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Citation Details

Szafranski, G., Granek, E. F., Hladik, M. L., & Hackett, M. (2023). Non-native Rhizophora mangle as Sinks for Coastal contamination on Molokai, Hawai'i. Environmental Advances, 100459.

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Non-native *Rhizophora mangle* as sinks for coastal contamination on Moloka'i, Hawai'i

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ARTICLE INFO

Keywords: Mangrove roots Microplastic pollution Non-native mangrove Organic contaminant Pesticide Sediment

ABSTRACT

Coastal mangrove forests provide a suite of environmental services, including sequestration of anthropogenic contamination. Yet, research lags on the environmental fate and potential human health risks of mangrovesequestered contaminants in the context of mangrove removal for development and range shifts due to climate change. To address this, we conducted a study on Moloka'i, Hawai'i, comparing microplastic and pesticide contamination in coastal compartments both at areas modified by non-native red mangroves (Rhizophora mangle) and unmodified, open coastline. Sediment, porewater, and mangrove plant tissues were collected to quantify microplastic and pesticide concentrations across ecosystem type. Average microplastics were similar between mangrove (8.89 items/kg) and non-mangrove areas (9.01 items/kg) in sediment and porewater, but mangrove roots were a substantial reservoir of microplastics (2004 items/kg). Additionally, there was a strong relationship between proximity to urban development and microplastics detected. Six pesticides were detected, most commonly the insecticide bifenthrin, found in most sediment samples (11.3 ng/g), all root samples (243.3 ng/g), and one propagule sample (8.60 ng/g). Other pesticides detected with appreciable concentrations include the neonicotinoid insecticide imidacloprid and the legacy insecticide transformation product, p,p'-DDE. The other detections, all at concentrations < 1 ng/g, were p,p'-DDT, trifluralin, and permethrin. The high concentrations of bifenthrin in roots compared to lower concentrations detected in sediment suggest that mangrove roots strongly accumulate some pesticides, indicating mangrove roots as a sink for organic contaminants. Study methods could be applied to other Hawaiian Islands and other locations where mangroves have been introduced to further examine the observed trends. Additional information is needed to investigate the fate and cycling of pesticides and microplastics adhered to mangrove roots, to better inform non-native mangrove removal efforts on Moloka'i and elsewhere.

1. Introduction

Mangrove ecosystems are brackish forests that, due to their unique circumstance as forests growing in the intertidal zone, are subject to highly variable tidal, temperature, and geochemical conditions (Bayen, 2012). Mangroves provide numerous and valuable ecosystem services in tropical and subtropical latitudes and are responsible for a disproportionate amount of coastal biodiversity (Saenger et al. 1983; Lefcheck et al. 2018). This is due in part to the complex and 3-dimensional root baffles formed by mangrove root systems, which provide shelter for numerous species of fish and invertebrates (Nagelkerken, 2009). These baffles also slow the flow of water, which reduces coastal erosion while contributing to the accretion of sediment and sheltering upland areas

from storm surges and extreme weather events (Augustinus 1995; Badola and Hussain 2005; Gilbert and Janssen, 1998). Mangrove root systems enrich the belowground organic matter (OM) of coastal areas, at the same time, sequestering contaminants (Kristensen et al., 2010; Vane et al., 2009) and sheltering downstream ecosystems such as reefs and kelp forests from contaminated runoff (Barbier et al., 2011).

Despite the ecological value that mangroves provide, they are in decline globally with some species already at risk for extinction (Duke, et al., 2007; Polidoro et al., 2010). This is due, primarily, to forest clearing to liberate space for urban development, aquaculture/ agriculture, rising sea levels, or as a source of charcoal (Saenger et al., 1983; Fortes, 1988; Malik et al., 2017; Sasmito et al., 2016). Further, mangroves are frequently proximate to urbanized areas, which applies other

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https://doi.org/10.1016/j.envadv.2023.100459

Received 10 September 2023; Received in revised form 8 November 2023; Accepted 22 November 2023 Available online 8 December 2023

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pressures on mangrove ecosystems such as the intensification of anthropogenic contamination (Defew et al., 2005; Kulkarni et al 2018; Girones et al., 2021). Mangroves possess physical characteristics that lead them to be an endpoint for many of these contaminants, including the physically complex aboveground and extensive belowground root structure produced by mangroves that trap waste and adsorb organic contaminants as well (Kristensen et al., 2010; Martin et al., 2020).

Unfortunately, ecotoxicological research in mangroves lags in many areas, including microplastics and organic contaminants (Deng et al., 2021; Bayen, 2012). Plastics are one class of pollutants frequently found in coastal environments generally, and mangrove ecosystems specifically (Luo et al., 2021). Plastic (both macro and micro plastics) enters the coastal environment from harbor activities, urban runoff, and sewage, and circulates through the environment via sea currents and tidal activity (Thushari and Senevirathna, 2020; Sbrana et al., 2022). Microplastics (MPs) are defined as any plastic equal to or less than 5 mm in length, with larger pieces being defined as macroplastics. When present in the environment, plastics can be consumed by fish and invertebrates such as corals, filling stomachs without providing nutrition (Reichert et al., 2018). Plastics can also adsorb chemical contaminants such as persistent organic pollutants, providing another Pathway for these substances to enter the food chain (Savoca et al., 2021). Further, plastic production has been increasing, currently exceeding 390 tons annually (Hachem, 2023). When plastic trash ends up in mangrove forests, mechanical forces such as weathering from tidal action and exposure to sunlight cause these larger plastics to fragment and break down, eventually into microplastics (Jahnke et al., 2017). While these microplastics may degrade further, potentially into nano-plastics, it is unknown if they will break down into non-plastic substances that may linger in coastal wetland environments such as mangroves for years (Davranche et al., 2020; Paduani et al., 2020).

Organic contaminants are a broad category of chemical species that come from a variety of origins including agriculture, industry, and personal use products (Girones et al., 2021). Contaminants of concern from this category include organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) (Mbusnum et al., 2020). Many of these chemicals have long half-lives and possess characteristics such as hydrophobia which together cause them to persist for long periods of time in mangrove sediment and water (Bayen, 2012). Additionally, organic contaminants have the potential to bioaccumulate up the food chain, threatening mangrove fauna and human populations that rely on mangrove forests for food (Bayen et al., 2019).

Sea level rise is another threat to mangrove forests, particularly those associated with urban environments where they may be subject to coastal squeeze (Schleupner, 2008). This occurs when coastal ecosystems are prevented from retreating landward from rising sea level due to hard urban structure along the landward edge, which is a common circumstance for mangroves (Schleupner, 2008; Tam and Wong, 2000). Simultaneously, warming temperatures due to global climate change (GCC) are allowing mangroves to expand poleward along their original ranges leading to the unique circumstance of non-native mangrove invading and modifying other kinds of ecosystems such as salt marshes (Alongi et al., 2015; Saintilan et al., 2014). Currently, there is little research into how the alterations to the mangrove environment from the changing environmental conditions around mangroves or how mangrove invasion affects the sequestration of pollution. If mangrove invasion increases a coastline's capacity to sequester contamination, it becomes important to understand how that contamination may be released when mangroves are cleared for urban development, harvested for charcoal, or pushed out by rising sea levels.

One area modified by mangroves is Moloka'i, Hawai'i, where red mangroves (*Rhizophora mangle*), hereafter referred to generally as mangroves were introduced in 1902 to address erosion resulting from feral grazing animals and agricultural activities (Allen, 1998). Following introduction, mangroves quickly spread to other islands, altering the Hawaiian coastline (Wester et al., 1981). These alterations include

increased ecosystem complexity, increased porewater salinity, higher sediment organic carbon concentrations, and finer sediments than adjacent non-mangrove ecosystems (Demopoulos and Smith, 2010). Due to their invasive status, mangrove removal has been ongoing around Hawai'i, necessitating an understanding of how mangroves influence the distribution of contamination in the intertidal zone and the risk of contamination release associated with removal.

This study aimed to determine how the presence of non-native mangroves influences the storage and distribution of pollutants in coastal environments to better understand the risk of contamination release during mangrove removal across the Hawaiian Islands. Specifically, this study aimed to answer the following questions:

- 1. Is contamination in areas with mangroves higher than adjacent areas without mangroves?
- 2. In what compartments (sediment, porewater, and coarse and fine roots) of the coastal environment are contaminants concentrating?
- 3. Are concentrations of contaminants correlated with land cover/land use variables?

2. Methods

2.1. Study sites

As mangroves have been removed from many areas in Hawai'i, the southern coastline of Moloka'i was selected due to the presence of remaining large mangrove stands (Fig. 1). Moloka'i also has ongoing agricultural activities, which creates a reasonable expectation for finding some level of pesticide contamination. Moloka'i is the fifth largest and fifth most populated Hawaiian Island with a total size of 673.4 km^2 and a population of 7,287 at the time of the study (U.S. Census Bureau, 2020). The population is concentrated onto a relatively small land area along the southern coastline where the largest town on the island, Kaunakakai, had a population of 3,419 when the study took place (U.S. Census Bureau, 2020). The primary industries on Moloka'i are agriculture and ranching, with tourism comprising only a small part of annual revenue. Agriculture is largely concentrated in the northwestern part of the island. The central and eastern parts of the island are dominated by the Moloka'i forest preserve and are relatively unpopulated. Fishponds dot the coastline along the area selected for the study, with some currently in active use for aquaculture and education. Fishponds operate by using stone blocks to create enclosed areas around a section of intertidal and shallow subtidal zone; entry and exit of water is controlled through gates, which prevents fish from exiting. Several study sites are located within these fishponds.

2.2. Field and Lab Quality Assurance/Quality Control

To prevent plastic contamination from entering samples while processing, several precautions were taken. Dyed pink clothing was used at all times in the lab and field, additionally all clothing and lab coats worn in the lab were cotton. All deionized (DI)-water used to rinse equipment or used as part of processing, as well as all reagents used for processing were passed through a 20 μ sieve before use. All equipment was triple rinsed before use, and for this methodology, rinsing refers to using 20 μ sieved DI water specifically. Airfall controls were prepared using a small amount of sieved DI water and rinsed glassware and were opened each time samples were exposed to the air to account for airborne microplastic particles; this includes glass jars used as environmental airfalls while collecting samples in the field. Finally, procedural controls were used with each round of processing; these controls received the same treatments and reagents as the samples, but did not contain any sample.

For pesticide analysis, all sampling equipment was rinsed daily with filtered DI water stored in acid washed mason jars. All glassware was acid washed by fully submerging in a covered tub containing 10% hydrochloric acid solution for 72 h, and then rinsed with filtered (20 μ m)



Area 1				Area 2				Area 3	Area 3 9-WCR 10-PLA 11-NIP 12-KPK 13-LEN Sea Sea Sea Sea			
1-FMS	2-CCG	3-NPT	4-MSK	5-ALL	6-MI9	7-OLW	8-KWU	9-WCR	10-PLA	11-NIP	12-KPK	13-LEN
Sea	Sea	Sea	Sea	Sea	Sea	Open	Sea	Sea	Sea	Sea	Sea	
	Open	Open	Open	Sea 2	Open		Sea 2	Open			Open	Open
				Open	Land		Open	Land				
							Open 2					

Fig. 1. Location of sampling sites split into three areas, abbreviated site names present in each area, and transect types (open coast=no mangroves, seaward edge of mangroves, and landward edge of mangroves) present at each site. ArcGIS Basemap Source: Esri, Latitude/Longitude: 21.0910° N, 157.0186° W. Specific sampling locations and names left out at landowner request.

DI water and dried prior to field work. Foil used to contain sediment and plant tissue samples was triple rinsed with tap water prior to use.

2.3. Field methods

Samples were collected in March of 2022 from 13 paired open coast and mangrove sites along the inhabited southern shoreline of Moloka'i. Sites were selected based on accessibility and included a continuum of urban development. At each site, up to three \sim 30 meter transects were measured parallel to the shore along open coastline (control), the seaward edge of mangrove stands (sea), and, when accessible, the landward edge of mangrove stands (land). In total, three sites contained all three transects, five sites contained paired mangrove and open coast transects, three sites contained just seaward edge mangrove transects, and two sites contained just open coast transects for a total of 27 transects.

Transects were arranged by placing pink plastic flags 10 meters apart. Sampling points were placed in the root zone along the seaward edge of the mangroves, and along the landward edge of the mangrove zone (land transects), where accessible. At open coast sites, sampling points were placed about one meter below the low tide line, roughly equivalent to the points sampled along seaward edge mangrove

transects.

To survey MPs, hydrophobic and hydrophilic pesticides, sediment and porewater were collected at all transect points. Additionally, root, leaf, and propagule tissue samples were collected at transects where mangroves were present. Porewater samples were collected by inserting an MHE stainless steel PPX36 PushPoint sampler ~25 cm into the sediment. A glass syringe which was used to draw out the porewater was attached to the sampler using Tygon® PVC Tubing. Porewater was then stored in 100 mL glass jars with metal screw tops. Aluminum foil rinsed with ambient sea water at the collection site was inserted below the cap to keep out any contamination from the plastic lining of the cap. Sediment samples were collected using a 20-inch Eijkelkamp Threaded Peat Sampler. To collect the sediment cores, the peat sampler was inserted into the surface of the sediment to a depth of about 30 cm. After collection, the core length and hole depth were recorded and the sediment cores were wrapped tightly in rinsed and pre-labeled foil.

To create a representative root sample, root balls were collected from three randomly selected areas within the root zone of a tree at the transect point. To collect the root balls, a four-inch sediment knife was inserted to the hilt and a roughly cube-shaped chunk of sediment was carved out. Then the three root balls from a single plot were combined and homogenized into one sample before being tightly wrapped in rinsed and pre-labeled foil. Leaf and propagule samples were collected by hand by plucking three randomly selected specimens from different branches or trees adjacent to the transect point. Specimens were then tightly wrapped in rinsed and pre-labeled foil. All samples were transferred to a cooler with ice upon collection.

2.4. Lab methods

2.4.1. Porewater

Porewater MP extraction was accomplished through a series of three sequential density separations where DI water was saturated with NaCl (Fisher Scientific®) to achieve a density of approximately 1.2 g/mL. Saturation was accomplished by slowly adding salt to a 2-liter glass mason jar that was being stirred on a hotplate until the salt no longer fully dissolved into solution. This salt solution was then filtered through a 20 µ sieve prior to use. Following each addition of salt solution, the samples were allowed to settle overnight. The samples were then filtered through a MilliporeSigmaTM IsoporeTM Polycarbonate Membrane Filters, 10 µm filter with the aid of a vacuum filtration apparatus. The filter was then transferred to a rinsed petri-slide for drying. To reduce waste the salt solution was reused for each overnight density separation in a sequence. To accomplish this, the salt solution was kept after the sample filtration process and salt was slowly re-added until saturation was achieved, indicated by the inability for salt to fully dissolve into solution. With the first addition of salt solution 1-2 drops of olive oil were added to the samples to prevent MPs from sticking to the surface of the glass beaker (Karlsson et al., 2017). 50 mL of sieved 30 % hydrogen peroxide solution was added with the third addition of salt solution in the density separation sequence to oxidize any remaining organic matter.

2.4.2. Sediment

Sediment cores were divided in a clean fume hood for organic contaminant and microplastic analysis prior to processing. Cores were first divided vertically into two segments, before being divided horizontally into three \sim 8cm sections using a clean sediment knife (Appendix K). This process resulted in a top, middle and bottom core section for each form of analysis. If the cores were shorter than 25 cm, due to compression or an inability to collect larger cores, the bottom section was omitted.

Microplastics were separated from sediment using a protocol described by da Silve Peas et al., 2022 with small modifications. Briefly, the samples were transferred to a drying oven set at 70° C to dehydrate to a constant weight over three days. After careful homogenization of the

dry sediment, a randomly selected 12-13 g were transferred from the sample to a clean centrifuge tube; this was repeated four times to run a total of \sim 50 g per sample. 35 mL of the prepared zinc chloride (ZnCl₂) solution (Nasco®, density 1.6 g/mL) was then added to each centrifuge tube. The tubes were shaken thoroughly for 1 min to mix the sample and reagent, then centrifuged at 3500 rpm for 15 min. After centrifugation, the supernatant from each tube associated with a single sample was poured through a rinsed 20 μ sieve to collect the MPs. The 20 μ sieve with the remaining sample was then thoroughly rinsed using sieved DI water into a clean and rinsed glass beaker. 50 mL of filtered 30 % hydrogen peroxide (H₂O₂) was then added to the samples to oxidize any adhered organic matter. After being left overnight in the H₂O₂ solution, the contents of the beakers were filtered onto a 10 μ MilliporeSigmaTM Isopore[™] Polycarbonate Membrane Filter using a rinsed vacuum filtration unit and stored in a Petri slide for drying and microscopy. The collected $ZnCl_2$ solution was then filtered through a 10 μ filter with the aid of the vacuum filtration apparatus and re-used up to three times to reduce the waste from and cost of the experiment.

2.4.3. Roots

Before processing, the roots were cleaned and separated for microplastic and organic contaminant analysis. Briefly, the roots were placed on the top of two stacked test sieves with 250- and 125 μ porosity and filtered DI-water was used to rinse away excess sediment and organic matter until no organic material from the root balls was left, then gently scrubbed with a cotton dish scrub, and then rinsed once more to remove any added cotton fibers. For microplastic analysis, roots were separated into coarse roots (thickness greater than a human hair), fine roots (thickness about that of a human hair), and ultra-fine roots (smallest root hairs<human hair). An air fall control was set up near the sink where rinsing took place and the time was recorded to identify how long each sample was exposed to the air. To prevent contamination between sites, sieves were rinsed between samples. Following separation, the roots were repackaged in clean, rinsed foil and frozen. The collected ultra-fine roots were excluded from MP analysis due to the inability to properly rinse off MP contamination that may have been atmospherically deposited from the lab while processing. For organic contaminant and pesticide analysis roots were subjected to the same extraction and quantification methodologies from Black et al. (2023) used for sediment analysis.

Roots were dehydrated to a constant weight in a drying oven at 70°C prior to processing for microplastic analysis. A protocol by Pfeiffer and Fischer (2020) was adopted using sieved sodium hypochlorite (NaClO). Samples were weighed and then placed in a clean glass beaker with 100 ml of bleach where the samples were agitated with a glass stir bar and kept at a heat of 50°C. All samples were digested for a minimum of four hours, if material remained after four hours, samples were digested for an additional hour. Following digestion, the samples were filtered onto a 10 μ filter with the aid of a glass vacuum filtration apparatus and stored in a clean petri-slide for later inspection.

2.4.4. Microplastic analysis

Microplastics were counted and identified using a Zeiss Primostar 3 microscope with Labscope software under both $10 \times$ and $4 \times$ magnification. Any particle that was at or less than 5 mm in length, without cellular structure, even throughout its length, and did not easily break under pressure from a metal probe was identified as a possible microfiber. Another type of microplastic found was films, which were characterized as uniform in color, and difficult to break. Suspected plastics were photographed, after which the color, size, and form were recorded. A subset of these plastics was sent to the Brander Lab at Oregon State University for chemical identification using microscope Fourier transform infrared spectroscopy (μ -FTIR) (Brander et al., 2020).

2.4.5. Standardization

The amount and length of the sediment cores varied due to

G. Szafranski et al.

compaction, which is a natural result of core collection. To account for this, a standardized total of items per kilogram for each transect was calculated through the following formula: sediment characteristics core taken from the middle of the transect and calculated as the difference between the dry weight of the sediment before and after combustion. Three replicates were taken from each of

(1000 / combined weight(g) from all samples intransect) X(Total MP(items) from all samples intransect)

The porewater was similarly standardized to items/liter with the following formula:

the top, middle and bottom sections.

USGS Streamstats (Ries et al., 2004) was used to determine basin characteristics drainage area, mean basin elevation, percent area with

(1000 mL/120 ml, which is the sum of the 40 ml collected from each sample in the transect) X (total MPs from transect)

Roots were standardized to 10 grams using the following formula:

slopes greater than 30 percent, and basin relief divided by basin perimeter for each site. R-studio v. 2022.12.0 (R Core Team, 2021) was

(10 / average weight of samples from transect) X(Total MP(items) of all samples in transect)

2.5. Pesticide analysis

Organic contaminant (pesticide) analysis was quantified by the U.S. Geological Survey Organic Chemistry Research Laboratory in Sacramento, California. In all, 37 sediment samples, 11 root samples, 12 porewater samples, 4 leaf samples, and 5 propagule samples were analyzed. The water samples were concentration via solid phase extraction (Gross et al., 2023). The other, solid samples (roots, leaves, sediment, propagules) were extracted using an organic solvent and co-extracted matrix interferences were removed with carbon (Black et al, 2023). Pesticides were quantitated using both liquid and gas chromatography coupled to tandem mass spectrometry (instrument details can be found elsewhere: Gross et al., 2023; Black et al. 2023). Laboratory QA/QC consisted off adding isotopically labeled surrogates to each sample (all recoveries were within the acceptable range of 70-130%), each batch had at least one laboratory blank and one laboratory replicate if there was sufficient sample mass.

2.6. Percent organic matter

Percent organic matter was calculated using the sediment loss on ignition protocol (Heiri et al., 2001) from a dedicated core collected from each transect. To perform percent organic matter analysis, each layer of the core was homogenized, and a portion of the layer was transferred to a tin weigh boat of known weight. After drying at 70°C for three days, the dry weight for each sample was recorded. Then samples were subjected to 550°C heat for four hours. The dry weight following this combustion process was recorded for each sample. This process was repeated three times. Percent organic matter was determined by calculating the percent difference between the dry weight of the sediment before and after combustion.

2.7. Statistical analysis

Average percent organic matter was determined from a dedicated

used to generate boxplots to visually check basin characteristics' significant differences between mangrove and non-mangrove sites. ArcGIS Pro v.2.8 was used to calculate population and urbanization (percent impervious surface) within one km of the sites. Due to the limited sample size, a generalized linear model (GLM) was selected over other analytical methodologies. This GLM was constructed to investigate how well MP abundance along the coast of Moloka'i was explained by mangrove presence using R-studio, with differences considered significant at or below the 0.05 level for roots, porewater, and sediment. The model included explanatory variables of level of urbanization within one kilometer of the site (percent impervious surface), percent organic matter of the transects, drainage area, average elevation, basin slope, basin relief, population within one kilometer of the site, and position along the coastline (longitudinal site coordinates). Variance inflation factor (VIF) was used to account for multicollinearity between predictor variables by removing variables from the model in a stepwise fashion until all variables had less than three VIF. The model was improved in a stepwise fashion with Akaike Information Criterion (AIC) until no further improvement was achieved (Appendices A).

3. Results

3.1. Microplastic Totals

Microplastics were found in sediment at 10 of the 13 sites, along 21 of the 27 transects in porewater at 12 of the 13 sites, and nine of the 27 transects, and in roots at (Fig. 2, Table 1). The average items of microplastics found per transect after standardization to items/kg (in sediment), items/L (in porewater), and items/10 g (in roots) were 8.49 items/kg in sediment; 16.36 items/L in porewater, 252.08 items/10 g in coarse roots and 122.66 items/10 g in fine roots (Table 2). Values from a single transect contribute disproportionately to the higher porewater concentration (Fig. 2). Sediment microplastics were most abundant at sites 2-CCG and 5-ALL, both of which were near the major population center of Moloka'i (Fig. 2). In porewater, the highest concentration of MPs was found at site 6-MI9 with plastics found only in the land transect of the site (Fig. 2). For sediment, sea transects had an average of 7.67



Fig. 2. Total microplastics for a) porewater (items/L) and sediment (items/kg) and b) fine and coarse roots (items/10g). Dashed lines indicate sea transects, solid bars represent land transects, and outlined bars represent open transects. Error bars represent standard error.

items/kg, while open transects averaged 9.01 items/kg and land transects averaged 10.12 items/kg (Table 2). In porewater there were an average of 10.89 items/L in the sea transect, 9.85 items/L in the open transect, and 63.89 item/kg in the land transect (Table 2). Fine roots had an abundance of 125.12 items/10 g at sea transects and 110.32 items/10 g at land transects (Table 2). Coarse roots on average had higher MPs than fine roots, with 212.12 items/10 g at sea transects and 451.89 items/10 g at land transects (Table 2). For both fine and coarse roots, 3-NPT-Land had the highest abundance of observed MPs. Roots overall had much higher plastic abundance when standardized to items per 10 grams than sediment and porewater microplastics (Fig. 2).

3.2. Microplastic form and color

Fibers, films, and fragments were the microplastic forms detected in this study, with fibers being predominant in sediment (93%), porewater (96%), and root (80%) samples (Fig. 3). Films were the second most commonly detected microplastic in sediment (6%) and porewater (3.6%), and there was a solitary fragment found in sediment. Fragments were the second most commonly detected microplastic in roots (20%), and films were not detected in roots. Microplastics were primarily clear

in sediment (\sim 55%), porewater (\sim 53%), and roots (89%). The next most frequent color was blue for porewater (35%), tan for sediment (27%), and yellow (9%) for roots. Gray fibers were also detected in low numbers in sediment and porewater, and orange was detected in roots (3%).

3.3. Relationship between sediment, porewater, and root microplastics, mangrove presence, and basin characteristics

No significant differences (p < 0.05) in basin characteristics were observed between mangrove and non-mangrove areas, nor between fishpond and open coast areas (Appendices C). The results for percent organic matter, percent impervious surface within one kilometer, and population within one kilometer are presented in Table 3. The relationships between sediment and porewater MPs and percent impervious surface, basin characteristics, population within 1 km, and fishpond presence were explored through linear regression. There was no significant relationship between the number of microplastics in porewater, coarse, or fine roots and any of these parameters (P >0.05) (Appendices D). Regression analysis using AIC identified the most appropriate predictor of sediment MP abundance as urbanization. A

Unadjusted total microplastics from all compartments: sediment, porewater, and coarse and fine roots (items/Transect).

Transect	Sediment Plastics (items/ Transect)	Porewater Plastics (items/ Transect)	Coarse Root Plastics (items/ Transect)	Fine Root Plastics (items/ Transect)
1-FMS-	0	3	2	0
Sea 2-CCG-	4	2	N/A	N/A
Open 2-CCG-	9	1	5	1
Sea 3-NPT-	8	1	N/A	N/A
3-NPT-	7	4	3	3
3-NPT-	6	0	2	0
4-MSK-	1	4	N/A	N/A
4-MSK- Sea	4	1	0	3
5-ALL- Sea	7	0	3	0
5-ALL- Open	2	1	N/A	N/A
5-ALL- Sea2	0	2	N/A	N/A
6-MI9- Open	1	0	N/A	N/A
6-MI9- Land	0	17	1	2
6-MI9- Sea	1	0	0	1
7-OLW- Open	3	2	N/A	N/A
8-KWU- Sea	1	2	N/A	N/A
8-KWU- Sea2	1	0	1	0
8-KWU- Open	2	3	N/A	N/A
8-KWU- Open2	5	0	N/A	N/A
9-WCR- Open	1	0	N/A	N/A
9-WCR- Land	4	2	N/A	N/A
9-WCR- Sea	2	0	N/A	N/A
10-PLA- Sea	1	0	4	3
11-NIP- Sea	0	3	0	1
12-KPK- Open	0	2	N/A	N/A
12-KPK- Sea	0	4	0	0
13-LEN- Open	4	1	N/A	N/A
Total	74	55	21	14

Table 2

Average micropl	astics for porewater	(items/L), sedimer	ıt (items/kg)	, fine roots
(items/10 g), an	d coarse roots (items	s/10g) at sea, open	, and land tr	ansects.

Average MPs by Transect Type									
Compartment	Sea	Open	Land	All					
Sediment (items/kg)	7.67	9.01	10.11	8.49					
Porewater (items/L)	10.89	9.85	63.89	16.36					
Fine Root (items/10g)	125.12	N/A	110.32	122.66					
Coarse Root (items/10g)	212.12	N/A	451.89	252.08					

positive linear relationship was found between sediment MPs and percent impervious surface within one kilometer ($R^2 = 0.33 P = 0.00096$, F=14,25) (Appendix J). The selected model meets the assumptions of linear regression, i.e., normality of residuals (Shapiro-Wilks: P=0.08, W=0.93263), Homoscedasticity (F test for variance: P=0.2967, ratio of variances=1.86), no multicollinearity (values with VIF>3 removed from model), and linearity in the relationship between predictor and response variables.

3.4. Microplastic type and contamination

18.3% of MPs found in this study across samples and controls were analyzed using μ FTIR, which identified ~47.5% of analyzed items as synthetic and semi-synthetic, ~29.5% as anthropogenically modified materials, and 23% as natural. The most common types of plastic were polyethylene terephthalate (PET) (31%), polyamide (24%), and polypropylene (13%). Other synthetics include polyvinyl alcohol, polyesterterpthalate, Teflon, and polyester. Analysis of procedural and airfall controls found that, on average, 1.4 MPs were present per control sample (Table 4). Most contamination was airfall from the laboratory.

For sediment and porewater analysis, when adjusted to MPs per transect, an average of 1.62 items/transect were found in the on-site airfall controls, 4.96 items/transect were found in the sediment sample partitioning controls, 0.73 items/transect were found in the sediment processing/MP extraction controls, and 0.73 items/transect were found in porewater processing/MP extraction. For roots, 10 MPs were found in the sample processing, digestions, and filtering airfalls for an average of 0.83 items/transect; and for the procedural controls, 4 MPs were found for an average of 0.33 items/transect. Porewater partitioning was done at the sample processing step as part of the methodology and was exposed to air very little during the collection process on Hawai'i. The average porewater MPs from processing was 0.73 MPs per transect.

3.5. Organic contaminants (Pesticides)

Each sample was analyzed for 178 pesticides as part of a multiresidue pesticide analysis; six different pesticides were detected across 69 samples. The average concentrations and number of detections per contaminant type is summarized in Table 5. The most commonly detected contaminant was bifenthrin, found in 96 % of sediment samples, 100 % of root samples, and 20 % of propagule samples, but not found in any leaf or porewater samples. The next most common detection was p,p'-DDE (N=7), followed by three contaminants which had 3 or less detections and average concentrations less than one ng/g. The most notable result was the substantial difference between the average concentration of Bifenthrin in roots (243.31 ng/g) versus in sediment (Open=9.29 ng/g; Sea=11.30 ng/g) (Table 5).

4. Discussion

Both MPs and organic contaminants are underreported from the Hawaiian Islands, and this study represents the first report of MPs and pesticides on Moloka'i. This study also represents one of the first reports of MPs and pesticides in non-native mangrove forest generally. Microplastic numbers observed in this study were numerically low compared to results reported globally (Deng et al., 2021). Previous work reports MPs in mangrove sediments ranging from a low of 1.22 items per kilogram to as high as 6390 items per kilogram (Deng et al., 2021). Approximately 5 of the 39 investigations reported fewer average plastics than found in this study (8.49 items/kg) (Deng et al., 2021).

4.1. Sediment and basin characteristics

As discussed above, Moloka'i is a relatively unpopulated island compared to others in the Hawaiian Islands. The population is clustered

Fig. 3. Distribution of microplastics in sediment, porewater, and roots by form and color.

Percent organic matter, impervious surface, and human population within one km of each site and basin characteristics of slope, relief, drainage area, and elevation by site.

Predictor Variable	1-FMS	2-CCG	3-NPT	4-MSK	5-ALL	6-MI9	7-OLW	8-KWU	9-WCR	10-PLA	11-NIP	12-KPK	13-LEN
Percent OM Average	5.23	6.07	15.05	7.72	7.83	7.54	5.86	5.77	7.92	4.47	5.48	5.47	11.18
Percent OM Top	5.23	9.42	5.28	6.25	8.20	6.29	11.27	5.88	9.88	7.16	5.19	17.66	6.29
Percent OM Middle		5.22	4.98	5.78	12.54	6.08	8.04	5.91	13.37		4.61	5.33	7.78
Percent OM Bottom		5.86	5.68	73.78				5.58	11.00			4.66	
percent impervious	0.63	3.09	9.44	3.69	1.47	0.77	0.98	1.09	2.17	1.2	0.74	1.14	0.96
population within 1 km2	0	242	647	253	30	1	34	13	99	1	14	67	19
Slope (Percent)	144	287	354	374	641	1020	820	924	1170	1160	1000	985	731
Relief (Feet)	1500	1030	4130	437	2270	2840	4950	1580	4600	4470	2140	2850	2010
Drainage Area (Mi ²)	14	0.99	9.04	0.21	0.97	1.59	6.66	0.46	2.02	1.72	0.63	0.54	0.49
Elevation (Feet)	498	388	1780	173	1100	928	2430	531	1920	1730	601	1060	812

around Kaunakakai, the main settlement on the island, which is also where the harbor is located. Elevation on Moloka'i increases towards the center of the island, and is higher on the eastern half of the island, which is dominated by an extinct volcano (Moore and Krivoy, 1965). Relief and elevation are highest in the basin containing the site 7-OLW, and are generally higher at the more centrally located sites. Similarly, the highest slopes are at the basins containing sites 9-WCR and 10-PLA and are generally higher at the eastern sites when compared to the western sites. However, these trends in slope and elevation did not correlate significantly with microplastic distribution further indicating that proximity to urban development is the most important factor controlling MP distribution along Moloka'i.

4.2. Microplastics

The linear relationship between impervious surfaces, population within one km and sediment MPs corresponds with global trends for mangrove MPs, and this trend combined with the generally higher concentrations at landward sites indicates a terrestrial rather than marine source of microplastics. In other studies, the strongest factors determining microplastic distribution in mangrove sediments are land use activities such as proximity to urban development, population dense areas, and outputs from sewage (Deng et al., 2021). Shipping is another source of MPs into the mangrove environment; with the main port of Molokai adjacent to Kaunakakai, shipping may contribute to the Molokai MP contamination along the coast (Deng et al., 2021). Further, the two most common types of polymers detected, PET (31 %) and polypropylene (13%), are frequent by-products of single use plastics (Chen et al., 2021). The other commonly detected plastic, polyamide (24 %), is a component of nylon and its source is associated with clothing and fishing nets. Together these plastics account for 68 % of plastics found in the sediment and porewater samples. The most commonly found microplastic shape found across all compartments was fibers (~80-93 %) followed by films in sediment (6 %) and porewater (4 %) and

Average microplastics (MP) contamination found in controls per transect from sample collection, sample processing, and sample partitioning. Control values were added together to calculate the total MPs contamination for each porewater and sediment sample (last column). *All samples associated to one control bottle that burst in freezer. **Lost in transport from Hawaii.

Site	On-site airfall control (# of MPs per transect)	Sediment Partitioning Airfall (Total # of MPs/ associated transects)	Sediment Processing Controls (# of MPs total/ transect)	Total MPs from processing and collection sediment (# of MPs/ transect)	PW Processing per transect
1-FMS-	1	6.94*	0.39	8.33	0.99
Open					
2-CCG-	1	N/A*	0.39	1.39	0.99
2-CCG-	0	N/A*	0.75	0.75	0.99
Sea					
3-NPT- Open	0	4	1.71	5.71	0.75
3-NPT-	5	0	1.2	6.2	0.75
Land	0	16 601		10.0	0.00
3-NPT- Sea	2	16.63*	1.17	19.8	0.99
4-MSK-	2	0	0.48	2.48	0.57
Open					
4-MSK- Sea	2	0	0.99	2.99	0.57
5-ALL-	0	4.43*	1.52	5.95	0.99
Sea	0		0.00	0.00	0.00
5-ALL- Open	0	N/A*	0.98	0.98	0.99
5-ALL-	2	7.5	0.91	10.41	0.75
Sea2	0	F 10t	0.00	F 41	0.00
Open	0	5.18	0.23	5.41	0.99
6-MI9-	0	5.18*	0.3	5.48	0
Land	1	11 6 4+	0.0	10.04	0.00
Sea	1	11.04	0.2	12.84	0.99
7-OLW-	1	5.91*	0.49	7.4	0
Open 8 KWII	n	25	0.75	6.25	0.00
Sea	2	5.5	0.75	0.23	0.99
8-KWU-	1	6.65*	0.77	8.42	0.57
Sea2 8-KWII-	1	35	0 44	4 94	0.57
Open	1	5.5	0.77	ч.)-т	0.37
8-KWU-	1	0	0.42	1.42	0.57
Open2 9-WCR-	0	6.4*	0.64	7.04	0.6
Open	0	0.1		,	010
9-WCR-	0	0	1.01	1.01	0.6
Land 9-WCR-	N/A**	6.4*	0.53	6.93	0.99
Sea		0.1	0.00	0150	0.55
10-PLA-	0	7.2*	0.59	7.79	0.99
Sea 11-NIP-	2	0	0.83	2.83	0.51
Sea					
12-KPK-	2	10.44*	0.56	13	0.99
Open 12-KPK-	0	7.5	0.44	7.94	0.51
Sea					
13-LEN-	16	0	0.91	16.91	0.57
Average	1.62	4.96	0.73	6.69	0.73

fragments in roots (20 %). Fibers being the predominant shape is typical for marine environments, and fibers are often created via shedding from clothing during the washing process or from fishing gear (Shim et al., 2018). The profile of MP shape and chemical species seen in this study suggests the MPs likely originated from single use plastics and clothing, which indicates the association between MPs and impervious surfaces is

logical. Clear plastics were the most commonly found color (53-88 %), followed by blue (35 %) for porewater, tan (27% for sediments), and yellow (9 %) for roots. Clear, white, black and blue plastics are some of the most frequently found colors for plastics in the marine environment (Hidalgo-Ruz et al., 2012; Ugwu et al., 2021). MP color has implications for its impact on the marine environment, with some studies finding that fish are more likely to confuse those colors with prey or other food items (Boerger et al., 2010).

MPs enter mangroves from both seaward and landward directions, which contributes to the high levels of MPs seen in mangroves broadly (Deng et al., 2021). The lower levels of contamination found in this study for both MPs and organic contaminants could be due to the small population on Moloka'i, which may contribute to lower contamination from land. The sheltered location of Moloka'i within the center of the Hawaiian Islands may also contribute to the smaller number of MPs seen in this study if the other islands are entraining MP contamination from oceanic sources. However, the lack of previous work investigating MPs across the islands complicates efforts to understand how localized currents may affect the distribution of MPs along the coast. Further research could help to understand if contamination is low on Moloka'i only or broadly throughout Hawaii.

Roots had much higher microplastic abundances compared to sediment (average sediment MPs = 9.2 items/kg, average coarse roots = 252.08 items/10 g, average fine roots = 122.66 items/10 g) (Appendix F). Roots were rinsed thoroughly prior to analysis and those that could not be rinsed were excluded, so plastics found in samples likely represent particles that were strongly adhered to root surfaces or partially embedded into root tissues. MP abundance was not significantly different between mangrove and non-mangrove sediment, but because roots were largely removed via centrifugation when processing sediment, roots may represent a notable MP sink in mangrove sediment (that were not represented in our sediment results due to the sample processing methodology used). Unlike for sediment samples, there was no significant relationship between population or impervious surface within one kilometer of the sites and root MP abundance. However, the highest plastics overall were found at the site 3-NPT which is the site closest to the main population center on Moloka'i. Given the small sample size of roots, future research may reveal that root plastics are correlated with proximity to urban area as seen with sediment MPs.

4.3. Pesticides

Bifenthrin, a pyrethroid insecticide that sees widespread use and is commonly found in the environment (Delgado-Moreno et al., 2011), was the most commonly detected pesticide in this study.Bifenthrin was found in all root samples at relatively higher concentrations (average=243.31 ng/g), but was found in most sediment samples at much lower concentrations (average = 11.17 ng/g) (Table 5). This suggests that bifenthrin is concentrated at mangrove roots. Bifenthrin is toxic to aquatic organisms, with lethal concentrations reported from *ex-situ* studies to be as low as 0.10 ng/g for some species of fish and 8 ng/g for aquatic invertebrates, suggesting that the concentrations seen in this study (0.16 – 870.79 ng/g) may be at environmentally toxic levels (Yang et al., 2018; Johnson et al., 2010; Anderson et al., 2008).

Pyrethroid insecticides including bifenthrin are hydrophobic and insoluble in water and tend to bind to organic matter in sediments. Higher OM content has been found to increase the retention of persistant organic pollutants (POPs) while reducing its bioavailability (Gammon et al. 2012; Maul et al., 2008; Zhu et al., 2014). Further, Hawaiian mangroves may lack the specialized communities of detritivores that have co-evolved to consume and break down the tannin- and lignin- rich detritus that mangroves produce (Kristensen et al., 2008; Demopoulos and Smith, 2007; Demopoulos and Smith, 2010). Understanding how the unique faunal assemblages interact with the accumulation and consumption of sediment OM may be important for understanding the bioavailability of bifenthrin and other organic contaminants in the

Average pesticide concentrations (by transect type and environmental compartment) detected in the subset of samples analyzed (ng/L for porewater; ng/g dry weight for sediment, roots, propagules and leaves), including sample size (N), and number of detections (following concentrations). Concentration ranges are not provided when only one sample had a detection.

Compartment (# of samples analyzed	Bifenthrin AVG (NG/G, NG/L) (RANGE, # OF DETECTIONS))	Imidacloprid AVG (NG/ G, NG/L) (Range, # of Detections)	Permethrin AVG (NG/ G, NG/L) (Range, # of detections)	P.P'-DDE AVG (NG/ G, NG/L) (Range, # of detections)	P.P'-DDT AVG (NG/ G, NG/L) (Range, # of detections)	Trifluralin AVG (NG/ G, NG/L) (Range, # of detections)
SEDIMENT SEA (N=22)	12.27 (0 - 94.96, 22)	0	0	0.62 (0.20 - 1.03, 2)	0	0.40 (1)
SEDIMENT OPEN (N=13)	9.29 (0.16 - 39.23, 13)	0	0.24 (1)	0.55 (0.32 - 0.95, 3)	0.32 (1)	0.30 (0.20 - 0.39, 2)
SEDIMENT LAND (N=2)	0.08 (0 - 0.16, 2)	0	0	0	0	0
Porewater (N=12)	0	37.1 (30.4 - 43.7, 2)	0	0	0	0
Roots (N=11)	243.31 (<i>34.95- 870.79,</i> 11)	0	0	11.57 (2.82 - 20.3, 2)	0	0
Propagule (N=5)	8.60 (1)	0	0	0	0	0
Leaf (N=4)	0	0	0	0	0	0

future.

Two other compounds were found at concentrations ≥ 1.0 ng/L, imidacloprid and p,p'-DDE. Imidacloprid, a hydrophilic, neonicotinoid insecticide that is thought to have lower toxicity to fish and mammals, but is highly toxic to insects (Pietrzak et al., 2020), was detected in porewater from KWU Sea and KPK Open transects. These sites are located away from Kaunakakai, but near a facility that is suspected to utilize imidacloprid. How imidacloprid cycles through the environment, including its relationship to sediment organic matter, is poorly understood (Pietrzak et al., 2020). P,p'-DDE, a highly persistent breakdown product of p,p'-DDT that adsorbs strongly to sediment (Pereira et al., 1996) and was banned in the United States in 1972, , was detected in a number of locations (N=7), at concentrations of 2.82 and 20.3 ng/g in the root compartment.

The scope of the organic contaminant analysis was conducted on a limited number of samples so additional studies could confirm how widespread contamination is along the coast of Moloka'i. Bifenthrin was found in 100 % of the roots analyzed, but was retained poorly in other ecosystem compartments in this study. This result matches results of a previous study that confirmed the tendency of the mangrove root layer to bind organic contaminants through various mechanisms such as the Fe plaque formed on roots through natural processes (Robin and Marchand, 2022). Based on these results, additional data could be collected and refined to determine the bioavailability and long-term fate of bifenthrin and other pyrethroid pesticides bound to mangrove roots to inform plans for ongoing and future mangrove removals. For example, we do not yet know whether bifenthrin remains adhered to roots while breaking down to less toxic substances over time. Cutting mangrove trees flush to the sediment may help to maintain the benefits of mangroves (pesticide sequestration, erosion control) while eliminating a portion of its detriments (overgrowing fishponds, harboring invasive species. If bifenthrin is found to remain bioavailable while adhered to roots, then complete removal of mangrove plants could be considered to prevent the contaminants from entering the food web. Further, mangrove removal generally may release contaminants into the environment, so ongoing mangrove removal on Moloka'i provides an opportunity to study how non-native mangrove systems store and then possibly release contaminants.

4.4. Study limitations

Lack of access to transects along the landward edge of the mangrove sites was a limitation of this study. Mangroves are known to filter and sequester contaminants moving across the intertidal zone from the land towards the sea (Kulkarni et al., 2018). The highest microplastics reported in the study were found within porewater at the land transect 5-MI9 porewater (141.67 items/L). Overall land transects had higher average MPs in sediment and much higher MPs in porewater (N=2), but

the high porewater concentrations were likely partially the result of a single high detection (Fig. 2). With access to only three land transects, the land data are limited. Greater access to land transects could have helped confirm whether higher concentrations of microplastics were measured landward of mangroves and would allow a deeper exploration of the role mangroves play in filtering contaminated runoff.

5. Conclusions

This study set out to determine which ecological compartments are sinks for contaminants along the coastline in Moloka'i and whether land use or non-native mangroves have a bigger role in determining contamination intensity. In this study it was found that mangrove roots are potentially significant sinks for both microplastic and pesticide contamination. However, the evidence for a large number of MPs adhered to roots is based off of a small sample size. For MPs not adhered to roots the amount of impervious surface around the site was more important, suggesting that land use may play an important role in determining MP concentrations along the coast. With averages of 8.49 items/kg for sediment and 16.36 items/L for porewater, contamination is overall lower on Moloka'i than other previously studied locations around the world (Deng et al., 2021; Maghsodian et al., 2022), indicating proximity to urban areas may be more important for the distribution of MPs along the coast than mangrove presence. Bifenthrin was the most frequently detected pesticide and was found in all but one of the samples and at all 15 transects analyzed. This study represents the first examination of these contaminants together specifically on Moloka'i, and more broadly in the Hawaiian Islands, providing important information on the relative concentrations and locations of contaminations across coastal compartments. Additional research could determine if decaying mangrove roots are an ongoing source of bifenthrin contamination or remain a sink until bifenthrin breaks down. Such information could help inform managers in Hawaii about the best approaches for future mangrove management. Larger sample sizes could also more clearly define the relationship between mangrove trees and coastal contamination on Moloka'i, particularly if additional samples were collected from behind mangrove stands. Additional information around all Hawaiian Islands could help contextualize the Moloka'i results. Collection of more root samples and further refinement of root digestion methodologies could help better understand how roots sequester microplastic and organic contamination. Further bifenthrin concentration assessments in mangrove species and adjacent ecosystems could determine its cycling through coastal ecosystems.

CRediT authorship contribution statement

Geoffrey Szafranski: Conceptualization, Methodology, Formal analysis, Data curation, Writing – original draft, Funding acquisition,

Investigation. Elise F. Granek: Writing – review & editing, Methodology, Project administration, Supervision, Resources. Michelle L. Hladik: Formal analysis, Writing – review & editing, Resources. Mia Hackett: Methodology, Visualization, Writing – original draft.

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.

Data availability

Data will be made available on request.

Acknowledgments

Funding for this project was provided by the Society of Wetland

Scientists, The Society of Integrative and Comparative Biology, Sigma Xi, and the Ed and Olive Bushby Scholarship. Thank you for your support. We thank Paul Hurst and Naomi Granek Brown for providing crucial assistance in the field with sample collection; Paul Hurst and Summer Traylor for training on microscope and chemical digestion methodology; Becca Wilson and the rest of the Portland State University Chemistry Stockroom team for their incredible patience and expertise while developed the microplastic processing protocols; Elise Yu for amazing assistance throughout much of the laboratory processing; Dr. Richard MacKenzie for guiding the the Moloka'i study design, and Dr. Yangdong Pan for assisting with statistical analysis. We are grateful for the analytical work conducted on our samples: pesticide sample extractions at the USGS California Water Science Center were done by C. Sanders, E. LaBarbera, M. De Parsia, and M. Uychutin; µ-FTIR analysis on microplastics samples were completed in Dr. Susanne Brander's Ecotoxicology and Environmental Stress Lab at Oregon State University by Jillian Ragno. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

Annendix A	AIC values fo	or sediment and	norewater from	hackwards	stenwise r	nodeling
appendix m.	mo values ie	n scument und	porewater nom	Duckwaras	stepwise i	noucing

Variables in Sediment Model	AIC
Transect + Fishpond Presence + Longitude + Impervious Surface + OM	-51.6
Transect + Longitude + Impervious Surface + OM	-53.57
Transect + Impervious Surface + OM	-54.61
Transect + Impervious Surface	-54.84
Impervious Surface	-55.54
Variables in Sediment Model	AIC
Transect + Fishpond Presence + Longitude + Impervious Surface + OM	33.21
Mangrove Presence + Fishpond + Longitude + Impervious Surface	31.22
Mangrove Presence + Longitude + Impervious Surface	29.23
Longitude + Impervious Surface	27.25

Appendix B. RStudio Code demonstrating linear model

mod.aic5<-lm(logsed1k~coast+mang+fishpond+Long+logperv+percent.om, data=proj) rst.l5<-summary(mod.aic5) round(rst.l5\$coefficients,5) summary(mod.aic5) vif(mod.aic5) mod.finalaic<-step(mod.aic5) summary(mod.finalaic) final<-median(predict(mod.finalaic)) (final1<-residuals(mod.finalaic)[predict(mod.finalaic)>final])

(final2<-residuals(mod.finalaic)[predict(mod.finalaic)<final]) var.test(final1,final2) shapiro.test(residuals(mod.finalaic)) anova(mod.finalaic, mod.aic5) vif(mod.finalaic) par(mfrow=c(1,1)) plot(logsed1k~logperv, data=proj, xlab="Impervious Surface", ylab="Sediment Microplastics") abline(mod.finalaic, lwd=3, col='red') Appendix C. Box plots demonstrating differences between basin characteristics for open (non-mangrove) and mangrove (present) sites, and fishpond (1) and non-fishpond (0) sites. Box plots depict the minimum, first quartile, median, third quartile, and maximum, with outliers depicted as single points

Appendix D. Non-significant relationships for porewater and roots

P-Values, degrees of freedom, and F-Statistics for non-significant linear relationships in sediment and porewater

Predictor	Porewater P-value	Porewater F-Statistic	Fine Roots P-Value	Fine Roots F-Statistic	Coarse Roots P-Value	Coarse Roots F-Statistic
Percent Impervious Surface	0.93	.0072, 25	0.6592	0.2066,10	0.5024	0.482,10
Population	0.819	0.0534, 25	0.8435	0.04107,10	0.1506	2.424,10
Percent OM	0.724	0.127,25	0.9944	5.269e-05,10	0.3851	0.8248,10
						(continued on next page)

(continued)

Predictor	Porewater P-value	Porewater F-Statistic	Fine Roots P-Value	Fine Roots F-Statistic	Coarse Roots P-Value	Coarse Roots F-Statistic
Fishpond Presence	0.963	0.00218,25	0.5337	0.4155,10	0.6681	0.1951,10
Slope	0.4944	0.481,25	0.3999	0.7732,10	0.9285	0.008473,10
Relief	0.2685	1.28,25	0.173	2.154,10	0.63	0.2468,10
Drainage Area	0.6209	0.2508,25	0.7758	0.08559,10	0.06204	4.412,10
Elevation	0.3169	1.043,25	0.2767	1.324,10	0.4346	0.6626,10

Appendix E. Table of raw data for transect physical characteristics

Sites	Fishpond	Percent Impervious	Population Within 1	Percent	Drainage Area	Elevation	Slope	Relief
	Presence	Surface	km ²	OM	(Mi ²)	(Feet)	(Percent)	(Feet)
1-FMS-Open	No	0.63	0	5.23	14	498	144	1500
2-CCG-Open	No	3.09	242	6.83	0.99	388	287	1030
2-CCG-Sea	No	3.09	242	5.31	0.99	388	287	1030
3-NPT-Open	No	9.44	647	28.6	9.04	1780	354	4130
3-NPT-Land	No	9.44	647	10.37	9.04	1780	354	4130
3-NPT-Sea	No	9.44	647	6.18	9.04	1780	354	4130
4-MSK-Open	No	3.69	253	9.66	0.21	173	374	437
4-MSK-Sea	Yes	3.69	253	5.79	0.21	173	374	437
5-ALL-Sea	Yes	1.47	30	11.42	0.97	1100	641	2270
5-ALL-Open	Yes	1.47	30	7.16	0.97	1100	641	2270
5-ALL-Sea2	Yes	1.47	30	4.9	0.97	1100	641	2270
6-MI9-Open	No	0.77	1	9.22	1.59	928	1020	2840
6-MI9-Land	No	0.77	1	7.04	1.59	928	1020	2840
6-MI9-Sea	No	0.77	1	6.37	1.59	928	1020	2840
7-OLW-Open	No	0.98	34	5.86	6.66	2430	820	4950
8-KWU-Sea	Yes	1.09	13	5.05	0.46	531	924	1580
8-KWU-Sea2	Yes	1.09	13	6.49	0.46	531	924	1580
8-KWU-	Yes	1.09	13	4.1	0.46	531	924	1580
Open								
8-KWU-	Yes	1.09	13	7.44	0.46	531	924	1580
Open2								
9-WCR-Open	No	2.17	99	5.65	2.02	1920	1170	4600
9-WCR-Land	No	2.17	99	10.72	2.02	1920	1170	4600
9-WCR-Sea	No	2.17	99	7.39	2.02	1920	1170	4600
10-PLA-Sea	Yes	1.2	1	4.47	1.72	1730	1160	4470
11-NIP-Sea	Yes	0.74	14	5.48	0.63	601	1000	2140
12-KPK-	Yes	1.14	67	5.54	0.54	1060	985	2850
Open								
12-KPK-Sea	Yes	1.14	67	5.4	0.54	1060	985	2850
13-LEN-	Yes	0.96	19	11.18	0.49	812	731	2010
Open								

Appendix F. Table of raw data for microplastic samples amount and results for each transect. N/A indicates samples that were not present

Transect	Grams of Sediment Processed Top Later	Grams of Sediment Processed Middle Later	Grams of Sediment Processed Bottom Later	Total Grams Processed Each Transect	Sediment Plastics (MP/ Transect)	Porewater Plastics (MP/ Transect)	Porewater Plastics (MPs/L)	Sediment Plastics (MPs/kg)	Sediment Plastics (MPs/ 100g)	Course Root Plastics (MPs/ 10g)	Fine Root Plastics (MPs/10 g)
1-FMS-	175	0	0	175	0	3	25	0	0.00	714.29	0.00
2-CCG- Open	136	150	63	349	4	2	8.33	13.11	1.15	N/A	N/A
2-CCG-	150	155	0	305	9	1	16.67	25.79	2.95	378.79	7.435
Sea 3-NPT- Open	97	163	90	350	8	1	8.33	23.12	2.29	N/A	N/A
3-NPT-	172	136	130	438	7	4	33.33	20	1.60	757.58	71.942
Land 3-NPT- Sea	112	128	106	346	6	0	0	13.7	1.73	114.29	0.00
4-MSK-	100	36	42	178	1	4	16.67	5.62	0.56	N/A	N/A
Open 4-MSK- Sea	176	50	0	226	4	1	8.33	17.7	1.77	0.00	130.78
5-ALL- Sea	85	64	0	149	7	0	0	26.82	4.70	273.72	0.00

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G.	Szafranski	et	al.	
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(continued)

Transect	Grams of Sediment Processed Top Later	Grams of Sediment Processed Middle Later	Grams of Sediment Processed Bottom Later	Total Grams Processed Each Transect	Sediment Plastics (MP/ Transect)	Porewater Plastics (MP/ Transect)	Porewater Plastics (MPs/L)	Sediment Plastics (MPs/kg)	Sediment Plastics (MPs/ 100g)	Course Root Plastics (MPs/ 10g)	Fine Root Plastics (MPs/10 g)
5-ALL- Open	128	40	0	168	2	1	8.33	13.42	1.19	N/A	N/A
5-ALL- Sea2	139	122	0	261	0	2	16.67	0	0.00	N/A	N/A
6-MI9- Open	165	172	50	387	1	0	0	2.48	0.26	N/A	N/A
6-MI9- Land	115	106	25	246	0	17	141.67	0	0.00	146.20	148.70
6-MI9- Sea	168	180	56	404	1	0	0	2.58	0.25	0.00	219.30
7-OLW- Open	175	125	50	350	3	2	16.67	8.57	0.86	N/A	N/A
8-KWU- Sea	135	141	20	296	1	2	16.67	2.98	0.34	N/A	N/A
8-KWU- Sea2	137	152	103	392	1	0	0	1.92	0.26	314.47	0.00
8-KWU- Open	168	167	186	521	2	3	25	6.76	0.38	N/A	N/A
8-KWU- Open2	136	148	52	336	5	0	0	12.76	1.49	N/A	N/A
9-WCR- Open	163	157	60	380	1	0	0	2.63	0.26	N/A	N/A
9-WCR- Land	174	152	71	397	4	2	16.67	10.34	1.01	N/A	N/A
9-WCR- Sea	129	155	103	387	2	0	0	5.04	0.52	N/A	N/A
10-PLA- Sea	97	135	85	317	1	0	0	3.15	0.32	325.60	652.17
11-NIP- Sea	160	151	71	382	0	3	25	0	0.00	0.00	241.55
12-KPK- Open	132	136	83	351	0	2	16.67	0	0.00	N/A	N/A
12-KPK- Sea	121	71	0	192	0	4	33.33	0	0.00	0	0
13-LEN- Open	155	160	60	375	4	1	8.33	10.67	1.07	N/A	N/A
Average	140.74	124.15	55.78	320.67	2.74	2.04	16.36	8.49	0.93	252.08	122.66

Appendix G. Table of raw data for porewater pesticide analysis

Cito Troppost	Madium	Mass (a) day	Difortheir (no (Imidaalaarid (no /	Down otherin (no (' DDT	Triffunction (no.)
Plot	Medium	weight	L)	L)	L)	p,p -DDE (ng/L)	p,p -DD1 (ng/L)	L)
NPT-Open2	Pore Water	0.0322						
	(Dissolved)							
NPT-Sea-2	Pore Water	0.0391						
	(Dissolved)							
FMS-Sea-2	Pore Water	0.0281						
	(Dissolved)							
KPK-Open-2	Pore Water	0.0423		30.4				
	(Dissolved)							
ALL-Sea-2	Pore Water	0.0199						
	(Dissolved)							
KWU-Sea-2	Pore Water	0.0427		43.7				
	(Dissolved)							
OLW-Open-2	Pore Water	0.0317						
	(Dissolved)							
WCR-Sea-1	Pore Water	0.074						
	(Dissolved)							
MSK-Sea-1	Pore Water	0.051						
	(Dissolved)							
MI9-Land-2	Pore Water	0.08						
	(Dissolved)							
MI9-Sea-2	Pore Water	0.072						
	(Dissolved)							

Appendix H. Table of raw data for plant tissue pesticide analysis

Site, Transect,	Plant Tissue	Mass (g) dry	Bifenthrin (ng/	Imidacloprid (ng/	Permethrin (ng/	p,p'-DDE (ng/	p,p'-DDT (ng/	Trifluralin (ng/
FIOL		weight	g)	g)	g)	g)	g)	g)
FMS-Sea-2	Roots	0.2154	298.05					
NPT-Sea-2	Roots	0.2131	94.79			2.82		
MSK-Sea-2	Roots	0.19	77.89					
KWU-Sea-2	Roots	0.0515	34.95					
ALL-Sea-2	Roots	0.2136	870.79					
MI9-Sea-2	Roots	0.0882	97.51					
KPK-Sea-2	Roots	0.2148	323.09					
CCG-Sea-2	Roots	0.2065	99.76					
NIP-Sea-3	Roots	0.1493	125.92					
NPT-Land-2	Roots	0.2067	69.67			20.3		
PLA-Sea-2	Roots	0.2161	583.99					
NPT-Sea-2	Leaves	0.2096						
FMS-Sea-2	Leaves	0.2115						
ALL-Sea-2	Leaves	0.2121						
Kwu-Sea-2	Leaves	0.2009						
NPT-Sea-2	Propagules	0.2013						
FMS-Sea-2	Propagules	0.2047						
ALL-Sea-2	Propagules	0.2071						
KWU-Sea-2	Propagules	0.2094	8.6					
KWU-Sea-1	Propagules	0.212						

Appendix I. Table of raw data for sediment pesticide analysis

Site, Transect, Plot	Core Segment	Volume (L)	Bifenthrin (ng/g)	Imidacloprid (ng/g)	Permethrin (ng/g)	p,p'-DDE (ng/ g)	p,p'-DDT (ng/ g)	Trifluralin (ng/g)
ALL-Open-2	Тор	5.0322	3.54					
ALL-Open-1	Тор	5.0395	20.28					
ALL-Sea-2	Тор	5.08	18.94					
ALL-Sea-2	Middle	5.0234	25.2					
ALL-Sea-1	Тор	4.985	29.13					
ALL-Sea-3	Тор	5.0052	4.68					
CCG-Open-2	Тор	5.0604	1.74					
CCG-Open-2	Middle	5.0578	0.16		0.24			
CCG-Sea-2	Тор	5.0466	4.04					
CCG-Sea-2	Middle	4.9608	3.14					
CCG-Sea-2	Bottom	4.9522	0.48					
FMS-Sea-1	Тор	4.9931	63.37					
KPK-Open-2	Тор	5.0064	39.23					0.2
KPK-Open-2	Middle	5.0938	0.27					0.39
KPK-Sea-2	Тор	5.0423	1.82					
KPK-Sea-1	Тор	5.0407	0.67					
KWU-Sea-2	Тор	5.0057	1					0.4
KWU-Sea-2	Middle	5.0117	0.16					
MI9-Land-2	Тор	4.9534	0.32					
MI9-Land-2	Middle	5.1487	0.16					
MI9-Land-2	Bottom	5.0495	0					
MI9-Sea-2	Тор	5.2787	0					
MI9-Sea-2	Middle	5.2002	0.69					
NPT-Open-2	Тор	5.0634	1.5			0.32		
NPT-Open-2	Middle	5.03	0.76			0.38		
NPT-Open-2	Bottom	5.0445	0.71			0.95	0.32	
NPT-Sea-2	Тор	5.02	6.14					
NPT-Sea-2	Middle	5.0015	4.32			0.2		
NPT-Sea-2	Bottom	5.0354	1.03			1.03		
OLW-Open-2	Тор	5.0762	16.47					
OLW-Open-2	Middle	4.998	9.68					
OLW-Open-2	Bottom	5.0494	22.34					
PLA-Sea-2	Тор	4.9345	94.96					
PLA-Sea-2	Middle	5.0388	10.99					
PLA-Sea-2	Bottom	4.9152	0.49					
WCR-Sea-2	Тор	4.911	0.77					
WCR-Sea-2	Middle	5.0924	1.85					

Statistic	Log % Impervious Surface within 1 ${\rm km}^2$
N	27
R ²	0.33
Р	0.000959
F	14,25

Appendix J. Relationship between impervious surface and sediment MPs

Appendix K. Schematic demonstrating how cores were divided prior to analysis

		4 Inches		
Тор	МР	МР	МР	Bottom
	РОР	РОР	POP	

Appendix L. Example microfiber at 4x magnification. Sample ID KWU-Sea 2 Porewater 2

Appendix M. Concentrations of Bifenthrin in mangrove roots (ng/g)

Appendix N. Root MP Abundance Standardized to 10 grams

Transect Name	Fishpond or	Fine Roots				Coarse Roots				Transect Total	
	open couci			Weiaht	MP Abundance			Weiaht	Abundance	Transcott Fotal	
		Total Site	MP	Standardized to	per Transect	Total Site		Standardized	per Transect		Transect Total
		Weight (g)	Abundance	10 grams	(10 g)	Weight (g)	MP Abundance	to 10 grams	(10 g)	MP Abundance	Abundance (10 g)
1-FMS-Sea	Open Coast	0.31	0.00	32.26	0.00	0.28	2.00	35.71	71.43	2.00	71.43
2-CCG-Sea	Open Coast	1.35	1.00	7.43	7.43	1.32	5.00	7.58	37.88	6.00	45.31
3-NPT-Land	Open Coast	0.42	3.00	23.98	71.94	0.40	3.00	25.25	75.76	6.00	147.70
3-NPT-Sea	Open Coast	1.70	0.00	5.88	0.00	1.75	2.00	5.71	11.43	2.00	11.43
4-MSK-Sea	Fishpond	2.29	3.00	4.36	13.08	0.73	0.00	13.66	0.00	3.00	13.08
5-ALL-Sea	Fishpond	4.84	0.00	2.07	0.00	1.10	3.00	9.12	27.37	3.00	31.45
6-M9-Land	Open Coast	1.35	2.00	7.43	14.87	0.41	1.00	24.15	24.15	3.00	29.49
6-M9-Sea	Open Coast	0.46	1.00	21.93	21.93	0.10	0.00	103.09	0.00	1.00	21.93
7-KWU-CVT	Fishpond	1.35	0.00	7.43	0.00	0.94	1.00	10.67	10.67	1.00	31.45
8-PLA-Sea	Fishpond	0.46	3.00	21.74	65.22	1.23	4.00	8.14	32.56	7.00	97.78
9-NIP-Sea	Open Coast	0.41	1.00	24.15	24.15	0.58	0.00	17.36	0.00	1.00	24.15
10-KPK-Sea	Fishpond	1.36	0.00	7.37	0.00	0.94	0.00	10.67	0.00	0.00	0.00

Appendix O. Average weights roots

Sample Site	Fine Roots	X - no sample	O - missing weight	Coarse Roots	X - no sample	O - missing weight
	Initial Weight (g)	Averaged Missing Weights (g)	Total Weight per Site (g)	Initial Weight (g)	Averaged Missing Weights (g)	Total Weight per Site(g)
1-FMS-Sea#1	0.26	0.26	0.31	0.16	0.16	0.28
1-FMS-Sea#2	0.05	0.05		Х	Х	
1-FMS-Sea#3	Х	Х		0.09	0.12	
2-CCG-Sea#1	0	1.345	1.345	0	0.66	1.32
2-CCG-Sea#2	Х	Х		Х	X	
2-CCG-Sea#3	Х	Х		0.66	0.66	
3-NPT-Land#1	0	0.139	0.417	0	0.132	0.396
3-NPT-Land#2	0	0.139		0	0.132	
3-NPT-Land#3	0.139	0.139		0.132	0.132	
3-NPT-Sea#1	0.56	0.56	1.7	0.16	0.16	1.75
3-NPT-Sea#2	0.98	0.98		0.08	0.08	
3-NPT-Sea#3	0.16	0.16		1.51	1.51	
4-MSK-Sea#1	0.241	0.241	2.294	0.211	0.211	0.732
4-MSK-Sea#2	0.978	0.978		0.416	0.416	
4-MSK-Sea#3	1.074	1.074		0.105	0.105	
5-ALL-Sea#1a	0.312	0.312	4.836	0.18	0.18	1.096
5-ALL-Sea#2a	0.624	0.624		0.173	0.173	
5-ALL-Sea#3a	0.414	0.414		0.127	0.127	
5-ALL-Sea#1b	1.45	1.45		0.186	0.186	
5-ALL-Sea#2b	0.94	0.94		0.28	0.28	
5-ALL-Sea#3b	1.096	1.096		0.15	0.15	
6-M9-Land#1	Х	Х	1.345	0.02	0.02	0.414
6-M9-Land#2	0	1.345		0	0.138	
6-M9-Land#3	Х	Х		0.256	0.256	
6-M9-Sea#1	0.031	0.031	0.456	0.032	0.032	0.097
6-M9-Sea#2	0.051	0.051		0.034	0.034	
6-M9-Sea#3	0.374	0.374		0.031	0.031	
7-KWU-CVT#1	0	1.345	1.345	0	0.106	0.318
7-KWU-CVT#2	Х	X		0.156	0.156	
7-KWU-CVT#3	Х	X		0.056	0.056	
8-PLA-Sea#1	Х	X	0.46	0	0.4095	1.2285
8-PLA-Sea#2	0	0.23		0.621	0.621	
8-PLA-Sea#3	0.23	0.23		0.198	0.198	
9-NIP-Sea#1	0.138	0.138	0.414	Х	X	0.576
9-NIP-Sea#2	0	0.138		0	0.288	
9-NIP-Sea#3	0.138	0.138		0.288	0.288	
10-KPK-Sea#1	X	X	Х	0.35	0.35	0.938
10-KPK-Sea#2	X	X		0.275	0.275	
10-KPK-Sea#3	Х	X		U	0.313	

Appendix P. Unadjusted totals accounting for all possible MPs found across samples and controls

Transect	Sediment Plastics (MP/Transect)	Porewater Plastics (MP/Transect)	Total MP Fine roots (mp/transect)	Total MP coarse roots (MP/Transect)	On-site airfall from field collection	Laboratory Controls (total)
1-FMS-Open	0	3	0	2	1	Sediment Partitioning Airfall Total MPs (All days) (continued on next page)

(continued)

Transect	Sediment Plastics (MP/Transect)	Porewater Plastics (MP/Transect)	Total MP Fine roots (mp/transect)	Total MP coarse roots (MP/Transect)	On-site airfall from field collection	Laboratory Controls (total)
2-CCG-Open	4	2			1	78
2-CCG-Sea	9	1	1	5	0	Sediment Processing and Airfall
						(All sites)
3-NPT-Open	8	1			0	18
3-NPT-Land	7	4	3	3	5	Porewater Processing and Airfall
						Control Total
3-NPT-Sea	6	0	0	2	2	20
4-MSK-Open	1	4			2	
4-MSK-Sea	4	1	3	0	2	
5-ALL-Sea	7	0	0	3	0	
5-ALL-Open	2	1			0	
5-ALL-Sea2	0	2			2	
6-MI9-Open	1	0			0	
6-MI9-Land	0	17	2	1	0	
6-MI9-Sea	1	0	1	0	1	
7-OLW-Open	3	2			1	
8-KWU-Sea	1	2			1	
8-KWU-Sea2	1	0	0	1	2	
8-KWU-Open	2	3			1	
8-KWU-	5	0			1	
Open2						
9-WCR-Open	1	0			0	
9-WCR-Land	4	2			0	
9-WCR-Sea	2	0			na	
10-PLA-Sea	1	0	3	4	0	
11-NIP-Sea	0	3	1	0	2	
12-KPK-	0	2			2	
Open						
12-KPK-Sea	0	4	0	0	0	
13-LEN-	4	1			16	
Open						
Total	74	55	14	21	42	116
Combined Total	359					

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G. Szafranski et al.

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