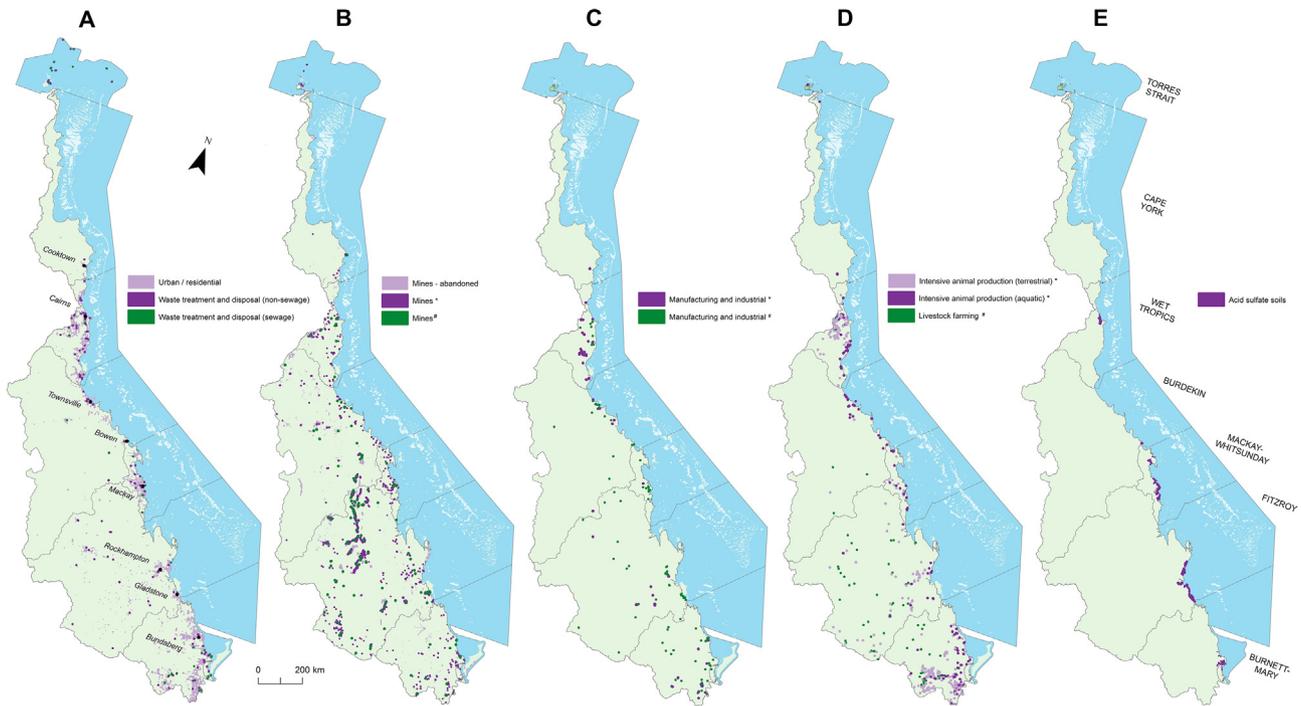


Fig. 2. Sources of contaminants to the Great Barrier Reef and Torres Strait marine ecosystems, including (A) urban, rural, and remote residential areas and associated waste treatment and disposal, (B) mining activity and abandoned mines, (C) manufacturing and industrial areas, (D) intensive animal production, and (E) acid sulfate soils, in the adjacent catchment area. Spatial data for (A–D) were sourced from the Queensland Land Use Mapping Program (*; accessed April 2017) (Queensland Government, 2017b) and the National Pollutant Inventory (#; accessed May 2019) (Department of the Environment and Energy, 2019), and classified according to the Australian Land Use and Management Classification Version 8 (ABARES, 2016) and the Australian and New Zealand Standard Industrial Classification (Trewin and Pink, 2006), respectively (see Supplementary Material Text S1 for more detail). Spatial data on abandoned mines and acid sulfate soils were sourced from Queensland Department of Natural Resources and Mines and the Queensland Department of Science, Information Technology and Innovation (both accessed April 2017), respectively. Other data sources and descriptions as presented in Fig. 1.

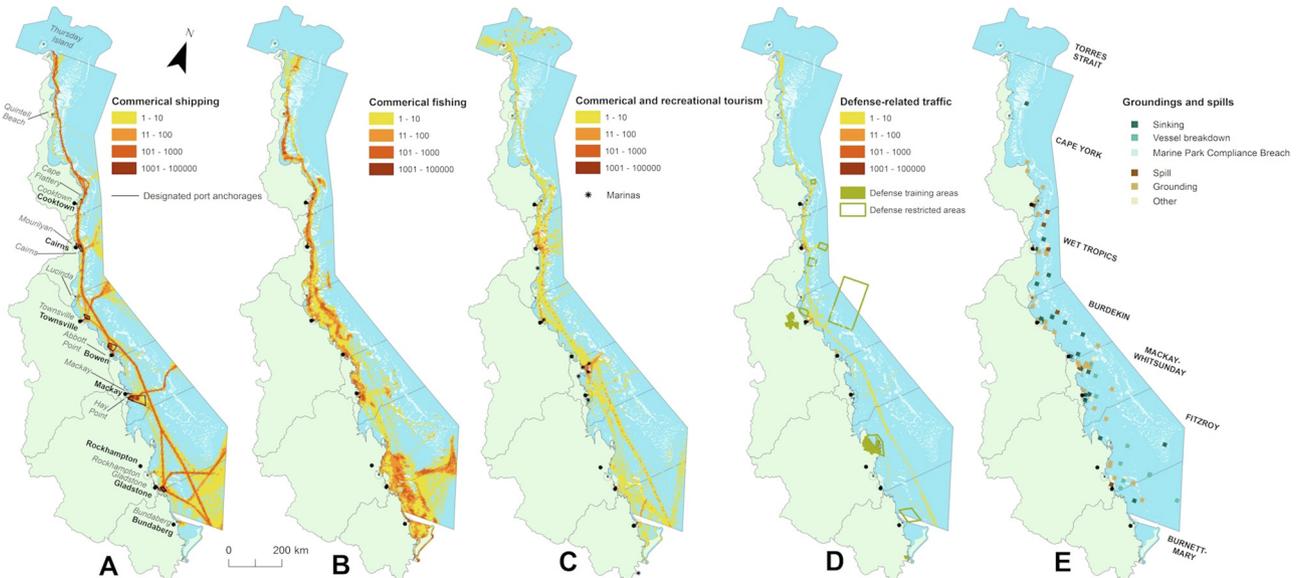


Fig. 3. Sources of contaminants to the Great Barrier Reef and Torres Strait marine ecosystems, including observed vessel traffic associated with (A) commercial shipping (maximum value 93,560), (B) commercial fisheries shipping (maximum value 40,019), (C) commercial and recreational marine tourism (maximum value 23,721), and (D) Defence activities within the GBR and TS marine environments (maximum value 3,197), as well as (E) vessel groundings and spills that caused significant damage to coral reef habitat and environments in the GBR Marine Park, in 2016. Observed vessel traffic was extracted from the Craft Tracking System database downloaded from the Australian Maritime Safety Authority (AMSA) website for each month in 2016 (accessed May 2017) (see Supplementary Material Text S1 for more detail). Vessel groundings and spills data were sourced from the Maritime Incidents Database 2016 [unpublished data], GBR Marine Park Authority, Townsville. The location of ports and designated port anchorages were sourced from the four Government-owned Port Corporations, namely North Queensland Bulk Ports Corporation, Port of Townsville Limited, Gladstone Port Corporation, and Ports North (accessed April 2017). Location of marinas were sourced from Queensland Department of Natural Resources and Mines (accessed May 2017). Defence facilities on land were extracted from the Queensland Land Use Mapping Program (accessed April 2017). Defence exclusion zones at sea were derived from the dataset ‘Defence Prohibited and Training Areas’ (© Copyright Commonwealth of Australia (Department of Defence) 2015). Other data sources and descriptions as presented in Fig. 1.

defence activities. The current population of northern Queensland exceeds 1.2 million people, with the majority living in Townsville (194,000 people), Cairns (160,000), Mackay (124,000), and Bundaberg (94,000), Rockhampton (84,000) and Gladstone (67,000); <5,000 people live in the Torres Strait region (Australian Bureau of Statistics, 2017) (Fig. 2a). This population is serviced by >50 operational WWTPs that discharge into rivers connected to the GBR marine environment (Hill et al., 2012), and at least 32 landfills that take waste from household, business and construction activities (Queensland Government, 2017b) (Fig. 2a). The population in the GBR catchment and on Torres Strait islands is projected to increase from 2011 to 2035 (Queensland Government, 2016c), with a forecasted annual growth of 1.6% for the GBR catchment (GBRMPA, 2014).

The GBR catchment is rich in mineral and energy (i.e. coal) resources and has long supported diverse mining activities (GBRMPA, 2014) (Fig. 2b). Queensland has > 15,000 abandoned mines with many occurring in the GBR catchment and a few in the TS (Queensland Government, 2016a) (Fig. 2b). More than 50 coal mines currently operate in Queensland with further large-scale coal mines being proposed in the GBR catchment (Department of Natural Resources and Mines, 2017a). Discharges from the Fly River (impacted by mining) and other, smaller coastal rivers flowing from Papua New Guinea also contribute contaminants and sediments to northern and north-central regions of the TS (Gladstone, 1996; Haynes and Kwan, 2002; Heap and Scaffi, 2008). Metal-related industries such as refineries are located in industrial areas near urban centres and ports such as Gladstone and Townsville (Department of State Development Infrastructure and Planning, 2014; Department of the Environment and Energy, 2019). Manufacturing and industrial areas (with the exception of mining) are not extensive but can be intensive at a local scale (Fig. 2c). In combination, facilities like mines, power stations and factories emit a range of contaminants into different environmental compartments, including air, land and water (Department of the Environment and Energy, 2019).

A total of 14 commercial ports are located on the coasts of the GBR and TS (Fig. 3a), with Abbot Point, Hay Point and Gladstone being among the largest coal export ports in the world (Department of Transport and Main Roads, 2016). In 2012–13, almost 11,000 movements of large commercial ships (length > 50 m) were monitored, with 94% of the 4,440 vessel arrivals berthing at the five major ports (Abbot Point, Cairns, Gladstone, Hay Point, Townsville) (North-East Shipping Management Group, 2014) (Fig. 3a). Movements of large vessels within the GBR and TS are highly regulated and managed, with designated routes, both inside the lagoon and between reefs to the outer routes in the Coral Sea (GBRMPA, 2014) (Fig. 3a). It is estimated up to 90% of the cargo tonnage transported from the north-east region comprises coal (Braemer Seascope, 2013; North-East Shipping Management Group, 2014). Shipping traffic was projected to increase ~2.6-fold by 2032 (based on 2011–12 numbers) (Braemer Braemer Seascope, 2013; PGM Environment, 2012), depending on future trade and marketing opportunities (Det Norske Veritas Ltd, 2013; North-East Shipping Management Group, 2014). Average vessel size of bulk carriers is also projected to increase from 2011 to 2025 (Braemer Seascope, 2013). The total anchorage area for bulk cargo and other trading vessels at the five larger ports adds to 2,881 km², with the area of Hay Point (1,573 km²) being the busiest (GHD Pty Ltd, 2013). At this area, 720 ships proceed to anchor each year with up to 60 ships anchored at any one time; ships can be at anchor for up to two months with an average stay of 19 days (GHD Pty Ltd, 2013). In addition, 83,000 privately registered recreational vessels and 485 commercial fishing trawlers were operating in the GBR and TS regions in 2012–13 (North-East Shipping Management Group, 2014) (Fig. 3b, c). Commercial marine tourism

in the GBR is predominantly vessel based, with >80% of day visits in 2013 concentrated in the Cairns and Whitsunday Planning Areas (GBRMPA, 2014) (Fig. 3c). The number of reported ship collisions and groundings in the GBR WHA ranges from zero to five per year since 1985 (GBRMPA, 2014) (Fig. 3e). All reported collisions were between ships and smaller vessels, while groundings include those within designated port areas (GBRMPA, 2014). Under current management arrangements in the GBR WHA, groundings of large vessels are predicted to occur once every 10 years (GBRMPA, 2014).

The Australian Defence Force operates and trains in the GBR and TS regions, including Australian Navy, Army and Air Force bases at Cairns and Townsville, and designated defence training areas on land, in the air and at sea (GBRMPA, 2014; PGM Environment and Eco Logical Australia, 2014) (Fig. 3d). Operational activities include ocean surveillance, maritime search and rescue missions, and hydrographic survey and charting. Two training areas, namely the Townsville Star and the Shoalwater Bay Training Area, are among the largest in Australia. The biennial Talisman Sabre exercises, a bilateral combined Australian and United States of America training activity, are held in the GBR with the majority of exercise activities taking place in the Shoalwater Bay Training Area (Department of Defence, 2019a). Both operational and training activities by the Australian Defence Force in the GBR region are expected to increase in the near future (GBRMPA, 2014; PGM Environment and Eco Logical Australia, 2014).

3. Contaminants of emerging concern

3.1. Antifouling paints

An 'antifouling system' (or antifoul) is defined as 'a coating, paint, surface treatment, surface, or device that is used on a ship to control or prevent attachment of unwanted organisms' (International Maritime Organization, 2005). Antifouling paints for use on ships' hulls generally contain metal oxides including copper (CuO), iron (FeO) or zinc (ZnO), and previously also arsenic and mercury (Almeida et al., 2007; Yebra et al., 2004). Copper is still widely used in antifouling paints although its application is being increasingly regulated following concerns about environmental impacts (Dafforn et al., 2011). Recent work has examined the use of CuO and ZnO nanoparticles in antifouling paints (Al-Fori et al., 2014; Anyaogu et al., 2008). Organic booster compounds (i.e. biocides) are added to antifouling paints to improve their efficacy (Almeida et al., 2007; Yebra et al., 2004). The detrimental environmental impacts of organotin compounds such as tributyltin (TBT) resulted in the adoption of the International Convention on the Control of Harmful Anti-Fouling Systems on Ships (the AFS Convention) in 2001 (International Maritime Organization, 2005). In Australia, the Convention is being enacted under the Protection of the Sea (Harmful Anti-fouling Systems) Act 2006 (Australian Government, 2013)². TBT, however, remains a CEC as TBT-based antifoulants (over-painted with contemporary coatings) are highly likely to be present on many ships traversing the GBR. This was highlighted by the severe TBT contamination of reef sediment following the grounding of the bulk coal carrier *Shen Neng 1* in the GBR in 2010 (GBRMPA, 2011). Following the adoption of the AFS Convention in 2001 (International Maritime Organization, 2005), eighteen compounds are currently used globally as antifouling biocides, namely benzomethylamide, chlorothalonil, copper pyrithione (CuPT), DCOIT (Sea-Nine 211), dichlofluanid, diuron, fluorofolpet, Irgarol 1051, Mancozeb, Polyphase, TCMS pyridine, TCMTB, thiram, Tolyfluanid,

² This Act stipulates that (i) organotin compounds that act as a biocide in an antifouling system are not to be applied to a ship in Australia, and (ii) non-complying ships are not to enter or remain in shipping facilities (i.e. a port, a shipyard, or an offshore terminal) in Australia.

Table 1

Land- and water-based sources for six different categories of contaminants of concern (CEC) known to be present, or likely to be present in the Great Barrier Reef (GBR) and Torres Strait (TS) marine ecosystems. Crosses indicate known or likely sources for each category of CEC, with shading indicating monitoring information exists (light grey) or does not exist (dark grey) for the GBR and TS regions. Empty cells indicate that a CEC is unlikely to be derived from a particular source. See Supplementary material for detailed monitoring information for each of the six CEC categories.

Sources	Antifouling paints	Coal dust and particles	Heavy/trace metals and metalloids	Marine debris	Pharmaceuticals and personal care products	Petroleum hydrocarbons
Agricultural land uses			x	x	x	x
Aquaculture	x		x	x	x	
Acid sulfate soils			x			
Urban land uses			x	x	x	x
Landfills			x	x	x	x
Wastewater treatment plants			x	x	x	
Mines (abandoned and current)		x	x			
Manufacturing and industrial areas			x	x		x
Commercial ports	x	x	x	x		x
Port anchorage areas	x		x	x	x	x
Marinas	x		x	x		x
Coastal/marine tourism	x		x	x	x	x
Shipping lanes	x		x	x	x	x
Shipping incidents	x	x	x	x		x
Shipping groundings	x	x	x			x

triphenylborane-pyridine, zinc pyrithione (ZnPT), zineb, and ziram (Almeida et al., 2007; Dafforn et al., 2011; Konstantinou and Albanis, 2004; Thomas, 2001; Voulvoulis et al., 1999; Yebra et al., 2004). Of these eighteen, seven (namely benzmethylamide, DCOIT (Sea-Nine 211), fluorofolpet, Irgarol 1051, polyphase, TCMS pyridine, and triphenylborane-pyridine) have not been registered or approved for use in Australia (Australian Pesticides and Veterinary Medicines Authority, 2015).

The main source of antifoulant in the GBR and TS marine ecosystems is shipping, principally large (>50 m) commercial and military vessels, commercial fishing and tourism boats, and recreational vessels (Table 1). Leaching and release of antifouling components from large vessels will occur while underway along shipping lanes, anchored in anchorage areas, and at berth in port (Fig. 3a–d). This could tally to substantial amounts, based on a total Cu leach flux of around 0.8 to 3.2 kg per day for an average bulk carrier for GBR coal ports (PGM Environment, 2012). It is likely to be much less for smaller vessels, but in high use areas such as marinas and ports could still be locally significant and contribute to the overall load of metals and biocides (Haynes and Loong, 2002). Catastrophic contamination of coral reefs in the GBR with antifouling components has occurred following groundings of large vessels (Fig. 3e), such as the *New Reach* in 1999, the *Bunga Teratai Satu* in 2000, and the *Shen Neng I* in 2010 (GBRMPA, 2011; Haynes et al., 2002; Haynes and Loong, 2002). This resulted in the release of antifouling components (including copper and TBT) several magnitudes higher than Australian and New Zealand default guideline values (DGVs)³ for sediment and marine water quality (ANZG, 2018) (Table S1). The effects of antifoul contamination may spread well beyond the ship grounding site depending on sediment transport and the mixing of local waters. The grounding of the *Bunga Teratai Satu* in 2000 was followed by extensive mitigation efforts to reduce the levels and extent of antifouling contamination (Marshall et al., 2002). More recently, the *Shen Neng I* caused the largest ship

grounding scar (approximately 400,000 m²) on the GBR resulting in severe contamination of the site with Cu, Zn and TBT (GBRMPA, 2011). Remediation is only just starting with an expected completion date of around 2022 (GBRMPA, 2019a). Ongoing monitoring has not occurred (or data are not publicly available) at any ship grounding site in the GBR or TS to determine the longer-term impacts of antifoul contamination, the effectiveness of any clean-up operations, and/or the potential for recovery of reef ecosystems.

In addition to shipping, antifouling paints are used on other marine infrastructure such as tourism-related structures (e.g. pontoons, jetties, underwater observatories, moorings) (GBRMPA, 2014). Further, antifouling paints are also used in commercial aquaculture operations (Fitridge et al., 2012) (Table 1, Fig. 2d), though no Queensland-specific information exists.

Components of antifouling paints, including metal oxides, TBT, and other booster biocides, have been detected in water and sediment of marine ecosystems in the GBR region (Table 1, S1a, b). Monitoring data for Cu, Zn as well as metals not associated with antifouling components is presented in section 3.3 ('Heavy/trace metals and metalloids'). A 1999 survey for the antifouling components Cu and TBT detected both in sediment at commercial harbours and mainland marinas, and Cu was detected at mooring sites at both mid-shelf islands and the outer reef (Haynes and Loong, 2002). More recent monitoring programs, associated with dredging and port developments, have demonstrated that TBT is still present in the water column and/or sediment in the five main ports (Department of Environment and Heritage Protection, 2012b; GHD Pty Ltd, 2005; Jones et al., 2005; Port Curtis Integrated Monitoring Program Inc., 2016; Port of Townsville Limited et al., 2013; Ports North, 2015). Exceedances of the DGVs for marine water quality for 95% species protection (0.006 µg Sn L⁻¹) and sediment quality (9 µg Sn kg⁻¹ dry weight, normalized to 1% organic carbon content) (ANZG, 2018) have been reported repeatedly for the 95% upper confidence limit (UCL) for TBT in water and sediment in Trinity Inlet (Cairns Port) (Ports North, 2015). Maximum TBT concentrations of individual sediment samples have exceeded the sediment DGV in all main ports (GHD Pty Ltd, 2005; GHD Pty Ltd, 2012c; Mortimer, 2004; Port of Townsville Limited et al., 2013; Ports North, 2015), except Gladstone (Department of Environment and Heritage Protection, 2012b). The biocides diuron

³ Guideline values to assess the quality of water and sediment for aquatic ecosystem protection are an integral component of the Water Quality Management Framework. This Framework 'provides users with a systematic approach to the management and assessment of water quality' for natural and semi-natural water resources. In the absence of more relevant guideline values, default guideline values can provide a generic starting point for the assessment of water and sediment quality.

and Irgarol 1051 have been detected in areas exposed to shipping (Ports North, 2015; Scarlett et al., 1999), with diuron regularly detected in sediment in Cairns Port with a maximum concentration of $5 \mu\text{g kg}^{-1}$ (Ports North, 2015). Monitoring information could not be found for the TS marine ecosystems or for biocides other than TBT, diuron and Irgarol 1051. Overseas studies suggest that at least some of these other biocides (e.g. Sea-Nine 211, dichlofluanid, chlorothalonil) are likely to be present including in ports, marinas and estuaries (Konstantinou and Albanis, 2004).

The effects of both acute and chronic exposures to antifouling paints have been examined for some GBR marine organisms. Toxicity of antifouling paints to hard and soft corals was observed near the grounding scar of the *Bunga Teratai Satu* on Sudbury Reef (Marshall et al., 2002). Laboratory exposures to Cu, Zn and TBT concentrations detected at GBR ship grounding sites found a significant reduction in coral fertilization and larval metamorphosis (Negri and Heyward, 2001; Reichelt-Brushett and Harrison, 2005; Victor and Richmond, 2005), coral recruitment (Negri et al., 2002), and major mortality of newly settled and coral branchlets (Smith et al., 2003). Comparable exposures in the field may thus delay the recovery of the resident coral community for many years unless the paint is removed (Negri et al., 2002; Smith et al., 2003). Acute exposure to the triazine herbicide Irgarol 1051 concentrations reflecting those detected in Caribbean marinas, harbours and coastal waters showed a reduction in photosynthesis of corals (Owen et al., 2002). Similarly, significant inhibition of photosynthesis was observed in coral symbionts exposed to environmentally relevant concentrations of diuron and Irgarol 1051 (Owen et al., 2003). More recent ecotoxicological tests on sub-tropical marine organisms showed that the effects of CuPT were comparable to that of TBT, while Irgarol 1051 was more toxic to autotrophic species than TBT (Bao et al., 2011).

Chronic exposure to antifouling paints has resulted in bioaccumulation of various biocides in marine plants and animals of GBR marine ecosystems (Table S1c). The biocide Irgarol 1051 was detected in seagrass tissue at concentrations considered to be potentially toxic in four of the five locations sampled along the GBR coast in 1997 (Scarlett et al., 1999). Butyltins were detected in muscle and liver tissue from silver trevally (*Caranx sexfasciatus*), stripey (*Lutjanus earponotatus*), black pomfret (*Apolectus niger*) and squid (*Loligo chinensis*) collected around Townsville in 1992 (Kannan et al., 1995). Bioaccumulation of TBT in oysters (scientific name not given) has been reported in Rosslyn Bay (Keppel Island), Cairns Port, and Gladstone Port (Mortimer, 2004). Around Port Curtis, TBT enrichment has been reported in oysters (*Saccostrea* spp.) and mud whelks (*Telescopium telescopium*) (Jones et al., 2005), with imposex being up to 43% in the whelk *Morula marginalba* in certain areas (Andersen, 2004). Corals are also sensitive to chronic, longer term exposures to low concentrations of TBT, diuron and Irgarol 1051 throughout their life history (Jones and Kerswell, 2003; Negri and Marshall, 2009; Watanabe et al., 2006). Diuron is also more commonly detected in the GBR lagoon as an agricultural contaminant and can affect all phototrophs, including seagrass at concentrations as low as $0.3 \mu\text{g L}^{-1}$ (Negri et al., 2015).

In summary, antifouling components including metal oxides (Cu, Zn) and biocides (TBT, diuron, Irgarol 1051) were reported in GBR marine ecosystems (Table S1a, b, c), including a biocide that has never been registered for use in Australia (Irgarol 1051). Recent monitoring has shown that TBT is still present in the water column and/or sediment in the five main ports, with overseas studies suggesting that other biocides are likely to be present. Locations where concentrations of metals and TBT exceeded DGVs for marine water quality and sediment quality include sites of ship groundings and coastal harbours. Given its phase-out the entry of TBT into the environment should eventually disappear but will remain a legacy issue for some time yet. In contrast, metal oxides are still being

used in antifouling components, released into the environment and likely accumulating in marine sediments. Disturbance of these sediments through activities such as dredging, ship movement, anchorage activity and cyclonic events, can thus be a potential source of toxic metals and legacy TBT. The lack of monitoring at GBR grounding sites following clean-up or no clean-up means that the long-term impacts of exposure to extreme concentrations of antifouling components are currently unknown. Designated port anchorages for bulk cargo and other trading vessels are also likely to contain elevated levels of metals and biocides, but monitoring information is not available for these areas. Given the substantial amounts of antifouling components that are leached continuously by bulk carriers along shipping lanes, at anchor, and at berth in port, as well as to a lesser extent around tourist and fishing areas, chronic exposure is likely to occur continuously throughout the year. This is of concern for habitat forming species such as coral and seagrass, and species for human consumption such as oyster, squid and fish.

3.2. Coal dust and particles

Coal is a combustible sedimentary rock which contains variable amounts and combinations of organic and inorganic constituents including polycyclic aromatic hydrocarbons (PAHs) and trace elements (e.g. Cd, Cu), respectively (Ward, 1984). Unburnt coal can be a potential source of these contaminants via leaching processes upon contact with water (Cheam et al., 2000), however, this process is highly variable. Trace metals and hydrocarbons are presented in more detail in sections 3.3 ('Heavy/trace metals and metalloids') and 3.6 ('Petroleum hydrocarbons'). The different types/ranks of coal vary in chemical composition and thus their energy content, use, and the potential for biological effects are different (Ahrens and Morrissey, 2005). Over the last three decades there has been a strong growth in Australia's mining sector due to global demand for coal and minerals. This growth stimulated increases in coastal development along the GBR, particularly related to port infrastructure and shipping activities (Grech et al., 2013). Australia is currently ranked first in global seaborne coal exports, with the country's largest reserves (~34 billion tonnes) found within Queensland (Department of Natural Resources and Mines, 2017b). Coal is transported by rail to the ports of Abbot Point, Gladstone, and Hay Point, all situated adjacent to the GBR WHA (Fig. 3a). Queensland coal exports through these three ports totalled 203 million tonnes for 2017 (Queensland Government, 2017a).

Unburnt coal enters the marine environment via a range of pathways such as the natural erosion of coal seams, and anthropogenic inputs during various stages of coal processing such as disposal of colliery waste, wind and water erosion of open coal stockpiles, accidental spillage, and coal carrier collisions and groundings (Ahrens and Morrissey, 2005) (Table 1). Colliery waste can enter river systems during major rainfall events when mines flood. In the GBR's Fitzroy River basin, all but two of the approximately 39 coal mines are permitted to discharge water under different environmental release conditions (Department of Environment and Resource Management, 2009). The likelihood of chronic coal exposure in the GBR marine ecosystems is greatest near the large coal export ports (Abbot Point, Gladstone, Hay Point) (Fig. 3a), where coal is stockpiled, conveyed and transferred. These coal stockpiles are not covered and coal dust and particles have the potential to be released into marine environments directly during windy conditions (GHD Pty Ltd, 2012a), cyclones (Department of Science Information Technology and Innovation, 2017) or heavy monsoonal rainfall (Queensland Government, 2019a). The concentrations of total suspended solids in stormwater runoff from coastal coal stockpiles into wetlands has been reported to be as

high as 33 mg L⁻¹ and 80 mg L⁻¹ in separate events (Department of Science Information Technology and Innovation, 2017; Queensland Government, 2019a). During a 2017 cyclone, concentrations as high as 800 mg L⁻¹ have been reported entering coastal waters adjacent to the GBR (Department of Science Information Technology and Innovation, 2017; Queensland Government, 2018). The total loads of suspended solids in these runoff events discharged into coastal waters are unknown. At Abbot Point, coal dust generation during offloading and accidental spillage during transfer activities has a probability of 95 to 100% to occur throughout a year (GHD Pty Ltd, 2012a), although no information about the actual amount lost is presented. Once in the marine environment, larger coal particles will settle close to the input source, while smaller particles have the potential to be carried long distances by currents (Jaffrenou et al., 2007; Johnson and Bustin, 2006). Oceanographic modelling suggests that coal particles on the ocean surface from coastal waters near Abbot Point could reach coral reefs off Mackay within three months under the influence of realistic wind conditions, tides and the East Australian Current (Andutta and Wolanski, 2012). Hence, an unknown proportion of the chronic coal contamination of waters in Queensland ports has the potential to migrate into GBR WHA waters. Chronic coal contamination is less likely in TS marine ecosystems due to their distance from coal port terminals. In the case of a collision or grounding (Fig. 3e), coal carriers could lose > 70,000 tonnes of coal (GBRMPA, 2011), representing a substantial potential source of coal to the marine environment.

Coal particles have been reported in sediments of port areas and adjacent coastal environments (e.g. wetlands) along the GBR coast (Table 1, S2). However, there is limited targeted monitoring and publicly available data relating to coal particles in GBR and TS marine ecosystems. For example, coal particles were not monitored in the Fitzroy River water quality assessment after the 2008 flood event (Tripodi and Limpus, 2010). Port development monitoring programs have measured sediment quality, including percentage coal particles, within coal ports and found varying sizes of coal particles in sediment samples in the proximity of loading facilities at Abbot Point (Toki et al., 2012; WBM, 2005). Coal particle concentrations ranged from nil at two reference sites 800 m east and west of the jetty, to <1 to 7% outside the berth area, and up to 45% below the coal-loading wharf. Surface sediments in wetlands adjacent to the Abbot Point coal stockpile contained up to 10% coal following a major cyclone-related discharge (Department of Science Information Technology and Innovation, 2017). PAHs with a pattern characteristic of coal have been measured in sediments and sediment traps in the central GBR region off the coast from Mackay and the Port of Hay Point coal terminals. Nearshore sediments contained the highest concentrations (~100–300 µg kg⁻¹ total PAHs), while offshore sediments as far as the continental shelf break contained PAHs with the same profile but at lower concentrations (Burns and Brinkman, 2011). Detection of elevated PAH concentrations (approximately 2,000 µg kg⁻¹ total PAHs) in sediment traps adjacent to midshelf reefs 65–85 nautical miles offshore (Burns and Brinkman, 2011) suggests the potential for widespread, albeit diluted transport within the GBR lagoon. However, hydrocarbon markers are not necessarily bioavailable or quantitatively correlated with coal particle contamination due to differences and uncertainties in leaching rates, transport and degradation.

The exposure and effects of both large coal spills and chronic coal inputs on marine organisms are largely unknown. Physical and/or chemical characteristics of coal particles have the potential to affect marine organisms (e.g. block light, clog, smother, etc.) (Ahrens and Morrissey, 2005). Accumulation of coal dust onto the upper and lower leaf/branch surfaces and trunks of mangroves growing around South Africa's largest coal-exporting port reduced photosynthesis by 17 to 39% (Naidoo and Chirkoot, 2004).

Controlled laboratory exposures of three taxa commonly found within the GBR (the hard coral *Acropora tenuis*, the reef fish *Acanthochromis polyacanthus*, the seagrass *Halodule uninervis*) to a wide range of coal concentrations over 28 d caused lethal effects in corals at relatively high concentrations ≥ 38 mg L⁻¹ (Berry et al., 2016). Reduced growth rates were reported for fish and seagrass at concentrations of 38 and 73 mg L⁻¹, respectively. Numerous early life history processes of the coral *A. tenuis*, such as fertilization and larval settlement, were substantially impaired by controlled exposures to a range of coal particle concentrations and scenarios (Berry et al., 2017). In contrast, coal leachate exposures during fertilization and larval settlement, and smothering of juvenile corals by coal had minimal effects. In general, studies on coal leachates report low concentrations of leached constituents and negligible bioavailability over relatively short periods (Bender et al., 1987; Berry et al., 2017; Jaffrenou et al., 2007).

In summary, coal particles and hydrocarbon markers potentially associated with coal particle contamination were detected in sediment nearby coal loading facilities at Abbot Point and in sediments of the central GBR, respectively. Monitoring information, however, is scarce despite the high likelihood of coal dust and particle generation at coal export ports (Abbot Point, Hay Point, Gladstone) and the large amount of coal exported annually through the GBR. The few studies that examined the effects of coal contamination on marine organisms suggest that the physical presence of coal particles is more detrimental than exposure to contaminants leached from coal. While these studies have identified thresholds for harm of coal particles to tropical marine species, the lack of available monitoring data makes assessing the risks posed by coal dust and particles to the GBR and TS marine ecosystems difficult to ascertain.

3.3. Heavy/trace metals and metalloids

Heavy/trace metals (hereafter metals) and metalloids occur naturally in rocks and soils; they enter the aquatic environment through weathering, erosion and atmospheric deposition. Human activities can augment the burden of metals [e.g. aluminium (Al), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), gallium (Ga), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), silver (Ag), tin (Sn), vanadium (V) and zinc (Zn)] and metalloids [e.g. antimony (Sb), arsenic (As) and selenium (Se)] in the environment (Stumm and Morgan, 1996). The presence of metals and metalloids in the water column usually reflects recent inputs, with many readily attaching to suspended particles and ultimately accumulating in sediments (Batley, 1995). Marine organisms accumulate metals and metalloids from the environment, with bioavailability and toxicity influenced by the metal/metalloid chemical form rather than total concentrations (ANZECC/ARMCANZ, 2000). Generally, ionic species are more bioavailable and toxic than those bound to particles and organic compounds. Notable exceptions include organic compounds of Hg and Sn, which are more toxic than their inorganic forms (Antizar-Ladislao, 2008; Boening, 2000). Although many metals and metalloids are essential micronutrients, all are potentially toxic to organisms above certain threshold concentrations (Batley, 1995; Kennish, 1997).

In the GBR and TS regions, sources such as agriculture and mining runoff, ports and harbours, drainage from acid sulfate soils (ASS), industrial effluents and emissions, atmospheric deposition, urban centres and wastewater discharge all contribute to elevated concentrations of metals and metalloids in receiving waters (Batley, 1995; Haynes and Johnson, 2000) (Table 1). Both abandoned (e.g. Mount Morgan gold mine, Horn Island gold mine) and operating mines (Fig. 2b) are potential sources of metals and metalloids to adjacent marine environment (GBRMPA, 2014;

Howley, 2012; Walker and Brunskill, 1996). In the northern and north-eastern regions of the TS, sediment contaminated with metals and metalloids is attributed to discharges from Papua New Guinean rivers, particularly the Fly River (Alongi et al., 1991; Baker and Harris, 1991; Baker et al., 1990; Gladstone, 1996; Haynes and Kwan, 2002; Heap and Scaffi, 2008). Metalliferous commodities and coal are handled and shipped in the ports of Abbot Point, Hay Point, Gladstone and Townsville, and metal-related industries such as refineries are located in Townsville and Gladstone (Department of State Development Infrastructure and Planning, 2014; Department of the Environment and Energy, 2019) (Fig. 3a). For both mines and refineries, an emerging issue is the large volume of water requiring emergency releases after high rainfall events (GBRMPA, 2014). Phosphate-based fertilizers and fungicides used in agricultural land uses in the catchments adjacent to the GBR (Fig. 1b) have been linked to elevated levels of As, Cd and Hg in GBR coastal sediments (Haynes, 2001; Walker and Brunskill, 1996). Both WWTP effluent discharge and landfill leachate are known to contain metals and metalloids (Batley, 1995; Kjeldsen et al., 2002), but no information is available for these sources in the GBR and TS regions. Disturbances of coastal ASS in the GBR and TS regions, generally related to agriculture, aquaculture and urban developments, will result in acidification, metal contamination, deoxygenation and iron precipitation in coastal receiving water (Cook et al., 2000; Powell and Martens, 2005; Waterhouse et al., 2013) (Fig. 2e), and subsequent remobilization of metals bound to particulate matter.

Metals and metalloids have been detected in water and sediment in the GBR and TS marine ecosystems, including the ports of Abbot Point, Cairns, Gladstone, Hay Point and Townsville (Table 1, S3, S4, S5). Based on available monitoring information⁴, dissolved metal and metalloid concentrations in GBR surface waters were typically low (Table S3a, S4a). Exceedances of DGVs for marine water quality (ANZG, 2018; Golding et al., 2015) have been reported for Al ($24 \mu\text{g L}^{-1}$), Cu ($1.3 \mu\text{g L}^{-1}$) and Zn ($15 \mu\text{g L}^{-1}$) (95th percentile concentrations) around the Port of Cairns (Ports North, 2015) and for Port Curtis (Andersen et al., 2005; Angel et al., 2010; Angel et al., 2012; Apte et al., 2005; Arango et al., 2013; Department of Environment and Heritage Protection, 2012a; Department of Environment and Heritage Protection, 2012b; Department of Environment and Resource Management, 2011; GHD Pty Ltd, 2009a; Mortimer et al., 2013; Vicente-Beckett et al., 2006; Vision Environment QLD, 2011). Rivers discharging into the GBR, such as the Fitzroy River, can contain elevated dissolved metals which could reflect a mixture of natural geological sources and industrial and other anthropogenic discharges (Apte et al., 2006). These contributions could add up to substantial amounts based on, for example, the highest average dissolved Al concentration ($64 \mu\text{g L}^{-1}$; Table S4a) (Port Curtis Integrated Monitoring Program Inc., 2016) and mean monthly discharges of 13,874 ML from the Calliope River (Queensland Government, 2016e). Similarly, exceedances that are one order of magnitude greater than relevant DGVs have been reported for several metals in marine waters receiving discharge from highly disturbed ASS, such as East Trinity near Cairns (Cook et al., 2000). Exceedances of DGVs for sediment quality (ANZG, 2018) have been reported from several GBR ports, as well as the TS region (Table S3b, S4b), including for 95% upper confidence limits for As, Hg, Ni, Pb and Se concentrations in the Cairns and Townsville Ports (Port of Townsville Limited et al., 2013; Ports North, 2015), and for individual samples for As, Cr, Ni, Hg and Pb from the Endeavour

River estuary, Port of Abbot Point and Port Curtis (Angel et al., 2012; GHD Pty Ltd, 2012b; Howley, 2012; Jones et al., 2005), and for As, Cr, and Ni in the Gulf of Papua and northern TS (Haynes and Kwan, 2002). In Townsville Port, exceedances were associated with shipping ores and refined materials containing Cu, Ni, Pb and Zn (Reichelt and Jones, 1994). Near Cairns, estuarine sediments impacted by ASS drainage can also be enriched with As, Cr, Fe, Ni, Pb and Zn (Broughton, 2008; Hicks et al., 1999; Johnston et al., 2010; Keene et al., 2010).

Metals and metalloids have been detected in a variety of marine organisms from the GBR and TS marine ecosystems, including in remote locations as well as near urban and industrial locations (Table S3c). Elevated metals (e.g. Cd, Co, Cu, Fe, Mn, Mo, Ni, Hg, Pb, Zn) and metalloids (e.g. As, Sb, Se) likely due to anthropogenic activities have been detected in seagrass (*Zostera capricorni*), crayfish (*Panulirus ornatus*), oysters (*Crassostrea echinata*, *Saccostrea amasa*, *S. commercialis*, *S. echinata* and *S. glomerata*), mud whelk (*Telescopium telescopium*), mud crab (*Scylla serrata*), tuxedo crab (*Australoplax tridentata*), fish (e.g. barramundi, *Lates calcarifer*; parrotfish, *Scarus dimidiatus*; mullet *Valamugil seheli*), dugong (*Dugong dugon*) and green turtle (*Chelonia mydas*) (Gladstone, 1996; Haynes et al., 2005; Jones et al., 2005; Mortimer, 2000; Negri et al., 2009; Port Curtis Integrated Monitoring Program Inc., 2016; Prange and Dennison, 2000; World Wildlife Fund, 2016). Increased bioaccumulation of Ag, Cd, Cu, Fe, Mn and Zn in oysters (*S. amassa*) coincided with increased bioavailability of metals during *Trichodesmium* blooms (Jones, 1992). Some concentrations of As, Cu, Se and Zn in mud crabs and oysters exceeded food code standards for these elements in crustaceans and molluscs (Food Standards Australia New Zealand, 2015) (Table S3c).

Metals can exert sub-lethal and toxic effects at concentrations measured in GBR and TS marine ecosystems. A reduction in growth has been reported following exposure to environmentally relevant metal concentrations, for the tropical microalga *Isochrysis galbana* (Cu) (Trenfield et al., 2015), the tropical dogwhelk *Nassarius dorsatus* (Cu, Al) (Trenfield et al., 2016), and for the marine diatom *Ceratoneis closterium* (formerly *Nitzschia closterium*) (Al) (Harford et al., 2011). Tissue mortality and effects on symbiosis were observed in tropical sponges experimentally exposed to Cu (Webster et al., 2001). In corals, exposure to metals such as Cd, Ni, Pb, Zn, and Cu in particular, can result in photosynthesis reduction in algal symbionts, zooxanthellae loss and changes in oxygen consumption (Bielmyer et al., 2010; Reichelt-Brushett and Harrison, 2005), as well as adversely affecting fertilization success, larval motility, larval settlement success and metamorphosis (Negri and Heyward, 2001; Negri et al., 2002; Reichelt-Brushett and Hudspeth, 2016; Reichelt-Brushett and Harrison, 1999; Reichelt-Brushett and Harrison, 2000; Reichelt-Brushett and Harrison, 2005; Reichelt-Brushett and Michalek-Wagner, 2005; Reichelt-Brushett and Harrison, 2004; Victor and Richmond, 2005). Embryo and larval development in oyster (*Saccostrea echinata*), mussel (*Mytilus edulis planulatus*) and barnacle (*Amphibalanus amphitrite*) are affected at Al concentrations that have been recorded in mid Boyne estuary and mid Calliope estuary (van Dam et al., 2016). Elevated concentrations of Co, Mn and Sb in green turtles (*Chelonia mydas*) correlate with clinical markers of inflammatory response and liver dysfunction (World Wildlife Fund, 2016). Several metals, such as Cd, Mn, and Hg (particularly methylmercury) are well-established endocrine disrupting chemicals (Kortenkamp, 2011; United Nations Environment Programme and the World Health Organization, 2013), and exposures to Cd, Hg, Pb, Se, and Zn have been implicated in the endocrine disruption of marine invertebrates (Depledge and Billingham, 1999). These metals and metalloids have been detected in marine organisms in the GBR and TS marine environments (da Silva et al., 2004; Denton and Burdonjones, 1986a, 1986b, 1986c; Haynes et al.,

⁴ For the commercial ports, monitoring information was typically obtained from the scientific and grey literature, and environmental assessment reports. The Port Curtis Integrated Monitoring Program also provided monitoring data specifically for this study. Monitoring data was requested but not made available by any of the other port corporations (North Queensland Bulk Ports, Ports North, Port of Townsville).

2005; Haynes and Johnson, 2000; Jones et al., 2005; Mortimer, 2000; Negri et al., 2009; Rayment and Barry, 2000; Berry et al., 2013; Gladstone, 1996) and could potentially disrupt endocrine systems in these species. Discharge of ASS containing Al and Fe can acidify receiving waters (Cook et al., 2000) potentially affecting migration of fish and invertebrate species (Kroon, 2005), and result in the formation and sedimentation of Al and Fe flocs that can smother benthic communities (Cook et al., 2000). Discharges from ASS have been linked to the seasonal occurrence of epizootic ulcerative syndrome in fish (Callinan et al., 1996; McClurg et al., 2009a; McClurg et al., 2009b; Powell and Martens, 2005), and extensive fish kills have been reported in both naturally (Brown et al., 1983) and artificially (Callinan et al., 1996; Dawson, 2002) drained ASS catchments in north-eastern Australia. Fish kills have also been reported for many GBR rivers, however, a causal link with ASS discharge has not been determined (McClurg et al., 2009a; McClurg et al., 2009b).

In summary, a range of sources related to human activities have contributed to elevated metal and metalloid concentrations in GBR and TS marine ecosystems. Locations where concentrations exceeded DGVs for sediment and marine water quality and/or food code standards include most of the five main ports, East Trinity near Cairns, the Endeavour river estuary, the TS region and the Gulf of Papua (Table S3, S4, S5). Only one port (Port Curtis) in the GBR provided monitoring data; a more complete assessment of metal/metalloid contamination would benefit from the contributions of all data custodians. Elevated levels of metals and metalloids were reported for a range of marine organisms in the GBR including species for human consumption (Table S3c). In addition, sub-lethal effects have been detected for reproduction, development and growth for a range of marine species following exposure to environmentally relevant concentrations of metals/metalloids. Other potential effects of metal and metalloid contamination on, for example, migration of fish and invertebrates, fish diseases and mortality, and disruption of endocrine systems, have been postulated but not been confirmed for the GBR and TS marine environments.

3.4. Marine debris

Marine debris (or marine litter) is defined as 'any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment' (United Nations Environment Programme, 2009). Marine debris originates from sea or ocean-based sources such as shipping, fishing and aquaculture, and oil and gas platforms and rigs, and land-based sources including landfills, sewage and storm water, and industrial effluent (United Nations Environment Programme, 2009). Around the world, plastic items are generally the most common type of marine debris (United Nations Environment Programme, 2009). To prevent and minimize marine debris pollution from ships, the Regulations for the Prevention of Pollution by Garbage from Ships (Annex V) entered into force 31 December 1988 under the International Convention for the Prevention of Pollution from Ships (MARPOL) (International Maritime Organization, 2019). Annex V imposes a complete ban on the disposal into the sea of all forms of plastics. In Australia, MARPOL is being implemented through the Protection of the Sea (Prevention of Pollution from Ships) Act 1983 and the Navigation Act 2012 (Australian Maritime Safety Authority, 2018a). Plastics, however, remain a contaminant of concern as exemplified by the fact that approximately three-quarters of marine debris along the Australian coastline is plastic with the remainder consisting of glass, metal, paper and wood (Hardesty et al., 2016; Hardesty et al., 2014). Globally the presence of oceanic microplastics (i.e. plastic items < 5.0 mm in size (Masura et al., 2015)) has been identified as an emerging issue of international

concern (GESAMP, 2016), and contamination in Australian marine waters is ubiquitous (Reisser et al., 2013).

Only a few studies have specifically examined the sources of marine debris pollution in the GBR and TS regions (Griffin, 2008; Hardesty et al., 2014; Haynes, 1997; Jensen et al., 2019) (Table 1). Most of the marine debris found on islands and cays in the Far Northern Section of the GBR is likely derived from oceanic and local shipping sources (Haynes, 1997) (Fig. 3a-d). Shipwrecks can be a major source of marine debris including in remote locations (Schiel et al., 2016), however debris contamination following groundings in GBR or TS waters (Fig. 3e) has not been assessed. On the other hand, high concentrations of floating marine plastics in GBR waters between Shoalwater Bay and Townsville in February 2013 were associated with large flooding events (Hardesty et al., 2014) due to Ex-Tropical Cyclone Oswald (Bureau of Meteorology, 2014), and thus most likely derived from land-based sources (Fig. 1b, 2a-d). The paths of many satellite-tracked global drifters suggest that the South Pacific and Coral Sea are at least a contributing source of marine debris washing up on the northern GBR and TS marine ecosystems (Griffin, 2008). Fragmentation of larger items is a likely source of marine microdebris in the GBR and TS regions, rather than specifically manufactured items such as microbeads or pre-production plastic pellets (Hall et al., 2015; Jensen et al., 2019; Kroon et al., 2018; Reisser et al., 2013). Most microfibrils detected in GBR waters and coral reef fish were deemed to be of textile origin likely derived from clothing and furnishing (Jensen et al., 2019; Kroon et al., 2018). Potential sources for microdebris range from riverine discharge for inshore reefs, and (un-)intentional discard and wastewater discharges from vessels in the GBR, to long-range atmospheric and oceanic transport for offshore reefs (Critchell et al., 2015; Jensen et al., 2019). Given the wide range of potential sources of marine debris it is likely that exposure in the GBR and TS ecosystems is widespread and continuous. Indeed, repeated clean-ups at the same GBR beach show that marine debris is present continuously, with strong seasonal variation (Kroon et al., 2015).

Marine debris has been detected along the coastlines and in marine waters of the GBR and TS (Table 1, S6). The presence of marine debris, including plastics, in the GBR Marine Park was first documented in the scientific literature in 1997 (Haynes, 1997). More recently, a total of ~1.8 million items of marine debris were collected in 1,121 beach clean-ups from January 2008 to October 2015 in the GBR and TS regions (Tangaroa Blue Foundation, 2015). These items were classified as plastic (81%), foam (6%), glass and ceramic (4%), rubber (4%), and metal (3%) (Kroon et al., 2015; Tangaroa Blue Foundation, 2014). Standardizing the marine debris data from 606 of these beach clean-ups to 100 m (Schulz et al., 2015) revealed that plastics were the most abundant item (58% to 91%) across all seven NRM regions overlapping the GBR and TS marine ecosystems (Kroon et al., 2015). Specifically, the abundance of plastic items ranged from an average of 398 to 1,558 items per 100 m of beach cleaned. Other items such as glass, metal and cloth comprised a relatively large proportion (19% to 33%) in some but not all the NRM regions. Smaller plastic particles, including microplastics, were first reported in surface waters of the GBR and TS, at concentrations ranging from 1 to 80,000 pieces per km², during surveys in September 2012 and February 2013. The highest concentrations (40,000 to 80,000 pieces per km²) were recorded between Shoalwater Bay and Townsville in February 2013 (Hardesty et al., 2014). Subsequent studies have confirmed microdebris contamination, including microplastics, of (sub-) surface waters in the central GBR (Hall et al., 2015), with contamination more prevalent near offshore compared to inshore reefs (Jensen et al., 2019). More than half of the items detected contained synthetic (i.e. plastic) polymers, the most common ones being polyester, nylon, and polyethylene (Jensen et al., 2019).

In the GBR and TS marine ecosystems, risks related to marine debris have known and potential impacts on matters of national environmental significance and Outstanding Universal Value (Ceccarelli, 2009; GBRMPA, 2014). Both entanglement and ingestion have been reported for turtles, cetaceans, dugong, and seabirds, including for species that are already threatened, vulnerable, endangered, or critically endangered (Ceccarelli, 2009). Based on evidence from overseas studies (Gall and Thompson, 2015; Wright et al., 2013), the impacts are likely to be much more widespread and include many more marine species than currently documented. For example, two recent studies reported a high frequency of occurrence of microdebris ingestion, including microplastics, in two species of coral reef fish captured in the GBR WHA (Jensen et al., 2019; Kroon et al., 2018). The effects, detrimental or otherwise, of physical, chemical and/or microbial exposure associated with the ingestion of marine debris on wild fish populations in the GBR and TS marine ecosystems are currently unknown. Micro-organisms and invertebrates have been detected on floating marine plastic in GBR waters (Reisser et al., 2014), representing a new dispersal pathway for marine organisms. Furthermore, disease on reef building corals increased with the presence of plastic waste (Lamb et al., 2018), including derelict fishing line as reported for the GBR (Lamb et al., 2015). Other potential ecosystem effects, such as changes of habitat and species assemblages, introduction of invasive species and alteration of marine food webs (Browne et al., 2015) have not been examined in GBR and TS marine ecosystems.

In summary, marine debris was reported for coastal and island beaches across GBR and TS marine ecosystems (Table S6). Across these different environments, plastics comprise by far the most common item of marine debris, followed by foam, glass and ceramic, rubber and metal. Locations where marine debris accumulates have not been determined but are likely to be influenced by the physical characteristic of the source location, the rate of degradation, and prevailing water currents and wind drifts. Both entanglement and ingestion have been reported in the GBR and TS regions and have known and potential impacts on matters of national environmental significance and Outstanding Universal Value. In addition, recent studies have demonstrated the prevalence of marine microdebris, including microplastics, in (sub-)surface waters and coral reef fish in the GBR. However, a lack of monitoring data across GBR and TS marine ecosystems means that the long-term impacts of exposure to marine debris are currently unknown. This is of concern for habitat forming species such as coral, species that are already threatened, vulnerable, endangered, or critically endangered, and species for human consumption such as coral trout.

3.5. Pharmaceuticals and personal care products

Pharmaceuticals comprise a broad range of human and veterinary medical products used for the diagnosis, treatment or prevention of various physical and mental conditions (Daughton and Ternes, 1999; World Health Organization, 2003). Many different conventions are used for the classification of pharmaceuticals, including systems based on their biological modes of action, chemical structures or intended use (World Health Organization, 2003). Examples of some major pharmaceutical classes include: antibiotics, psychotropic drugs (e.g. anti-depressants, mood stabilizers), non-steroidal anti-inflammatory drugs (NSAIDs), anti-hypertensives and anti-cholesterolemic, and anti-convulsants, as well as illicit (recreational) drugs. Personal care products (PCPs) comprise a large array of consumer products generally used on the human body, such as cosmetics, toiletries or fragrances that contain active ingredients to prevent diseases or alter odour, appearance, touch or taste (Daughton and Ternes, 1999). Similar

to pharmaceuticals, PCPs are represented by a broad range of classes of compounds, both in terms of function and physicochemical properties (Molins-Delgado et al., 2015). Some of the common classes of PCPs include surfactants and their bioactive transformation products (e.g. nonylphenol, quaternary ammonium compounds), disinfectants and antimicrobials (e.g. parabens, triclosan), insect repellents (e.g. N,N-diethyl-*meta*-toluamide/DEET), musks and fragrances (e.g. galaxolide, tonalide), UV filters (e.g. benzophenone, octocrylene), plasticizers (e.g. bisphenol A (BPA), diethylhexylphthalate (DEHP), phthalates), anticorrosives (e.g. benzotriazoles) and food products (e.g. artificial sweeteners, caffeine). Pharmaceuticals and personal care products (PPCPs) can be considered high volume products (produced in thousands of tons) that are released into waste streams from population centres (e.g. WWTPs) or at sites associated with human activities (e.g. shipping, tourism or recreation activities) (Kaplan, 2013). Due to their potential ecotoxicological effects even at low concentrations (Christen et al., 2010; Daughton and Ternes, 1999; Melvin, 2015; Melvin et al., 2014; United Nations Environment Programme and the World Health Organization, 2013), PPCPs are now widely recognized as emerging threats to aquatic animals and ecosystem health (Boxall et al., 2012; Rudd et al., 2014), including marine ecosystems (Minguez et al., 2014).

The main sources of PPCPs in the GBR and TS marine ecosystems are wastewater discharges from WWTPs into (rivers that are connected to) coastal waters and from vessels discharged directly into marine waters (Table 1). Wastewater discharge generally consists of untreated or treated sewage (i.e. drainage from toilets, medical premises and spaces containing living animals) and greywater (i.e. drainage from showers, sinks and laundries). The distribution of the human population and locations of WWTPs (Fig. 2a) in the region, combined with the distribution of vessel traffic associated with bulk shipping (Fig. 3a), commercial fishing (Fig. 3b), commercial and recreational tourism (Fig. 3c) and Australian Defence Force operations (Fig. 3d), would therefore have the greatest influence on the presence of PPCPs in the GBR and TS marine ecosystems. Dilution following discharge from land-based WWTPs is likely to be significant, greatly reducing concentrations in the GBR and TS marine environments. For example, data for Sydney Harbour revealed relatively low (ng L^{-1}) concentrations of all measured compounds despite its proximity to a major urban centre (Birch et al., 2015). Discharge for treated and untreated sewage in the GBR Marine Park is generally permitted with restrictions around certain vessels (e.g. cruise ships; GBRMPA, 2018) and distances to a person in the water, to an aquaculture fisheries resource, and to a reef, island or the mainland (Maritime Safety Queensland, 2019). Discharge estimates for a large cruise ship (3,000 passengers and crew) are 800,000 L for sewage and 4,000,000 L for greywater for a typical one-week voyage (Copeland, 2005). Although dilution is also likely to be significant, the lack of monitoring information in receiving waters means that potential environmental risks from PPCP loads in wastewater discharges from land-based WWTPs (Fig. 2a) and from the combined vessel traffic (Fig. 3a-d) cannot be discounted. In addition, production of livestock such as cattle, pigs and poultry and land-based aquaculture in north Queensland present potential sources of veterinary pharmaceuticals, especially antibiotics (Gaw et al., 2014; Sarmah et al., 2006) (Table 1). Landfills and seafills, urban storm water and septic tanks can also leach PPCPs into receiving waters particularly in high rainfall areas (Gaw et al., 2014; Masoner et al., 2014). Finally, direct inputs of PPCPs into the marine environment in high use areas for tourism and recreational activities are also likely (Brausch and Rand, 2011). Approximately two million people visit the GBR annually using commercial tourist operators, with the Cairns and Whitsunday planning areas receiving approximately one million visitations each (GBRMPA, 2019b).

Direct input represents a potential source of PPCPs in areas of in-water activities in close proximity to coral reefs (e.g. snorkelling areas), including through leaching after external applications (e.g. sunscreens, perfumes). For example, two sunscreen applications per day by 2,700 visitors in the Cairns planning area could result in the release of 4,050 g of organic UV filters across the multiple reefs visited during that day (Danovaro et al., 2008; Kroon et al., 2015; Therapeutic Goods Administration, 2016)⁵. The final concentration would depend on the volume of water surrounding a reef and associated extent of dilution in the marine environment. At Hawaii's Waikiki Beach, seawater concentrations of the common UV-blocker benzophenone-3 reached a maximum of $0.136 \mu\text{g L}^{-1}$ (Mitchellmore et al., 2019).

Limited monitoring data are available on PPCPs in or around the GBR and TS marine ecosystems, and most of this is reported in treated domestic wastewater (O'Brien et al., 2014) rather than actual environmental concentrations (Scott et al., 2014) (Table 1, S7). At the time of this review, only these two studies had been published providing information for just 29 products, measuring effluent from two of the >50 operational WWTPs (Hill et al., 2012) (Fig. 2a) and receiving waters in two out of the 35 river basins discharging into the GBR (Fig. 1a). PPCPs measured in treated sewage were at concentrations between 10 and 500 ng L^{-1} , which is consistent with storm water and WWTP effluent concentrations measured in other Australian studies not located in the GBR and TS region (French et al., 2015; Sidhu et al., 2012). Exceptions include several PPCPs observed in the Wet Tropics region, including the artificial sweetener acesulfame ($4.4 \mu\text{g L}^{-1}$), the anticonvulsant gabapentin ($1.8 \mu\text{g L}^{-1}$), the diuretic hydrochlorothiazide ($1.1 \mu\text{g L}^{-1}$), the X-ray contrast agent iopromide ($2.3 \mu\text{g L}^{-1}$), the analgesic tramadol ($2.0 \mu\text{g L}^{-1}$) and the psychotropic venlafaxine ($1.2 \mu\text{g L}^{-1}$) (O'Brien et al., 2014). In river water, paracetamol was reported in the Fitzroy region at $4.1 \mu\text{g L}^{-1}$ (Scott et al., 2014). Many of these measurements come from a single sampling event, so the reliability of these concentrations as representative for these regions is limited. A recent national analysis of WWTP effluent representing 48% of Australia's population demonstrated loads of total UV-filters (5 organic UV-filters) of $819 \pm 647 \text{ mg per 1,000 person per day}$ and a maximum UV-filter effluent concentration of $8.48 \mu\text{g L}^{-1}$ (O'Malley et al., 2020). Of the measured UV filters being released into the environment through effluent, Ensulizole and Sulisobenzone were the most prevalent. The WWTP sites were not named but loads were highest in tropical locations which may include the GBR. The number of PPCPs entering the GBR and TS coastal environments through WWTP and storm water discharges, as well as directly entering the marine environment through vessel discharges and direct input in areas of in-water activities is likely to be vastly greater. For example, an additional 39 pharmaceuticals have been detected in Australia-wide monitoring studies in wastewater and receiving environments (Kroon et al., 2015), and 13 licit (e.g. alcohol, nicotine, oxycodone, fentanyl) and illicit (e.g. cocaine, methylamphetamine) drug types have been detected in sewage inflow at WWTPs located in Queensland (Australian Criminal Intelligence Commission, 2018).

Many PPCPs are persistent (e.g. artificial sweeteners, X-ray contrast agents), bio-accumulative (e.g. synthetic fragrance musks, some UV filters) and toxic (e.g. antimicrobials) (Molins-Delgado et al., 2015), and may therefore impacts on the health of receiving marine ecosystems. We are not aware, however, of any studies

examining the effects of PPCPs on marine organisms in the GBR and TS ecosystems. Given the potentially large number of PPCPs present in these systems, we used hazard quotients (HQs; highest measured or predicted environmental concentration, i.e. MEC or PEC, divided by the predicted no-effect concentration, i.e. PNEC) for WWTP effluent to assess the potential ecological risks associated with PPCPs (Kroon et al., 2015). We followed an established framework (Cristale et al., 2013; Sánchez-Avila et al., 2009), where $\text{HQ} < 1$ indicates no risk, $1 \leq \text{HQ} < 10$ represents a low risk, $10 \leq \text{HQ} < 100$ signifies a high risk, and $\text{HQ} \geq 100$ indicates that adverse outcomes are expected. These HQ values are indicative only as (i) limited effluent concentration data for WWTPs were available for the GBR, (ii) additional effluent concentration data was used from other WWTPs in Australia, and (iii) many of the derived PEC and PNEC data have a great deal of uncertainty related to them, including being derived from freshwater species. PPCPs identified with $\text{HQ} \geq 1$ are the antibiotic salinomycin ($\text{HQ} = 1,700$), the antimicrobials benzalkonium chloride (21) and triclosan (23 water; 450 sediment), the statin atorvastatin (18), the fibrate gemfibrozil (5.8), the musk fragrance galaxolide (4.2), a non-ionic surfactant by-product nonylphenol (2.3), the psychotropic amitriptyline (1.9), the analgesic tramadol (1.2), and the psychotropic venlafaxine (1.2) (Table S7). While the actual ecological risks of these PPCPs following dilution in the marine environment may be significantly lower, paucity of monitoring data including from sources other than WWTPs means that HQs cannot be reliably estimated for the GBR and TS marine environments. In addition, numerous studies have demonstrated sub-lethal outcomes in marine organisms from exposures at ng L^{-1} concentrations, such as behavioural effects (Fong and Ford, 2014), antibiotic resistance (Zhu et al., 2017) and endocrine disruption (United Nations Environment Programme and the World Health Organization, 2013), which can also have implications for higher-level biological processes (e.g., survival, health and population fitness) (Ankley et al., 2010; Groh et al., 2015). For example, the UV-filter benzophenone-3, commonly found in sunscreens and cosmetics, has been reported to affect coral larvae at moderately low concentrations of $6.5 \mu\text{g L}^{-1}$ (Downs et al., 2016), coral bleaching at very high concentrations of $10,000 \mu\text{g L}^{-1}$ (Danovaro et al., 2008), and mortality across a range of high concentrations ($49\text{--}1,000 \mu\text{g L}^{-1}$) (Downs et al., 2016; He et al., 2019). These and other PPCPs can directly enter the marine environment in tourist hotspots, including popular reef viewing areas, which highlights the need for better monitoring of PPCPs in the GBR and TS marine environments. Given the large number of potential PPCPs present, such monitoring should be informed by predictive methods for risk prioritization to develop a short-list of potentially high-risk compounds (Dong et al., 2013; King et al., 2015).

In summary, PPCPs were reported for treated domestic wastewater from two WWTPs in Cairns, and in receiving waters of the Fitzroy River. We are not aware of any other available monitoring information on PPCPs for water, sediment or biota in the GBR and TS marine environments. Indeed, a recent review of worldwide pharmaceutical risks to marine environments demonstrates the extremely limited monitoring data available for Australia compared to other parts of the world (Fabbri and Franzellitti, 2015). Locations where concentrations of PPCPs may pose potential ecological risks include marine environments receiving discharge from land-based WWTPs (Fig. 2a) and from the combined vessels in high traffic shipping lanes and at designated port anchorages (Fig. 3a–d) and discharge areas, as well as around tourist hotspots with in-water activities in close proximity to coral reefs (e.g. snorkelling areas). The preliminary estimates of HQ values, developed in lieu of any effect studies on marine organisms in the GBR and TS ecosystems, would suggest that at least some PPCPs in wastewater discharges may be causing adverse effects at the most contami-

⁵ Based on (i) 2,700 visitors per planning area per day (~ one million visitors annually) (Great Barrier Reef Marine Park Authority, 2019b), (ii) two applications of sunscreen per person per day, of which ten g can be released into the environment (Danovaro et al., 2008), and (iii) the maximum concentration (15%, w/w) permitted for organic UV filters (Therapeutic Goods Administration, 2018).

nated sites. Hence, targeted studies that examine environmental concentrations and potential effects of chronic exposure to potentially high-risk PPCPs in GBR and TS marine ecosystems are warranted.

3.6. Petroleum hydrocarbons

Petroleum hydrocarbons are compounds that contain carbon and hydrogen atoms in a vast array of molecular combinations. The most common hydrocarbons are petroleum crude oils, fuels or other refined petroleum products, which contain various proportions of alkanes (paraffins), cycloalkanes (naphthenes) and aromatic hydrocarbons containing one or more aromatic rings (Petrov, 2012). Petroleum hydrocarbons that contain one to eight carbon atoms are gases or highly volatile liquids and persist in the marine environment for only hours to days, while higher molecular weight hydrocarbons are less volatile and can persist on or in seawater or sediments for days to months (Gong et al., 2014). These larger, more persistent petroleum hydrocarbons generally have low solubility in water and following surface spills initially float on the seawater surface and later become associated with sediments (Gong et al., 2014). Petroleum hydrocarbons can also become entrained in the water column as oil droplets due to the wave action on oil slicks at the water surface, by high current velocities, or due to pressurized releases from underwater wellhead blowouts or pipeline ruptures (Reed et al., 1999). The components of petroleum hydrocarbons that are most hazardous to marine life are the aromatic hydrocarbons including the monocyclic hydrocarbons (MAHs) and PAHs (French-McCay, 2002). Indeed, petroleum hydrocarbons are often reported for monitoring purposes as total PAHs associated with aquatic sediments because this component is the most toxic and can be relatively persistent (French-McCay, 2002).

The presence of petroleum hydrocarbons in the GBR and TS regions is associated with natural sources, runoff and discharge from industry and urban sources, atmospheric deposition, vessel and port operations, and shipping incidents and accidents (Det Norske Veritas Ltd, 2013; Haynes and Johnson, 2000; Waterhouse et al., 2013) (Table 1). Oil exploration or extraction does not occur in the GBR and TS regions. Numerous natural sources of hydrocarbons exist in the marine environment, including seepage from sub-surface reservoirs (Hornafius et al., 1999), and recent biogenic synthesis by plants and microorganisms (Blumer et al., 1971). Hydrocarbons from natural sources such as terrestrial plant material have been identified in an inner GBR sediment sample (Shaw and Johns, 1985), but their presence in the GBR and TS marine ecosystems more broadly has not been assessed. Similarly, atmospheric deposition of burnt fuels and bushfires has not been quantified. Spikes in petroleum and natural sources of hydrocarbons may occur after cyclones due to excess runoff (Sandstrom, 1988). Activities that can contribute to contamination within ports include direct discharge of burnt fuel and lubricants from vessels, loading of cargo (e.g. coal which contains PAHs), and loss from coal stockpiles (Maher and Aislabie, 1992; Queensland Government, 2016b; Smith et al., 1985) (see section 3.2 'Coal dust and particles'). Most petroleum hydrocarbon contamination around ports from these sources is likely to accumulate in sediments close to loading facilities (ANZG, 2018; GHD Pty Ltd, 2009b; WBM, 2005). Shipping activities have resulted in >500 spills of diesel oil, hydraulic oils and heavy fuel oils in ports, coastal waters and the GBR from 2002 to 2016, with half of these occurring within ports or port limits (Queensland Government, 2016b). The largest spills include 25 tonnes of heavy fuel oil from the *Global Peace* within Gladstone Harbour (18 tonnes recovered) after a collision in 2006 (Andersen et al., 2008; Melville et al., 2009); 14 tonnes of diesel fuel from a sinking tourist catamaran off Cape Tribulation in 2013 (Queensland Government, 2016b); 10 to 15

tonnes from an unnamed ship in 2015 off Cape Upstart (Queensland Government, 2016d); and up to 4 tonnes of fuel oil from the *Shen Neng I* which ran aground on Douglas Shoal off Gladstone in 2010 (GBRMPA, 2011). In 1970 the tanker *Oceanic Grandeur* collided with an uncharted rock in the heavily transited Torres Strait, losing 1,100 tonnes of crude oil (Australian Maritime Safety Authority, 2018b). The largest plausible spills in the GBR and TS marine environments would be from large oil tankers which could potentially lose over 50,000 tonnes (all tanks ruptured) of refined or crude oil, while large general cargo vessels could lose up to 5,000 tonnes of fuel oil (Australian Maritime Safety Authority, 2015; Burgherr, 2007). Recent risk assessments indicate 'total oil spill risk' annually within the GBR and TS regions of 418 tonnes petroleum hydrocarbons from all ship and accident types (Det Norske Veritas Ltd, 2013). This risk was projected to more than double by 2032 based on expected increases in ships transiting the region (PGM Environment, 2012), including passages of very large coal vessels and liquefied natural gas (LNG) tankers (North-East Shipping Management Group, 2014). Locations facing the highest risk of major oil spills are those where shipping traffic is highest (Fig. 3a–d), including the West Torres Strait, North and Middle Inner Route, north of Yeppoon and Hydrographers Passage.

Across the GBR and TS marine environments, total PAHs are not routinely monitored except as part of the environmental planning and management activities for port and dredging developments (Table S8a, b, c). When sampling has been performed in diverse locations across the GBR, total PAH concentrations in the sediments was highest in the larger multi-use ports (total PAH generally less than < 200 $\mu\text{g kg}^{-1}$ but some samples up to 13,400 $\mu\text{g kg}^{-1}$), followed by rivers with urban inputs (<200 $\mu\text{g kg}^{-1}$) and island locations visited by small boats (<25 $\mu\text{g kg}^{-1}$), and finally undetectable at offshore coral reefs (Smith et al., 1987; Smith et al., 1985) (Table S8b). Given that the Australian and New Zealand DGV for sediment quality for total PAHs of 10,000 $\mu\text{g kg}^{-1}$ (normalized to 1% OC) (ANZG, 2018) has rarely been exceeded, total PAHs have been considered a low risk to the highly disturbed port environment by port operators (GHD Pty Ltd, 2009b; North Queensland Bulk Ports Corporation, 2012; WBM, 2005). However, this DGV was developed from expected bioavailability and toxicity largely to non-tropical species (ANZG, 2018), and should be validated further with tropical species and under tropical conditions. The detection of PAHs in sediments (<200 $\mu\text{g kg}^{-1}$) and waters (<11 $\mu\text{g kg}^{-1}$) of river mouths, in the absence of large vessel and industrial operations, likely reflect inputs from agricultural and urban sources (Humphrey et al., 2007). Following the oil spill from the *Global Peace* in Gladstone Harbour in 2006, maximum concentration of total PAHs in sediments close to the spill were 9,800 $\mu\text{g kg}^{-1}$ (normalized to 1% OC) a month after the spill (Andersen et al., 2008; Melville et al., 2009). We are not aware of other monitoring following large fuel spills from shipping accidents in the GBR or TS regions, including ongoing monitoring to determine the long-term impacts of petroleum hydrocarbon contamination, the effectiveness of any clean-up operations, and/or the potential for recovery of marine ecosystems.

Acute exposure to high concentrations of oil, fuel and coal spills from ship collision, grounding or sinking can have detrimental and long-term effects on tropical marine ecosystems (Peterson et al., 2003). For example, an approximate 10,000 m^3 spill of medium crude in coastal waters of Panama in 1968 had extensive long-term effects on mangroves, seagrasses and corals (Jackson et al., 1989), with very little evidence of recovery of coral reefs observed another five years on (Guzman et al., 1994). Floating oil slicks can cause particular harm to (i) reefs which emerge at low tide or by interacting with floating coral gametes during mass spawning (Loya and Rinkevich, 1980; Negri and Heyward, 2000); (ii) fish

stocks through mortality of eggs and larvae (Langangen et al., 2017); (iii) intertidal mangrove forests where oil degradation rates are slow and contamination is likely to be long-term (Duke, 2016); and (iv) waders and seabirds (Piatt et al., 1990), and marine reptiles and mammals that surface to breathe (Geraci and St. Aubin, 1990). A major oil spill on coastal or reef ecosystems in the GBR or TS is thus expected to result in widespread habitat degradation, acute wildlife mortality and contamination lasting for decades (Burns et al., 1993; Guzman et al., 1994; Haapkylä et al., 2007; Jackson et al., 1989). In addition to detrimental effects of surface slicks, the dissolved fraction of PAHs is particularly toxic in particular to pelagic species and life history stages such as eggs and larvae (French-McCay, 2002; Sweet et al., 2018). Default guideline values for marine water quality are available for only a few MAHs and PAHs (ANZG, 2018), and are therefore inadequate for assessing the toxicity of complex dissolved MAHs and PAHs mixtures that would be present in the environment following an oil spill. Recent laboratory tests have shown that coral larvae are relatively sensitive to dissolved MAHs and PAHs, being negatively affected at concentrations as low as $24 \mu\text{g L}^{-1}$ (total MAH + PAH) (Negri et al., 2016; Nordborg et al., 2018), within the hazard threshold range of 10 to $100 \mu\text{g L}^{-1}$ total PAHs proposed for the Deepwater Horizon Spill (French-McCay et al., 2018). Chemical dispersants which reduce the persistence of surface slicks (National Research Council, 2005) were used on the *Oceanic Grandeur*, *Global Peace* and *Shen Neng 1* spills. However, the use of dispersants near sensitive habitats such as coral reefs, mangroves and seagrass meadows is generally not recommended as this can increase exposure to PAHs (Irving and Lee, 2015; Hook and Lee, 2015). Finally, chronic exposure to contaminated sediments associated with ports and large population centres can result in bioaccumulation, lethal and sub-lethal impacts in marine organisms, and in alteration of habitats (Moore and Dwyer, 1974). Despite generally being rapidly metabolized (Kleinow et al., 1987), PAHs or biomarkers for exposure have been identified in coastal and marine organisms across the GBR region, including sea cucumber (*Holothuria* spp.), coral (*Acropora* spp.), giant clam (*Tridacna maxima*), mud crab (*Scylla serrata*) and fish (barramundi, *Lates calcarifer*) (Coates et al., 1986; Humphrey et al., 2007; Negri et al., 2009; Smith et al., 1984; van Oosterom et al., 2010) (Table S8c).

In summary, petroleum hydrocarbons were detected in sediments of GBR marine ecosystems; however, very few exceedances of DGVs for marine water quality and sediment quality were reported (Table S8a, b). These DGVs should be validated further with tropical species under tropical conditions. While petroleum hydrocarbons can originate from a range of natural and human sources in the GBR and TS regions, shipping activities represent a considerable source contributing to over 500 spills in ports, coastal waters and the GBR from 2002 to 2016. The contribution of atmospheric deposition of burnt fuels and bushfires to petroleum hydrocarbon contamination, including exhaust from vessel traffic associated with bulk shipping (Fig. 3a), commercial fishing (Fig. 3b), commercial and recreational tourism (Fig. 3c) and Australian Defence Force operations (Fig. 3d), has not been quantified. Although the likelihood of a very large petroleum spill in the GBR and TS from an oil tanker is very low, such an occurrence would result in widespread habitat degradation, acute wildlife mortality and contamination lasting for decades.

4. Synthesis

Previous studies have examined the presence of CECs in the marine environments of the GBR and TS regions, including antifouling paints (Haynes et al., 2002; Haynes and Loong, 2002), marine debris (Haynes, 1997), metals (Haynes and Johnson,

2000; Haynes and Kwan, 2002), and petroleum hydrocarbons (Haynes and Johnson, 2000). The current review of more recent data indicates that 20 years on, these CECs are still relevant hazards to the GBR and TS, and that other CECs including coal, microplastics and PPCPs require further monitoring and hazard studies. While our focus was on the tropical marine ecosystems along the east coast of Queensland, Australia, covering the GBR Marine Park and WHA and associated reef ecosystems extending into the TS region bordering Papua New Guinea, we envisage that our findings on sources, presence and potential effects of the six different categories of CECs will be relevant to other, less studied, tropical marine ecosystems around the world.

The known and likely sources for the six different categories of CECs reviewed are relatively well understood across the GBR and TS regions (GBRMPA, 2014) (Figs. 1, 2, 3). For example, monitoring information demonstrates that commercial ports, marinas, and shipping incidents and groundings are sources of antifouling paints, metals and petroleum hydrocarbons (Table 1, S1, S3, S4, S5, S8). Moreover, the main source for PPCPs is likely to be wastewater discharge from WWTPs and vessels (Table 1, S7) (Copeland, 2005; GBRMPA, 2018; Maritime Safety Queensland, 2019; O'Brien et al., 2014), although contributions from agricultural and urban land uses, aquaculture, landfills, and coastal and marine tourism cannot be discounted based on information from (inter-) national studies (Brausch and Rand, 2011; Gaw et al., 2014; Masoner et al., 2014; Sarmah et al., 2006). Indeed, the relative contribution of known and likely sources to the CECs considered here, and the associated risks these potential threats pose to the receiving environments, remains largely unknown. Our spatial representation of the known and likely sources of the six CEC categories in this review is a first step to future ecological risk assessments of these CECs to the GBR and TS marine environments.

While the presence of the CECs reviewed was broadly confirmed based on available monitoring data (Table 1, S1–S8), spatio-temporal coverage across the GBR and TS marine ecosystems varied greatly within and among the six categories. For example, the few large datasets we were able to acquire consisted of monitoring information with high temporal resolution at a single location (e.g. metals; (Port Curtis Integrated Monitoring Program Inc., 2015) or, conversely, information with high spatial resolution at a single point in time (e.g. marine debris; (Tangaroa Blue Foundation, 2015)). The routine monitoring that is conducted on some of the CECs considered here, such as organotins, metals, and total PAHs, is mostly done as part of environmental planning and management activities for GBR port and dredging developments (e.g. Department of Environment and Heritage Protection, 2012b; GHD Pty Ltd, 2005; Port of Townsville Limited et al., 2013; Ports North, 2015). However, such monitoring information, often published in voluminous environmental impact statements, is not readily accessible or made publicly available by most port corporations. In contrast, other CECs such as antifouling components other than organotins and metals, coal dust and particles, marine debris, and PPCPs, are not routinely monitored at all. Indeed, monitoring information for either water, sediment, or biota is not available for any of the CECs across the geographical scale of the GBR and TS marine ecosystems with sufficient temporal coverage to support a regional assessment of potential risk. This is surprising given that the majority of contaminant data in the GBR was already considered dated 20 years ago (Haynes and Johnson, 2000).

Our review has revealed the relatively poor understanding of potential risks of exposure to tropical marine organisms and ecosystems for the majority of the CECs reviewed. The clear exceptions are organotin compounds such as TBT which have been banned based on their detrimental environmental impacts (Australian Government, 2013; International Maritime Organization, 2005). Legacy issues associated with organotin con-

tamination of water, sediment, and biota, however, are still present in GBR and TS marine ecosystems (Table S1). In addition to antifouling components, other CECs such as metals and metalloids, marine debris and microplastics, and PAHs have been detected in a variety of organisms from the GBR and TS marine ecosystems, including in remote locations as well as near urban and industrial locations (Ceccarelli, 2009; Jensen et al., 2019; Kroon et al., 2018) (Table S3c, S8c). Field observations or experiments that link exposure patterns and associated bioburdens with potential adverse effects at different biological levels of organization to assess ecological risk (Ankley et al., 2010) are rarely conducted on tropical organisms in GBR and TS marine ecosystems with few exceptions (Humphrey et al., 2007). Controlled laboratory exposure experiments are more commonly conducted on tropical marine organisms but need to consider endpoints at whole of organism (i.e. development, growth, survival) that are meaningful to assessing ecological risk (Ankley et al., 2010). Bioaccumulation of CECs such as TBT, some metals and metalloids, and PAHs was also reported in species for human consumption, in some cases exceeding Australian food standard guidelines (Table S1c, 3c, 8c). For those locations a re-appraisal of potential risk from seafood consumption is in order, given that most of these data are more than ten years old and Australian food standard guidelines are non-existent for CECs such as TBT, marine plastics, or PAHs.

Finally, our review focused on six different categories of CECs known to be present, or likely to be present in GBR and TS marine environments. Our initial consideration of potential categories of CECs to be examined also included (i) nanomaterials, (ii) per- and poly-fluoroalkyl substances (PFAS), and (iii) unexploded ordnance, explosive ordnance waste and a wide range of disposed war materials. However, monitoring information on these three categories appear to be either non-existent or even less available and/or accessible compared to the six categories of CECs examined here. The increased use of engineered nanomaterials (i.e. materials measuring 1 – 100 nm in at least one dimension) in industrial applications and in consumer and medical products (Batley et al., 2013; Vance et al., 2015) and their occurrence in wastewater (Neale et al., 2013) has raised concerns relating to their potential effects on marine organisms including coral, algae, macro-invertebrates and fish (Matranga and Corsi, 2012). PFAS have been used in industrial processes such as fire-fighting foams, and in common household products and specialty applications (Ahrens and Bundschuh, 2014). Environmental contamination around Australian Defence properties (Department of Defence, 2019b) and commercial ports and airports (Queensland Government, 2019b) is currently being investigated, although the distribution and effects of PFAS on the GBR and TS ecosystems is currently unknown. Unexploded ordnance and munitions have been dumped in coastal marine environments around the world (Beck et al., 2018), including around Australia (Plunkett, 2003) and in the GBR (PGM Environment and Eco Logical Australia, 2014). Corrosion of metal munition housing, and associated release of organic explosive compounds or chemical warfare agent into the marine environment, may result in acute and sub-lethal toxicity to marine organisms (Beck et al., 2018). Given the potential ecological effects of these three additional categories of CECs, we recommend that their presence and distribution are further examined through targeted monitoring studies to determine potential exposure of GBR and TS marine environments.

4.1. Areas of further research

The information presented in this review, in particular the spatial representation of the known and likely sources, will help inform future ecological risk assessment and management of the CECs known to be present, or likely to be present in GBR and TS

marine environments. In contrast to the considerable progress made linking the effects of sediment, nutrients and pesticides on GBR ecosystems to their catchment sources and transport pathways (Brodie et al., 2012; Kroon et al., 2016), relatively little progress in these fields has been made for CECs since the studies conducted by Haynes and colleagues in the late 1990s and early 2000s (Haynes, 1997; Haynes et al., 2002; Haynes and Johnson, 2000; Haynes and Kwan, 2002; Haynes and Loong, 2002). Our review has provided an update on the current understanding of known and potential sources, presence and effects of CECs in the study region. Based on our findings, we recommend the following key areas of research to further progress future ecological risk assessments of CECs to the GBR and TS marine ecosystems.

4.1.1. Identify potential high-risk CECs for targeted monitoring

For CECs such as PPCPs, continuously discharged in wastewater from WWTPs and from vessels, very little to no monitoring data exists for the study region and our review relied primarily on information from other Australian or international studies. Similarly, the lack of monitoring at vessel grounding sites following clean-up (Marshall et al., 2002) or no clean-up (GBRMPA, 2011) means that the long-term impacts of exposure to extreme concentrations of antifouling components are currently unknown. This highlights the need for targeted monitoring campaigns in the GBR and TS marine environments to examine acute and chronic exposures to CECs, and to reduce the uncertainty in future ecological risk assessments. Such monitoring should be informed by the development of a short-list of potential high-risk CECs using predictive methods for risk prioritization (Dong et al., 2013; King et al., 2015). Such a strategy will also help avoid the pitfalls of the Matthew Effect (commonly known as the 'bandwagon effect'), where available monitoring and toxicity data influences and biases future research activities (Daughton, 2014; Grandjean et al., 2011). Once a comprehensive suite of CECs has been identified for monitoring using this process, collaboration on regional, national and international scales will be necessary to generate a comprehensive database of compounds posing a risk to tropical marine environments, and particularly the high value GBR and TS environments.

4.1.2. Integrate monitoring data into publicly available databases

For some of the six categories of CECs reviewed, additional datasets were identified but, despite repeated requests, were not made available for our study by the respective custodians. This means that not all existing environmental datasets were included in our review, with a potential flow-on effect on policy and management not being based on a complete picture of existing relevant information. Moreover, much of our effort was focused on repeatedly trying to access relevant datasets rather than conducting associated analyses to determine presence and distribution of CECs. We recommend that monitoring data for CECs are placed into the public domain and made available in integrated databases for developing marine baselines for the GBR and TS marine environments (National Marine Science Plan 2015–2025, 2015). Depending on the CEC, this would include data from governments and government agencies, universities, research organizations, agricultural industries, shipping industries, port authorities and associated industries, engineers, consultants, NGOs and community groups.

4.1.3. Derive guideline values for tropical marine environments

DGVs for contaminants in marine water and sediment are generally derived from toxicity data that do not reflect environmental variables typical of tropical marine environments, or include species that are representative of tropical marine ecosystems (ANZG,

2018). The bioavailability and toxicity of contaminants can be influenced by variables such as temperature (Pathiratne and Kroon, 2016) and UV (Nordborg et al., 2018) which are generally higher in tropical environments. Moreover, tropical marine ecosystems often contain unique habitat-forming species such as corals and sponges for which toxicity data are mostly absent, limiting the relevance of DGVs. Hence, we recommend that both field observational and controlled field and laboratory exposure studies are conducted on tropical species to derive site-specific guideline values for GBR and TS marine ecosystems. Tropical environmental conditions, including high seawater temperature and UV that can modulate the toxicity of high-risk CECs should be accounted for in such studies.

4.1.4. Assess the ecological risk of priority CECs

The potential risks of exposure to enhanced levels of land-based pollutants such as suspended sediments, nutrients and pesticides to GBR coastal and marine ecosystems has been assessed several times (Waterhouse et al., 2017). In contrast, the potential risk of the CECs considered in this review have only been considered in a qualitative, cursory manner largely due to a lack of information (Kroon et al., 2015). While regional-wide monitoring information is scarce, our review shows that traditional contaminants such as organotins, metals and PAHs are still present, low dose exposures to contaminants of emerging interest such as PPCPs and coal dust and particles are likely, and emerging contaminants such as microplastics appear to be ubiquitous in the GBR and TS marine environments. It is likely that the sources and diversity of contaminants released into these environments will increase given the current and projected agricultural, urban and industrial developments in the GBR region (Det Norske Veritas Ltd, 2013; GBRMPA, 2014). Hence, an assessment of the ecological risks of priority CECs, including the risks relative to those posed by suspended sediments, nutrients and pesticides is warranted to ensure mitigation efforts are focused on those contaminants posing the highest threat now and into the future. This should include a characterization of exposure that evaluates potential or actual co-occurrence or contact of the contaminant with one of more ecological entities, and a characterization of ecological effects that evaluates the ability of a contaminant to cause adverse effects to one of more ecological entities (U.S. Environmental Protection Agency, 1998). We envisage that our spatial representation of the main sources of the six categories of CECs will contribute to the characterization of exposure in particular, and to future ecological risk assessments of CECs to the GBR and TS marine ecosystems in general.

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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Author statement

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Appendix A. Supplementary data

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