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Trace metal pollution gradients in a tropical seagrass ecosystem

Teresa Baptista Nobre[a,b,*], Luka Seamus Wright[b], Dominik Kneer[a], Dody Priosambodo[d], Sebastian C.A. Ferse[a,c,e]

[a] Leibniz Centre for Tropical Marine Ecology (ZMT) GmbH, Fahrenheitstrasse 6, 28359, Bremen, Germany
[b] Oceans Institute, The University of Western Australia, 35 Stirling Highway, Perth, WA 6009, Australia
[c] Marine Ecology Department, Faculty of Biology and Chemistry, University of Bremen, Leobener Strasse, 28359, Bremen, Germany
[d] Department of Biology, Faculty of Mathematics and Natural Sciences, Universitas Hasanuddin, Kampus Tamalanrea, JI. Perintis Kemerdekaan KM. 10, Makassar, 90245, Indonesia
[e] Faculty of Fisheries and Marine Sciences (FPIK), Bogor Agricultural University (IPB), Kampus IPB Dramaga, Jl. Agatis 1, Bogor 16680, Indonesia

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ABSTRACT

Trace metals are one of the most serious pollutants in tropical seagrass meadows given their persistence and toxicity. Whereas quantity is frequently measured, there is no information on the spatial extent of metal pollution in these systems. Here, we use an island in Indonesia (Barang Lompo) as a model system to study the impact radius of two major and eight trace metals in sediment and seagrass leaves. We provide evidence for exponential decay in both the metal pollution index and concentrations of most metals with increasing distance from the island ($k = -0.01$ to -0.08 m^{-1}). Consequently, there is an impact radius of approximately 100 m around the island. The comparative analysis of both seagrass species further revealed interspecific differences in metal loads. This study highlights the importance of assessing the spatial extent of metal pollution in addition to its quantity.

1. Introduction

Marine coastal zones lying adjacent to populated urban areas are highly impacted by land-based activities and coastal development. In many places of the world, the effects of terrestrial-derived inputs in coastal waters have resulted in substantial spatial gradients in water quality and pollutant accumulation (Edinger et al., 1998; Fabricius et al., 2005; Vallejo Toro et al., 2016). In order to support effective environmental planning and management of ecosystems, an assessment of their pollution status is required. This is especially the case for estuarine (de Souza et al., 2016) and marine ecosystems (Costa-Böddeker et al., 2020), which are exposed to an array of organic and inorganic pollutants, such as heavy metals. In recent years, there has been increasing ecological and global public health concerns associated with environmental contamination by heavy metals (Tchounwou et al., 2012; Shah, 2021). Given their persistence, bioaccumulation, magnification and high toxicity characteristics, heavy metals are one of the most serious pollutants in the marine environment (Schneider et al., 2018; Shah, 2021; Jeong et al., 2021; Han et al., 2021).

Sources of heavy metal pollution include natural weathering of the Earth's crust, soil erosion, mining and smelting operations, industrial effluents, urban and agricultural runoff, paint additives, refineries, nuclear power stations and high-tension lines (Morais et al., 2012; Naser, 2013). In addition to these sources, electronic waste is an emerging environmental problem (Alabi et al., 2021). Recent estimates place the global e-waste production at 50 million tonnes a year and on track to reach 120 million tonnes per year by 2050 if current trends continue (World Economic Forum, 2019). In Indonesia, it is estimated that more than 9500 tonnes of waste are produced annually from cell phones alone, an amount that is steadily increasing and is likely to be similar for other electronic devices (Panambunan-Ferse and Breiter, 2013). The lack of appropriate waste management and recycling of e-waste have contributed to an increase in the level of heavy metals in the environment (Quan et al., 2014; Diarra and Prasad, 2021).

In estuaries and coastal environments, metal enrichment can ultimately affect the distribution and density of benthic organisms, as well as the composition and diversity of infaunal communities in marine sediments (Bryan and Langston, 1992; Blankson et al., 2021; Rao et al., 2023). The degree of trace metal contamination in environmental matrices has been measured and reported in numerous studies designed to determine the quality of marine ecosystems (Lee et al., 2023; Mishra et al., 2023). Macrophytes such as seagrasses tend to have even higher

* Corresponding author. Leibniz Centre for Tropical Marine Ecology (ZMT) GmbH, Fahrenheitstrasse 6, 28359, Bremen, Germany.
E-mail address: teresa.baptistanobre@research.uwa.edu.au (T. Baptista Nobre).

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metal concentrations than the sediment they are rooted in (Mance, 1987; Lee et al., 2023) due to the uptake from not only the sediment via the root system but also from the surrounding water (Fö;rstner & Wittmann, 1981; Bonanno and Borg, 2018). This analysis of organismal tissue can present a time-integrated measurement of bioavailable trace metals and offer an early indication of environmental disruption (Rainbow, 1995). Owing to their high bioaccumulation capacity, seagrasses have been used extensively as a bioindicator for metal monitoring purposes, especially in the tropics (Ahmad et al., 2015b; Sidi et al., 2018; Suratno and Irawan, 2018, Cai et al., 2024). Our recent (July 2023) literature search revealed extensive knowledge on the concentration of various metals in the two most common tropical seagrasses, *Thalassia hemprichii* and *Enhalus acoroides* and their sediments (Table 1).

The present study aims to (i) determine the concentrations of aluminium (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), lithium (Li), nickel (Ni), lead (Pb), and zinc (Zn) in two widespread tropical seagrass species in the Spermonde Archipelago, in Indonesia, (ii) understand the spatial on-off-shore gradient of heavy metal pollution in the local meadows by analysing both the seagrass leaf tissue and the surface sediments. In addition, in order to obtain a first indication of the potential trophic transfer of contaminants in the study region, we iii) assessed metal concentrations in a small sample of two species of commercially important fish.

2. Methods

2.1. Study area

The present study was performed in Barang Lompo (05.048°S, 119.328°E) an island in the Spermonde Archipelago located off southwest Sulawesi, Indonesia (Fig. 1) which lies on a seagrass covered platform reef crowned by a coral cay. The Spermonde Archipelago covers approximately 2500 km^2 and is comprised of about 67 low-lying coral atoll islands, 54 of which are populated (Glaeser, 2017). Barang Lompo has an area of 0.14 km^2 and is inhabited by around 5000 people (Sur et al., 2018) most of whom are almost exclusively dependent on marine resources for their livelihood (Glaeser, 2017). Barang Lompo's lack of waste management infrastructure has been previously detailed (Sur et al., 2018).

2.2. Species and sampling method

Seagrass and sediment samples were collected between April and August 2010 along four transects radiating outward from the island approximately towards the north, east, south, and west (Fig. 1). Sampling locations were recorded on-site as GPS coordinates and were roughly as follows: mid-tide line on the beach, edge of the seagrass meadow, 10 m into the meadow, 100 m into the meadow, middle of the meadow, and 100 m from the reef-ward edge of the seagrass meadow. As the meadow to the east is narrower, only four locations were sampled in this direction. On the southern transect, there were seagrass free areas in the meadow, one due to boat scarring and one due to a blowout, a natural erosive feature common in high energy seagrass meadows (Patriquin, 1975). In both areas sediment and seagrass samples were collected at slightly more distant locations. A total of 23 sites were sampled. The seagrass meadows were all eu- or infra-littoral. The eastern meadows were slightly deeper, approximately 1 m below the tide line.

In the study area, several seagrass species were present, however, they mostly occurred at a single station making comparisons within species and across sample sites unreasonable, hence only two species were sampled. *Thalassia hemprichii* (Ehrenb. Asch) and *Enhalus acoroides* (Linnaeus f. Royle) are the two most widely distributed species in the western Pacific (Mukai, 1993) and particularly in the Spermonde Archipelago (Verheij and Erftemeijer, 1993). For this reason, they were used as bioindicator species in this study.

At each sampling site, six shoots (material above the sediment) from individual seagrass plants were collected from each species. The seagrass samples were rinsed with seawater to remove any sediment and epiphytes that frequently attach to the leaves. The different species' shoots were placed in separate sealed polyethylene bags and transported to the laboratory in a cool box (<4 °C) and preserved at −20 °C prior to laboratory analysis. Three sediment samples were collected from each site using an acrylic plastic corer to avoid metal contamination. The cylindrical vial collected the top 10 cm of sediment. Sediment samples were carefully separated from any visible fauna and debris (broken coral pieces, gastropod shells, etc.) which were picked by hand and removed from the sample. All sampling was done via snorkelling.

To determine the potential trophic transfer of heavy metal contaminants from the environment and primary producers to locally commercially important fish, six white-spotted spine-foot fish, *Siganus canaliculatus* (Park, 1797) and six blue tail mullet, *Crenimugil buchanani* (Bleeker, 1853), were collected in August 2010. Whilst not the focus of this work, the analysis of heavy metals present in fish muscle tissue offers additional information regarding local contaminants. Spine-foot fish were collected at the southern transect while blue tail mullet were collected at Sari Laut beach in the city of Makassar, located approximately 10 km away from the study area. The fish were caught by line-fishing, labelled, and stored at −20 °C until dissection. Detailed sampling information and number of samples analysed can be found in Tab le S10.

2.3. Sample processing and analysis

All laboratory materials used were prepared following rigorous cleaning procedures before and in between samples. Polyethylene and Teflon beakers were immersed in 2% hydrochloric acid at room temperature for 24 h prior to use and later rinsed thoroughly with Milli-Q H_2O. All chemical reagents used were of Merck suprapure grade. The samples were analysed at the Leibniz Centre for Tropical Marine Research (ZMT) in Bremen, Germany.

Seagrass blades were cut using a ceramic knife, freeze-dried, ground, and homogenised in a ceramic mortar, and kept in sealed vials until analysis. Sediment samples were freeze-dried and ground to uniform particle size. To measure metal concentration, the frozen fish were thawed at room temperature and dissected using a ceramic knife. The axial muscle meat (without skin) was filleted from each fish (about 5–10 g). The fish muscle tissues were then dried to a constant weight in an oven at 40 °C for approximately 24 h (see Gu et al., 2015). Fish muscle tissue and sediment samples were homogenised in a Retsch MM400 mill (Retsch GmbH, Germany) with ceramic spheres. All samples were weighed before and after freeze-drying to get an accurate proportion of lost water weight.

The sediment was digested according to Hu et al. (2019) with minor modifications. Briefly, approximately 0.1 g (dry weight) of sediment ground to analytical fine powder (<2 μm) was placed in an acid-washed Teflon digestion vessel with a mixed acid (5 mL HNO_3 and 1.8 mL HCl). The mixture was heated on an AHF EvapoClean heating block (AHF analysentechnik, Germany) at 110 °C for 16 h. After cooling down to room temperature, the temperature was again raised to 120 °C to evaporate the concentrated acid to incipient dryness. After that, the residue was dissolved in 10 mL of 0.5 M HNO_3 and heated in a closed Teflon digestion vessel at 120 °C until the solution become clarified.

Both the seagrass and fish muscle tissue samples were treated in the following way. Known quantities of powdered sample (~0.15 g) were placed in an acid-washed Teflon digestion vessel with a mixed acid (5 mL HNO_3 and 1.8 mL HCl) at room temperature. After an hour, 200 μL of H_2O_2 was added. After another hour, the mixture was placed in the AHF heating block at 40 °C for an hour, then raised to 82 °C for another hour. The vessels were then closed, and the temperature raised to 130 °C overnight. Finally, the vessel was cooled to room temperature and after

Table 1
Global summary of metal concentrations (μg g^{-1} dry mass, or ppm) in the tropical seagrasses *Enhalus acoroides* and *Thalassia hemprichii* and their sediment, collated from an exhaustive list of 30 publications. Concentrations are given as mean ± s.d. where each observation is a mean value from the published literature. Where only one published mean is available, s.d. is omitted. Superscripts denote references cited as footnotes in order of appearance. A more detailed version of this table with sample sizes and ranges is provided in the supplement as Ta ble S1.

	Al	As	Cd	Co	Cr	Cu	Fe	Hg	Li	Mg	Mn	Ni	Pb	Ti	Zn
China															
Sediment			0.47 ± 0.26[a-c]		1.3[c]	7.8 ± 12[a-c]						1.6[c]	7.1±3.2[a,c]		36 ± 33[a-c]
E. acoroides			1.1±0.97[a,c]		33±15[c]	8.2±6[a,c]						3.6±1.4[c]	7.4±7.6[a,c]		26±11[a,c]
T. hemprichii			1.3 ± 1.1[a-c]		63±27[c]	21 ± 23[a-c]						3.7±0.5[c]	6.8±5.5[a,c]		93 ± 87[a-c]
India															
E. acoroides	830[d]		0.71 ± 0.99[d-f]	0.7±0.4[d,e]	30±38[d,e]	20 ± 16[d–g]	510±410[d,e,g]			730[e]	270±250[d,e,g]	1.6±0.17[d,e]	2.2±1.3[d,e,f]		21 ± 12[d-g]
T. hemprichii	470 ±210[d,h]		1.4±1.2[d,e,h,i]	1.1 ±0.77[d,e,h]	43±45[d,e,h]	44±49[d,e,h–j]	500±420[d,e,h–j]			3600 ±4300[e,h]	270±320[d,e,h-j]	4.6±4.4[d,e,h,i]	7.2±4[d,e,h]		27±20[d,e,h-j]
Indonesia															
Sediment	960 ±760[k]	0.82±0.26[k]	1.3±1.3[l-s]		4±4.3[k]	6.6±4.5[k-m,q,r,t]	8800 ±19000[k,l]	0.64±0.72[l,u,v]			12±4.2[k]	0.73±0.32[k]	5.6±5.4[k-n,p,q,s,t,v]	8.4±6.5[k]	18±46[k-m]
E. acoroides	0.58 ±0.4[k]	0.0018 ±0.0044[k]	0.4±0.38[l-n,q-s,w]		0.0027 ±0.0029[k]	1.7±2[k-m,q,r,t,w]	650 ±1300[k,l]	0.12±0.19[l,u,v]			0.029 ±0.039[k]	0.00089 ±0.0022[k]	2.1±2[k,m,n,q,t,v,w]	0.0036 ±0.0028[k]	13±28[k-j,w]
T. hemprichii			0.29±0.2[o,p,s,w]			5±2.1[w]		0.044±0.027[u]					0.9±0.99[p,s,w]		18±4.5[w]
Malaysia															
Sediment		32[x]	32[x]			51[x]		27[x]					250[x]		
E. acoroides		14±3.3[x,y]	26±25[x-z]			16±14[x-z]		44±20[x,aa]				4.8±1.2[z]	120±110[x,z,aa]		14±7.5[z]
T. hemprichii		9.7±3.5[y]	29±15[y]			21±10		21±2.3[aa]					99±15[aa]		
Micronesia															
Sediment		5.5[ab]	0.036[ab]		16[ab]	7.5[ab]		0.027[ab]				11[ab]	7.7[ab]		20[ab]
E. acoroides		3.6±0.81[ab]	0.038 ±0.024[ab]		0.84±0.29[ab]	2.5±1.6[ab]		0.0075 ±0.00071[ab]				1.8±0.72[ab]	1.2±0.13[ab]		19 ±6.6[ab]
T. hemprichii		3.5±1.3[ab]	0.064 ±0.032[ab]		0.63 ±0.071[ab]	2±1.1[ab]		0.0075 ±0.0035[ab]				1.8±0.91[ab]	0.81 ±0.33[ab]		19 ±4.1[ab]
Palau															
Sediment		13[ac]	0.018[ac]	2.8[ac]	62[ac]	8[ac]		0.015[ac]	12[ac]			14[ac]	1[ac]		11[ac]
E. acoroides		10±7[ac]	0.035 ±0.021[ac]		4±0.21[ac]	2±1.3[ac]		0.008 ±0.0014[ac]				2±0.85[ac]	0.5±0[ac]		13±6.7[ac]
Vietnam															
Sediment			0.056 ±0.042[ad]			11±15[ac]							5.5±7.3[ad]		14±16[ad]
E. acoroides			0.41 ±0.33[ac]			12±34[ac]							1.9±4.6[ad]		18 ±9.4[ad]

[a]Li and Huang (2012), [b]Zheng et al., (2018), [c]Zhang et al., (2021), [d]Thangaradjou et al., 2010, [e]Kannan et al., (2011), [f]Gopi et al., (2020), [g]Mathevan Pillai 1990 in Thangaradjou et al., (2010), [h]Thangaradjou et al., (2013), [i]Gopinath et al., (2011), [j]Kannan et al., 1992 in Thangaradjou et al., (2010), [k]Zamani et al., (2018), [l]Suwandana et al., (2011), [m]Ambo-Rappe (2014), [n]Sugiyanto et al., (2016), [o]Tuapattinaya et al., (2016), [p]Tupan and Uneputty (2017), [q]Wijayanti and Putra (2019), [r]Yona et al., (2020), [s]Rosalina et al., (2022), [t]Werorilangi et al., (2016), [u]Suratno and Irawan (2018), [v]Natsir and Rijal (2020), [w]Nienhuis (1986), [x]Ahmad et al., 2015b, [y]Ahmad et al., (2015a), [z]Sidi et al., (2018), [aa]Ahmad et al., (2014), [ab]Jeong and Ra (2022), [ac]Jeong et al., (2021), [ad]Nguyen et al.(2017).

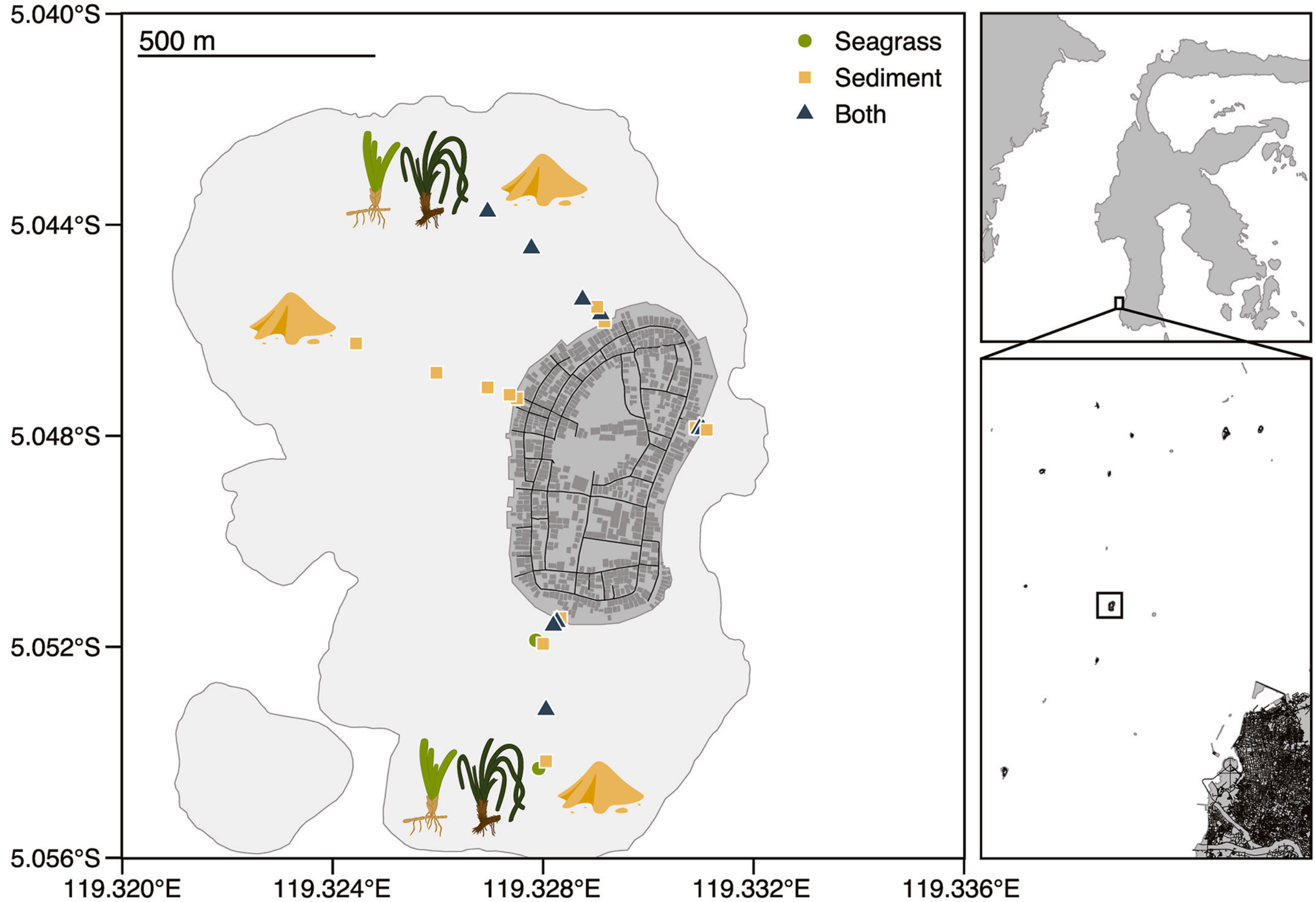

Fig. 1. Location of the island Barang Lompo (left) within the Spermonde Archipelago off Makassar (bottom right) in Sulawesi, Indonesia (top right). Shapes and icons indicate sampling sites for seagrass (circles), sediment (squares) and both materials (triangles). The shaded area around the island shows the extent of the lagoon and the grey outline the reef edge. Map projection: WGS 84. Base map source: OpenStreetMap.

that, heated to 120 °C until the concentrated acid was evaporated.

Al and Fe were determined in the digested samples with a Ciros Vision ICP-OES (Spectro Analytical Instruments GmbH, Germany) given its good accuracy and precision for elements present at higher concentrations. Cd, Cr, Co, Cu, Pb, Li, Ni and Zn were analysed with the PlasmaQuant Elite ICP-MS (Analytik Jena GmbH, Germany) which is better equipped to measure trace concentrations due to its higher sensitivity. The instruments were in a temperature-controlled laboratory and stabilised for a sufficient period (approximately 1 h before measurements were taken). The instrumental parameters of ICP-OES and ICP-MS, as well as the wavelength of detection and isotopes analysed for each element, can be found on the supplementary material.

2.4. Quality assurance

Reagent blanks were analysed concurrently with samples to validate the methods used for metal analysis. No metals were detected in reagent blanks. Instrument calibration was performed using standard solutions with defined concentrations. Dilutions of each element (1 μg L^{-1}, 2 μg L^{-1}, 5 μg L^{-1}, 10 μg L^{-1}, 20 μg L^{-1}, 50 μg L^{-1}) were used as working standard solutions. A blank standard drift was performed in between every five samples. To evaluate the quality of the analytical method and the accuracy of metal analysis on sediment, seagrass, and fish tissue samples, five certified reference materials were analysed (SRM 3232: kelp powder, BCR 129: hay powder, PACS-2: marine sediment, MESS-3: marine sediment, ERM CE278k 129: mussel tissue). The recovery rates and measured element concentrations of the certified reference materials (listed on Tab les S5–S9) indicated good consistency.

2.5. The metal pollution index (MPI)

To evaluate the current pollution levels at each sampling site, the Metal Pollution Index (MPI) was obtained from the following equation (AMA 1992; Usero et al., 2005; Li and Huang, 2012):

$$\mathrm{MPI} = (\mathrm{C}_1 \times \mathrm{C}_2 \times \ldots \mathrm{C}_n)^{\frac{1}{n}} \tag{1}$$

where *Cx* is the concentration of metal *x* in the sample of seagrass or sediment (μg g^{-1}) and *n* is the total number of metals analysed. The MPI values indicate the metal pollution index of each site. Since several extreme outliers had to be removed during the visual data exploration phase, the *Cx* and *n* values in the above equation were adjusted for affected samples. If three or more metals in a given replicate were outliers, the replicate was removed from the MPI dataset because at this low number of metals the index was deemed untrustworthy.

2.6. Data analysis and visualisation

Data analysis and visualisation were performed in R v4.0.4 (R Core Team, 2021) using the integrated development environment RStudio v1.4.1106 (RStudio Team, 2021). Distance from shore of each site was then calculated as geodesic distance assuming an Earth ellipsoid (WGS 84) using the package geosphere v1.5-14 (Hijmans et al., 2021). The map of Sulawesi was plotted using rworldmap v1.3-6 (South, 2016) and rworldxtra v1.01 (South, 2012).

To test whether metal pollution declined with distance from shore, linear and nonlinear models were fit to the sediment and seagrass data. Starting with exponential decay as the theoretically optimal choice, model choice was further influenced by visual data exploration, model convergence and goodness of fit. Extreme outliers were deemed technical outliers due to instrument error and removed prior to analysis. The removal process is transparent in the R script (see Data and code availability). Each model included the continuous response variable metal concentration (μg g^{-1}), the continuous explanatory variable distance (m) and the categorical explanatory variable material (sediment, *E. acoroides* and *T. hemprichii*).

Exponential decay was selected based on theory and determined to be the optimal model in most cases based on visual data exploration and model convergence. The three-parameter exponential decay function describes the change in metal concentration (C, μg g^{-1}) with distance (D, m) as

$$C = P_0 \times e^{-k \times D} + B \tag{2}$$

where P_0 is the metal pollution in excess of the baseline on the coast, or mathematically the difference between the intercept and the asymptote, k is the exponential rate of decay and B is the baseline metal concentration, or mathematically the asymptote. Overall starting values for these parameters were obtained using the *SSasymp* and *nls* functions from the R stats package. Based on these starting values, a new nonlinear least squares model, augmented with the material variable, was built with the *nls* function. Assumptions of normality and homogeneity were again assessed visually (Ritz and Streibig, 2008). Since all nonlinear data were normally distributed but heterogenous, a generalised nonlinear least squares model with a power-of-the-mean variance function (Ritz and Streibig, 2008) was applied using the *gnls* function of nlme v3.1-153 (Pinheiro, 2021).

Simple linear models ($y = \beta x + \alpha$) were fit using the *lm* function of the R stats package, and assumptions of normality and homogeneity of variance were tested visually (according to the steps outlined in Zuur et al., 2009). Data mostly conformed to the assumption of normality. In the single case of Cd in seagrass, data were right-skewed and the Gaussian distribution was consequently ill-fitting. Since transformation affects the scale of the response variable and the test result and should therefore be avoided (Zuur et al., 2009), a gamma distribution with a logarithmic link function was fit to seagrass Cd data using the *glm* function from the R stats package. When heterogeneity was encountered, categorical differences in residual variance were modelled by the categorical explanatory variable material (Zuur et al., 2009). This weighted linear modelling was achieved with generalised least squares using the *gls* and *varIdent* functions of nlme v3.1-153 (Pinheiro, 2021).

All metal concentrations from fish muscle tissue were analysed separately, as distance from shore was deemed negligible given fish mobility. This was done using simple linear models with metal concentration (μg g^{-1}) as continuous response variable and fish species (*S. canaliculatus* and *C. buchanani*) as categorical explanatory variable. The assumptions of normality and homogeneity of variance were again tested visually. In the case of deviation from normality or homogeneity, a generalised linear model with a gamma distribution or a generalised least squares model were fit to the data as described above.

Omnibus sums of squares hypothesis tests (type III for models with two explanatory variables and type II for models with one explanatory variable) were performed with the *Anova* function of car v3.0-11 (Fox and Weisberg, 2019) for all linear models. For contrasts between all three materials, pairwise tests were performed with the *summary* function of the R base package. No straightforward or reliable omnibus hypothesis tests are available for nonlinear models (Ritz and Streibig, 2008), so the statistical difference from zero of the exponential rate of decay (k) for each material and pairwise contrasts between materials for parameters C and A were tested using t tests. 5% was chosen as the α level for all tests.

Descriptive statistics (means ± s.e.m.) were calculated using the *describeBy* function in psych v2.1.9 (Revelle, 2021). Linear model predictions and 95% confidence intervals were calculated using the *predict* function of the R stats package. Generalised least squares 95% confidence intervals were calculated as model predictions $\pm z \times$ s.e.m., which was implemented in R according to the procedure outlined by Bolker (2021a). In the case of generalised linear models, predictions and confidence bounds were calculated on the link scale and then converted to the response scale with the inverse of the link function. The best way to illustrate confidence around nonlinear model fits is by bootstrapping the data (Bolker, 2021b). 95% confidence intervals around nonlinear model fits were bootstrapped with 1000 resamples. All data were visualised using the package ggplot2 v3.3.5 (Wickham, 2021), and plots juxtaposed with cowplot v1.1.1 (Wilke, 2020).

3. Results

3.1. General metal pollution trends

Heavy metal concentrations generally changed with distance from the island but varied in their patterns across the off-shore gradient. Heavy metal concentrations in sediment and seagrasses were mostly found to be the highest closest to the coast, the general trend being exponential decay with distance from the coast (Table 2). The degree of metal pollution, summarised by the Metal Pollution Index (MPI) (**Equation (1)**), increased toward the island at an exponential rate (k) of 0.04 m^{-1} in *E. acoroides* ($t = 2.84$, $p = 0.005$) and 0.03 m^{-1} in sediment ($t = 5.09$, $p < 0.001$) (Fig. 2). Although there was also a visible MPI increase in *T. hemprichii* (Fig. 2), k was not statistically different from zero at the 5% α level. MPI ranged between 1.36 and 19.26 for sediment, 1.09 and 10.48 for *E. acoroides*, and 1.13 and 6.36 for *T. hemprichii*. Closest to shore, MPI was 61% higher in sediment than in *E. acoroides* and 124% higher than in *T. hemprichii*. Away from the island, however, MPI was similar across both seagrasses and sediment.

3.2. Metals which followed the general trend

The overall trend in metal pollution observed in Fig. 2 is explained by a selection of metals, mostly those of highest concentration and therefore strongest influence on the MPI (**Equation (1)**, Fig. 3). These metals, Fe, Al, Pb, Cu and Co, show a similar trend of exponential increase towards the coast of Barang Lompo in both seagrasses and sediment.

For Fe and Al, the metals with the highest concentrations, the amount in the two seagrasses differed closest to the island but converged away from it. The coastal excess concentration (P_0, Equation (2)) of Fe in *E. acoroides* was 29% higher than that in *T. hemprichii* ($t = 2.4$, $p = 0.02$). The baseline concentration away from the island (B, Equation (2)), however, was not different between species (Table 2, Fig. 3b). P_0 for Al was 74% higher in *E. acoroides* than in *T. hemprichii* ($t = 4.61$, $p < 0.001$), yet B did not differ between the two species (Table 2, Fig. 3c). In both seagrasses, [Fe] increased towards the island at an exponential rate (k) of 0.03 m^{-1} (*E. acoroides:* $t = 6.67$, $p < 0.001$; *T. hemprichii:* $t = 4.69$, $p < 0.001$). [Al] in *E. acoroides* increased towards the island at k of 0.01 m^{-1} ($t = 3.47$, $p < 0.001$), but only marginally changed with distance in *T. hemprichii* ($t = 1.95$, $p = 0.05$). In the case of both Pb and Cu, the concentrations did not vary between seagrasses across distance, therefore neither P_0 nor B were different (Table 2, Fig. 3d, e). In both seagrasses, k for Pb was -0.04 m^{-1} (*E. acoroides:* $t = 4.06$, $p < 0.001$, *T. hemprichii:* $t = 4.07$, $p < 0.001$) and for Cu -0.02 m^{-1} (*E. acoroides*: $t = 3.16$, $p = 0.01$, *T. hemprichii*: $t = 2.91$, $p = 0.01$). Conversely, P_0 for Co was not different between the two seagrass species, yet B was higher for *T. hemprichii* ($t = 2.75$, $p = 0.01$) (Fig. 3f). In seagrass, [Co] increased significantly towards the island at an exponential rate of 0.03 m^{-1} in *E. acoroides* ($t = 5.13$, $p < 0.001$) and 0.02 m^{-1} in *T. hemprichii* ($t = 3.21$, $p = 0.01$).

Table 2
Summary of metal concentration descriptive statistics and model parameters (mean ± s.e.m.) for sediment, the seagrasses *Enhalus acoroides* and *Thalassia hemprichii*, and the fishes *Crenimugil buchanani* and *Siganus canaliculatus* in this study. Accompanying test statistics are summarised in Ta bles S2 and S3 and model equations in Tab le S4.

	Al	Cd	Co	Cr	Cu	Fe	Li	Ni	Pb	Zn
μ (μg g^{-1})										
Sediment		0.11 ± 0.0041	0.48 ± 0.036	5.9 ± 0.22	7.5 ± 1.3	2117 ± 382	2.1 ± 0.066	8.5 ± 0.19	5.7 ± 0.81	25 ± 3.9
E. acoroides	93 ± 7	0.19 ± 0.02	0.31 ± 0.022	0.67 ± 0.033	5 ± 0.42	183 ± 13	0.16 ± 0.0092	1.4 ± 0.045	1.4 ± 0.21	47 ± 2.1
T. hemprichii	66 ± 4	0.18 ± 0.02	0.38 ± 0.023	0.63 ± 0.032	5.2 ± 0.43	171 ± 8.4	0.1 ± 0.0061	3 ± 0.088	1.6 ± 0.22	40 ± 1.8
C. buchanani	57 ± 3.6	0.13 ± 0.036	0.033 ± 0.007	0.22 ± 0.024	1.2 ± 0.11	47 ± 5.2	0.043 ± 0.0034	0.07 ± 0.0075	0.086 ± 0.011	13 ± 0.9
S. canaliculatus	77 ± 8.3	0.15 ± 0.014	0.07 ± 0.0042	0.22 ± 0.027	0.76 ± 0.02	39 ± 4.1	0.041 ± 0.0038	0.095 ± 0.0068	0.16 ± 0.032	19 ± 2.4
***P_0* (μg g^{-1})**										
Sediment		0.049 ± 0.0096	0.53 ± 0.08	2.7 ± 0.4	17 ± 1.9	5451 ± 1005			13 ± 1.2	67 ± 6.3
E. acoroides	128 ± 12		0.47 ± 0.071		8.6 ± 1.7	277 ± 33	0.15 ± 0.017		4.8 ± 1.1	58 ± 50
T. hemprichii	54 ± 9.9		0.37 ± 0.055		8.4 ± 1.7	180 ± 24	0.067 ± 0.015		5.3 ± 1.1	11 ± 5.8
***k* (m^{-1})**										
Sediment		−0.047 ± 0.018	−0.062 ± 0.014	−0.016 ± 0.0076	−0.036 ± 0.0051	−0.083 ± 0.012			−0.041 ± 0.0064	−0.051 ± 0.0049
E. acoroides	−0.013 ± 0.0036		−0.027 ± 0.0053		−0.019 ± 0.0061	−0.025 ± 0.0038	−0.011 ± 0.0044		−0.035 ± 0.0087	−0.07 ± 0.058
T. hemprichii	−0.016 ± 0.0084		−0.015 ± 0.0048		−0.018 ± 0.0061	−0.033 ± 0.0071	−0.011 ± 0.0089		−0.038 ± 0.0093	−0.009 ± 0.018
***B* (μg g^{-1})**										
Sediment		0.091 ± 0.0038	0.31 ± 0.017	4.4 ± 0.19	0.68 ± 0.087	539 ± 32			0.8 ± 0.16	1.5 ± 0.17
E. acoroides	43 ± 6.7		0.19 ± 0.0088		2.4 ± 0.27	109 ± 3.8	0.096 ± 0.014		0.4 ± 0.081	40 ± 2.5
T. hemprichii	45 ± 5.3		0.24 ± 0.016		2.5 ± 0.32	120 ± 4.2	0.074 ± 0.013		0.44 ± 0.09	35 ± 6.1
***α* (μg g^{-1})**										
Sediment							2.2 ± 0.084	8.8 ± 0.24		
E. acoroides		−2±0.17 (0.13)*		0.79 ± 0.047				1.4 ± 0.069		
T. hemprichii		−2.1 ± 0.14 (0.13)*		0.63 ± 0.046				3.4 ± 0.11		
C. buchanani	57 ± 3.6	0.13 ± 0.036	0.033 ± 0.007	0.22 ± 0.024	1.2 ± 0.11	47 ± 4.7	0.043 ± 0.0037	0.07 ± 0.0075	−2.5 ± 0.16 (0.086)*	13 ± 0.9
S. canaliculatus	77 ± 8.3	0.15 ± 0.014	0.07 ± 0.0042	0.22 ± 0.027	0.76 ± 0.02	39 ± 4.7	0.041 ± 0.0037	0.095 ± 0.0068	−1.9 ± 0.17 (0.16)*	19 ± 2.4
***β* (μg g^{-1} m^{-1})**										
Sediment							−0.00046 ± 0.00053	−0.0026 ± 0.0015		
E. acoroides		0.0022 ± 0.00091*		−0.00084 ± 0.00025				−0.00027 ± 0.00037		
T. hemprichii		0.0025 ± 0.00089*		−0.000015 ± 0.00025				−0.0027 ± 0.00061		

μ: average across all observations, P_0: excess coastal metal pollution above the baseline, *k*: exponential decay of metal concentration with distance from coast, *B*: baseline metal concentration, *α*: coastal metal concentration, *β*: linear change in metal concentration with distance from coast, $P_0 + B$ gives the predicted metal concentration at the coast, equivalent to *α*. *gamma generalised linear model estimates on the logarithmic link function scale (for *α* the exponentially transformed mean is given in brackets).

Concentrations of each element in sediment varied in relation to the concentration of the same element in seagrass. Concentration of Fe in sediment increased towards the island at an exponential rate of 0.08 m^{-1} ($t = 6.79, p < 0.001$). At the island, [Fe] in sediment was an order of magnitude higher than in seagrass (Fig. 3a); more specifically, [Fe] in sediment was 15 times higher than in *E. acoroides* ($t = 6.67$, $p < 0.001$) and 19 times higher than in *T. hemprichii* ($t = 6.67$, $p < 0.001$) at the same location. For this reason, they were plotted separately. *B* for Fe dropped considerably but was still 4 times higher than in *E. acoroides* ($t = 13.23$, $p < 0.001$) and 3.5 times higher than in *T. hemprichii* ($t = 12.9$, $p < 0.001$). Al content in sediment could not be measured as the values were below detection range.

Pb concentration in sediment increased towards the island at an exponential rate of 0.04 m^{-1} ($t = 6.39, p < 0.001$) (Fig. 3d). Similarly to Fe, at the island [Pb] in sediment was much higher than in seagrass; it was 156% higher than in *E. acoroides* ($t = 4.79$, $p < 0.001$) and 132% higher than in *T. hemprichii* ($t = 4.34$, $p < 0.001$). *B* for Pb in sediment away from the island was double that of *E. acoroides* ($t = 2.29, p = 0.02$) and 82% higher than in *T. hemprichii* ($t = 2.01$, $p = 0.046$). Co concentration in sediment was also higher at the island, towards which it increases at an exponential rate of 0.06 m^{-1} ($t = 4.60$, $p < 0.001$) (Fig. 3f). It also remained higher than in seagrass away from the island, where it was 63% higher than in *E. acoroides* and 29% higher than in *T. hemprichii.* Cu concentration increased towards the island at an exponential rate of 0.04 m^{-1} ($t = 7.06, p < 0.001$) (Fig. 3e). Unlike the previous metals, baseline sediment [Cu] was 35% lower than the

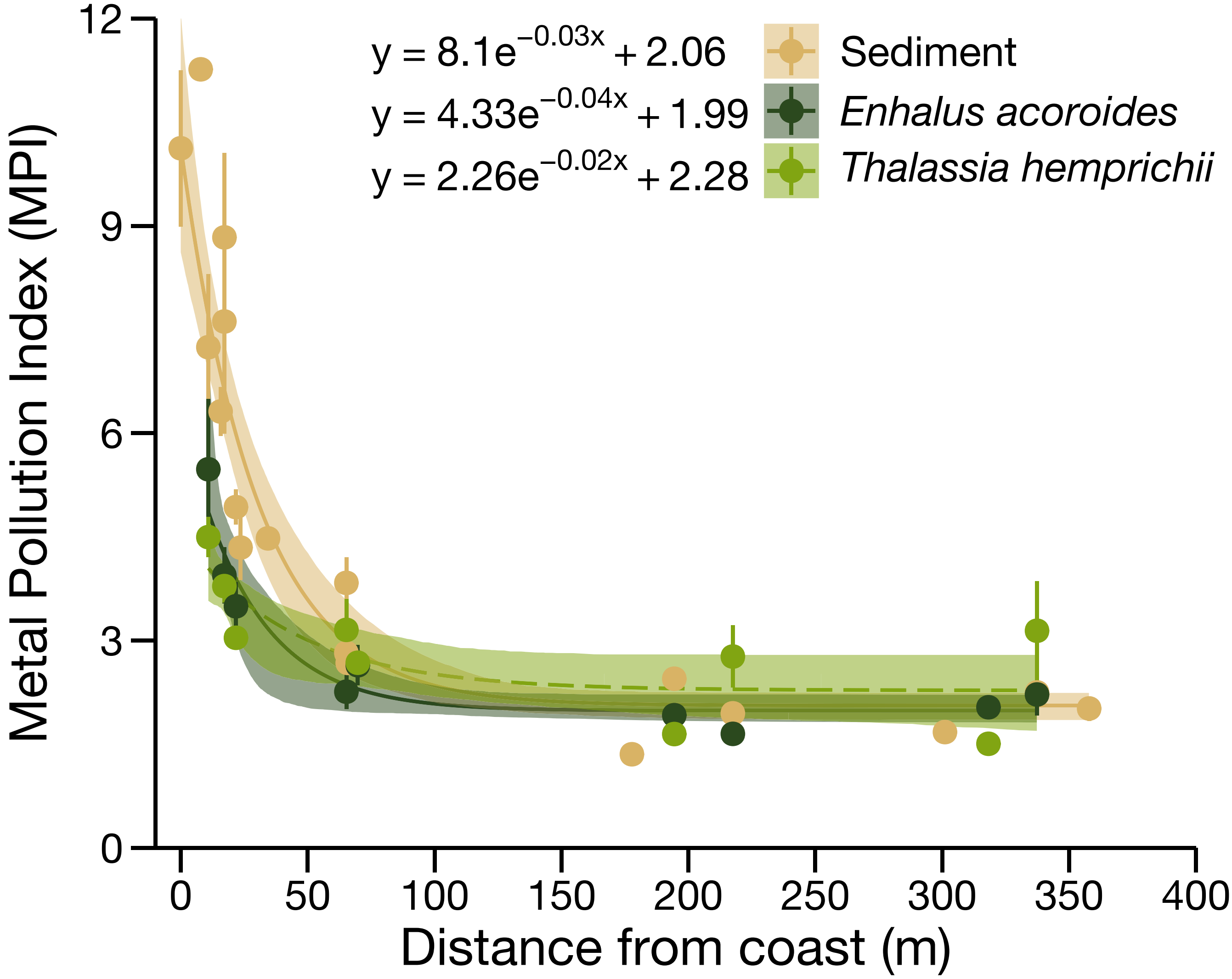

Fig. 2. Trend in Metal Pollution Index (MPI) with distance from the coast of Barang Lompo. Point ranges are means ± SE and indicate MPI at individual sampling sites. Lines and shaded areas are model predictions and 95% confidence intervals respectively. Solid lines represent significant slopes at the 95% confidence level, while dashed lines indicate no significant change with distance.

concentration in *E. acoroides* and *T. hemprichii* at the same location.

3.3. Metals which did not conform to the general trend

While the metals outlined in section 3.2 followed the overall trend in metal pollution well, other elements did not conform to this general trend (Fig. 4). The concentrations of some of the elements did not increase exponentially towards the island but were best described by linear models. In the case of Zn, both seagrass species showed very similar concentration values throughout the studied area. In both cases, [Zn] did not change significantly with distance from the island (Fig. 4a). Closest to the island, [Zn] of *E. acoroides and T. hemprichii* differed significantly ($t = 2.4$, $p = 0.02$). However, *B* for Zn was not different between species. Ni concentration in seagrass followed a linear trend with distance from the coast. [Ni] did not change with distance in *E. acoroides*, but did significantly increase in *T. hemprichii* by 0.0027 μg g^{-1} m^{-1} towards the island (X^2_1, $152 = 20.23$, $p < 0.001$) (Fig. 4b). The [Ni] of *T. hemprichii* was 137% higher than in *E. acoroides* ($t = 14.81$, $p < 0.001$). Cr concentration in seagrass increased by 0.0008 μg g^{-1} m^{-1} towards the island in *E. acoroides* ($F_{1,\,105} = 10.94$, $p = 0.001$), but did not change in *T. hemprichii* (Fig. 4c). Closest to shore, [Cr] was 25% higher in *E. acoroides* than in *T. hemprichii* ($F_{1,\ 105} = 5.52$, $p = 0.02$). Li concentration in seagrass increased toward the island at an exponential rate of 0.01 m^{-1} ($t = 2.53$, $p = 0.01$) in *E. acoroides*, but did not change significantly in *T. hemprichii* (Fig. 4e). Li in *E. acoroides* was 79% higher than in *T. hemprichii* ($t = 3.84$, $p < 0.001$). *B*, however, was not significantly different between species. Cd was the only element where there was a decrease in concentration away from the island. In both *E. acoroides* and *T. hemprichii*, [Cd] increased at exponential rates of 0.002 and 0.003 m^{-1} away from the island in *E. acoroides* and *T. hemprichii*, respectively (*E. acoroides*: $X^2_{1,\ 110} = 6.37$, $p = 0.012$, *T. hemprichii*: $X^2_{1,\ 110} = 9.05$, $p = 0.003$).

Throughout the study area, the concentration levels of these metals in sediment were always higher than in seagrass, except for Zn. In sediment, [Zn] increased towards the island at an exponential rate of 0.05 m^{-1} ($t = 6.79$, $p < 0.001$) (Fig. 4a). At the island, [Zn] in sediment did not differ significantly from that of *E. acoroides* but was 48% higher than that of *T. hemprichii* ($t = 6.53$, $p < 0.001$). Further from the island, sediment [Zn] had a sharp decrease and was 96% lower than that of both *E. acoroides* ($t = 15.14$, $p < 0.001$) and *T. hemprichii* ($t = 5.57$, $p < 0.001$). In sediment, Ni concentration did not change significantly with distance. [Ni] in sediment was found to be five times higher than in *E. acoroides* ($t = 29.53$, $p < 0.001$) and 1.6 times higher than in *T. hemprichii* ($t = 20.33$, $p < 0.001$) (Fig. 4b). In sediment, [Cr] increased significantly towards

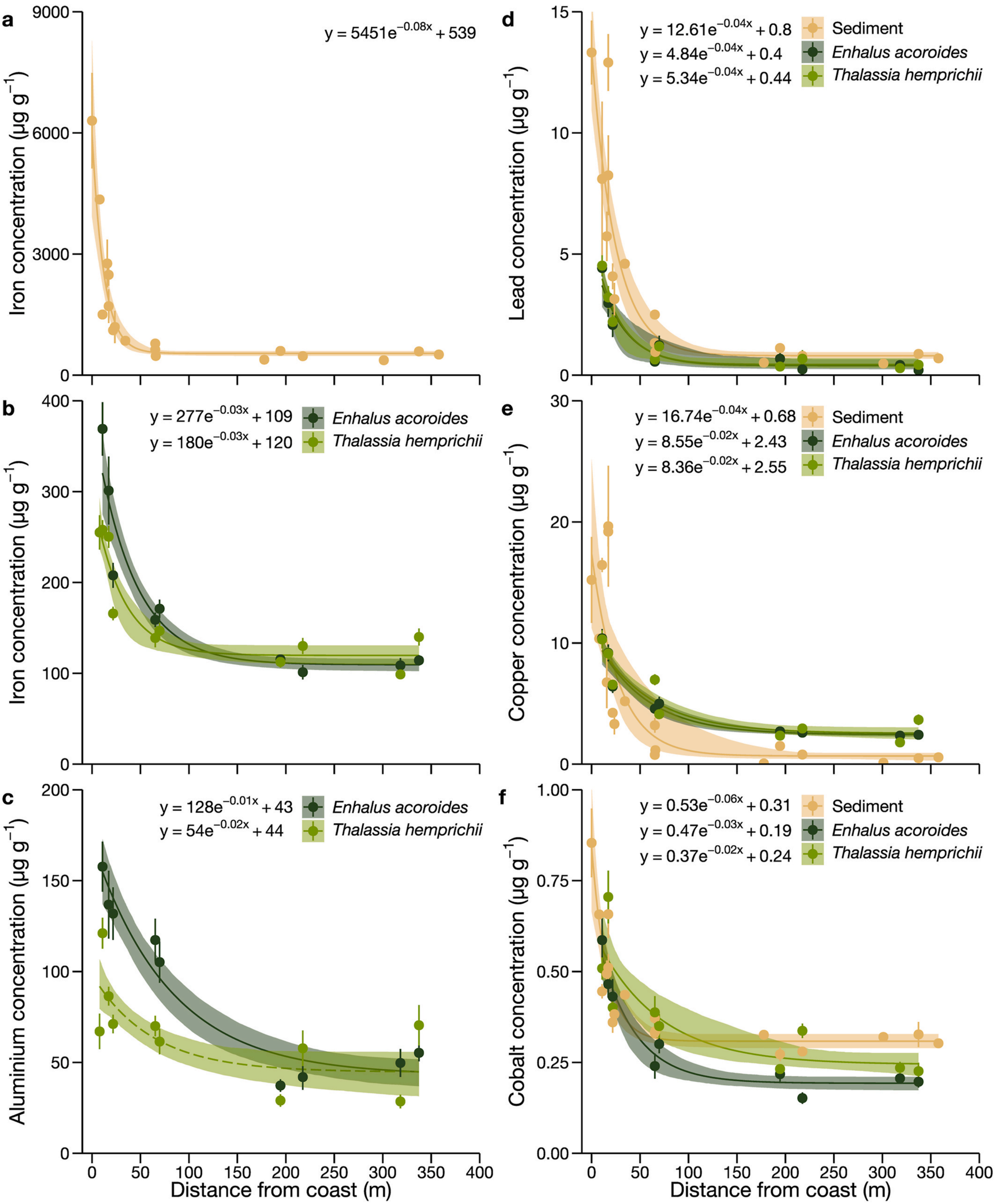

Fig. 3. Iron (**a**, **b**), aluminium (**c**), lead (**d**), copper (**e**) and cobalt (**f**) concentrations with distance from the coast of Barang Lompo. Point-ranges are means ± SE and indicate the concentration at individual sampling sites. Lines and shaded areas are model predictions and 95% confidence intervals respectively. Solid lines represent significant slopes at the 95% confidence level, while dashed lines indicate no significant change with distance.

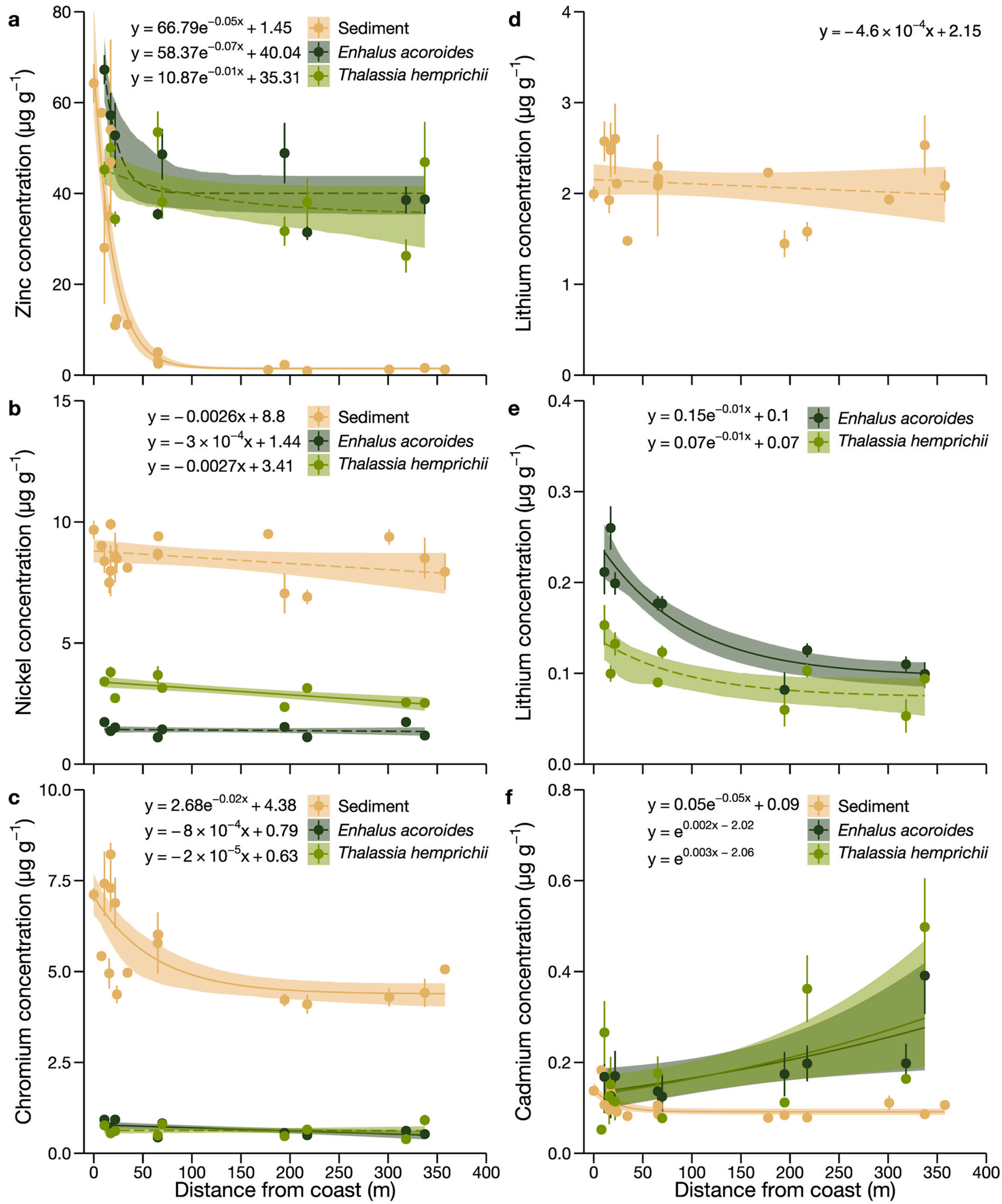

Fig. 4. Zinc (**a**), Nickel (**b**), Chromium (**c**), Lithium (**d**, **e**) and Cadmium (**f**) concentrations with distance from the coast of Barang Lompo. Point-ranges are means ± SE and indicate the concentration at individual sampling sites. Lines and shaded areas are model predictions and 95% confidence intervals respectively. Solid lines represent significant slopes at the 95% confidence level, while dashed lines indicate no significant change with distance.

the island at an exponential rate of 0.02 m^{-1} (t = 2.15, p = 0.04) (Fig. 4c). At its highest, sediment [Cr] was an order of magnitude higher than that of seagrass. Lithium concentration in sediment was, overall, an order of magnitude higher than that in seagrass. [Li] in sediment remained the same regardless of distance from the coast (Fig. 4d). [Cd] in sediment, in an opposite trend to seagrass, increased towards the island at an exponential rate of 0.05 m^{-1} (t = 2.57, p = 0.014) (Fig. 4f).

3.4. Metal content in fish muscle tissue

The MPI revealed a 43% higher overall metal content in *S. canaliculatus* when compared to *C. buchanani* ($X^2_{1,\ 12} = 5.27, p = 0.02$) for the ten metals tested (Fig. 5). The concentrations of the following metals were significantly higher in *S. canaliculatus* than in *C. buchanani*: Al ($X^2_{1,\ 12} = 4.89$, $p = 0.03$), Zn ($X^2_{1,\ 12} = 5.43$, $p = 0.02$), Pb ($X^2_{1,\ 11} = 6.62$, $p = 0.01$), Ni ($X^2_{1,\ 12} = 6.02$, $p = 0.01$) and Co ($X^2_{1,\ 11} = 21.15$, $p < 0.001$). Only [Cu] was significantly higher in *C. buchanani* when compared to *S. canaliculatus* ($X^2_{1,\ 11} = 16.56$, $p < 0.001$). The concentration of Cr, Cd and Li was not significantly different between the two species.

4. Discussion

This study shows that the proximity to a densely populated island substantially affects the surrounding seagrass ecosystem by exponentially increasing heavy metal concentrations in sediment and seagrass. Our model primary producers, the common tropical seagrasses *E. acoroides* and *T. hemprichii*, responded very differently to environmental increases in heavy metal contamination. The former had a much greater affinity for assimilating heavy metals and therefore showed a stronger increase in the concentration of contaminants towards the island. Further, this research provides strong indication of heavy metal contamination in the primary consumers associated with these seagrass meadows, suggesting potential knock-on effects for local fisheries and human health.

Metal concentrations in sediments from around Barang Lompo obtained in this study are intermediate to previous reports worldwide. For example, Cu concentrations from sediment in this study ranged between 0.06 and 35.34 μg g^{-1}. These were much higher than results obtained by Amin et al. (2008) from an area in Dumai, Indonesia classified as unpolluted to moderately polluted (1.67–10.55 μg g^{-1}). However, the results shown here were much lower than those reported from the coast of Semarang, Indonesia, contaminated by wastewater outfall from an electroplating factory (33–72 μg g^{-1}) (Takarina et al., 2004). Cd levels in sediment were below those described in Fanga'uta Lagoon, Tonga and in Laucala Bay, Fiji, both of which have been described as having no significant metal contamination problems despite some degree of anthropogenic enrichment of land-origin (Morrison and Brown, 2003; Morrison et al., 2001). The concentrations in sediment reported here for Fe, Co, Cr, Cu, Ni, Pb and Zn were far lower than those obtained by Chand and Prasad (2013) in Lami Coast, Fiji, an area with significant input from surrounding industries. The surface sediments analysed contained high concentrations of Fe and Al (beyond detection range), which was to be expected as both are major lithogenous components in marine sediment (Fö;rstner & Wittmann, 1981). Nonetheless, mineralogical studies would be required to determine the origin of Fe and could help explain the steep change in the sediment composition away from the island. The strong trend observed for all metals in sediment, except Li and Ni, could be an indication that the sources of those metals' input, both natural and anthropogenic, were similar, while the source of Li and Ni may differ or be affected by different factors (Thibon et al., 2021; Malea et al., 2021).

The comparison of the MPI from each analysed site reflects the overall trend of local metal enrichment along the coastline. The highest MPI values for both sediment and seagrass were observed within 100 m of Barang Lompo (Fig. 2). According to the original proponent of this

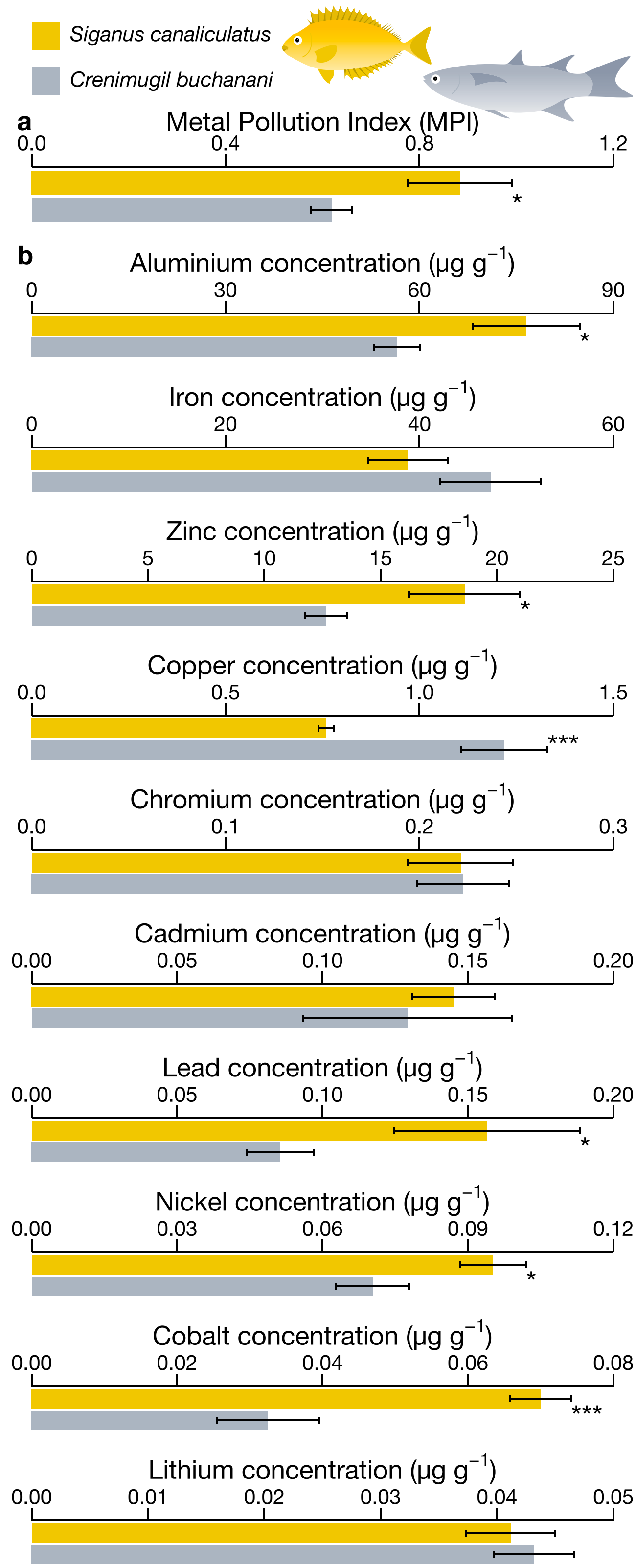

Fig. 5. Metal Pollution Index (**a**) and concentrations of individual metals (**b**) in the muscle tissue of the two primary consumer fish species *Siganus canaliculatus* (yellow) and *Crenimugil buchanani* (grey). Bars and error bars indicate means ± SE. Asterisks represent significance at the 95% confidence level (p < 0.05 *, <0.01 **, <0.001 ***).

pollution index, a value of zero would indicate pristine unpolluted conditions, a value of one would indicate baseline levels of pollutants, and values above one would indicate progressive environmental quality deterioration (Tomlinson et al., 1980). Based on the calculated values, all sites would be deemed contaminated, though to different degrees. Generally, this index does not compare contaminants with any baseline or guidelines, and its proposed threshold classification to differentiate between unpolluted and highly polluted is much too broad for clear definitions. However, the sampling design used here means one can assume levels away from the island - where they remain mostly constant - can be used as a baseline against which coastal levels can be compared. Then, coastal enrichment and pollution increase closest to the coast become evident. Overall, the average metal concentration found in the sediment around Barang Lompo was within the guidelines for protection of aquatic biota according to the Australian and New Zealand Guidelines for Marine Water Quality (ANZECC, 2000). Indonesia has no established concrete limits for toxicity in environmental matrices. When compared to the levels ascertained in the government regulation No. 18/1999 for Toxicity Characteristic Leaching Procedure (TCLP) which determine the quality standards of contaminants in waste, however, the values identified here are far higher than those: Cd = 0,05 $\mu g\ g^{-1}$; Cu = 0,19 $\mu g\ g^{-1}$; Pb = 2,5 $\mu g\ g^{-1}$; Zn = 2,5 $\mu g\ g^{-1}$ (Government of the Republic of Indonesia, 1999).

Metal contents in seagrass leaf tissue were also comparable to those found by previous studies. For example, the average Cu concentration in *E. acoroides* observed here was nearly 3 times higher than the mean value reported for this species in Indonesia (Tables 1 and 2). This study has found higher concentrations of Al, Cr, Ni, and Zn in *E. acoroides* and *T. hemprichii* than ever reported before in Indonesia. Zn concentration, for example, was still lower than those reported in *Posidonia oceanica* in an area of the Mediterranean deemed uncontaminated (114–171 $\mu g\ g^{-1}$) (Campanella et al., 2001). The high Zn and Cu uptake observed in seagrass, compared to the low levels in the environment, is in agreement with previous studies (Schlacher-Hoenlinger and Schlacher, 1998). Both Zn and Cu are required micronutrients for plant and animal life (Schneider et al., 2018). Tracing studies on Zinc accumulation pathways in seagrasses show that this metal is taken up by the surface membrane as a result of exchange adsorption by passive processes and transported into intercellular space where it accumulates (Henkin, 1984; Malea et al., 1995). The same mechanism is involved in the uptake of Cd by the seagrass *Heterozostera tasmanica* (Fabris et al., 1982). Despite not being an essential metal, the concentration of Cd in seagrass increased regardless of the constant low concentration in the surrounding sediment. Whilst this contradicts the finding of linear accumulation described in laboratory settings by Warnau et al. (1996), it may align with the observation that when growing in sediment low in cadmium, seagrasses accumulate Cd primarily in the leaves when exposed to elevated Cd concentrations in the water (Fabris et al., 1982). An analysis of Cd levels on water in the Spermonde Archipelago would be required to confirm this. Unlike Zn and Cd, the uptake of Cu involves ion-exchange processes (Schlacher-Hoenlinger and Schlacher, 1998b) and once absorbed it has a high translocation capacity, as most of the Cu in the plant is in soluble form (Brix and Lyngby, 1982). Cu is believed to have acropetal translocation (Lyngby and Brix, 1982), which is consistent with the correlation between Cu concentrations in the leaves of *E. acoroides* and *T. hemprichii* and the sediment. We found that neither one of the two species analysed here had consistently higher metal concentrations than the other. This was consistent to the findings summarised in Table 1 (see Cd and Zn; Cr,Cu, and Pb in India).

The Indonesian National Standards (INS) (2009) establishes the levels of four metals tin (Sn), mercury (Hg), Cd and Pb, below which fish are deemed safe for human consumption. The mean Cd concentration found in *Siganus canaliculatus* and *Crenimugil buchanani* were 45% and 30% higher than the limit set by the INS (0.1 $\mu g\ g^{-1}$), respectively. The mean Pb concentration of both species, on the other hand, were below the 0.3 $\mu g\ g^{-1}$ limit set by the INS. Further studies, taking biometry and ontogeny into consideration, should be carried out to fully understand the extent of metal pollution in fish in this area.

In conclusion, this study provides baseline levels and accumulation patterns of ten metals in the Spermonde archipelago for the first time. It shows that densely populated coastal areas exponentially increase environmental trace metal content in its surroundings. This study also evidences differences in the uptake and consequent concentration of certain trace metals between the seagrass species *Enhalus acoroides* and *Thalassia hemprichii* leading to differences in vulnerability which can bear ecological repercussions. The spatial gradient observed here may not affect the ecosystem in a distinct fashion but rather expose reef organisms to gradually changing abiotic conditions which could prove detrimental and exclude organisms without the physiological capacity to cope with the changing environment (Teichberg et al., 2018). Further research to identify locally important sources of heavy metal input in the coastal area can prove beneficial in informing local management and legislation.

CRediT authorship contribution statement

Teresa Baptista Nobre: Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Luka Seamus Wright:** Visualization, Formal analysis. **Dominik Kneer:** Methodology, Investigation, Conceptualization. **Sebastian C.A. Ferse:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare no competing interests.

Data availability

The complete datasets and code are available in the GitHub project repository at github.com/TeresaNobre/metal-gradients.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marenvres.2024.106632.

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