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Microplastic pollution in the intertidal and subtidal sediments of Vava'u, Tonga

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ABSTRACT

Plastic pollution research on a global scale intensified considerably in the current decade; however, research efforts in the South Pacific are still lagging. Here, we report on microplastic contamination of intertidal and subtidal sediments in the Vava'u archipelago, Tonga. While providing the first baseline data of its type in Tonga, the study also advances methods and adjusts them for low-budget research. The methods were based on density separation of microplastics from the sediment using $CaCl_2$, a high-density salt which due to its high solubility, low cost and availability. Once separated, microplastics were quantified by microscopic analysis and polymers characterized via FTIR spectroscopy. Microplastics in intertidal and subtidal sediments were found in concentrations of 23.5 ± 1.9 and 15.0 ± 1.9 particles L^{-1} of sediment, respectively. The dominant type of microplastics in both intertidal (85 %) and subtidal sediments (62 %) were fibres.

1. Introduction

Small island developing states (SIDS) depend on the marine environment in many ways and are highly susceptible to human impacts such as climate change and marine pollution (Rawlins et al., 1998; Sareer, 2017). Recognised as an environmental problem decades ago, marine plastic pollution continues to prompt questions in relation to human and environmental health as the annual global production and consumption of plastics steadily increases (PlasticsEurope, 2021). In response, research on the levels and impacts of plastic pollution in the world's oceans has noticeably intensified in the recent years.

Plastic debris and microplastics have been documented in the digestive system of marine organisms, which are regularly consumed by humans in the Pacific, such as fish and shellfish (Van Cauwenberghe and Janssen, 2014; Rochman, 2015; Forrest and Hindell, 2018; Markic et al., 2018; Alfaro-Núñez et al., 2021). Ingested plastic can either have a direct physical impact on the organism, such as blockage and rupture, or cause a physiological response to plastic-related toxicity, such as complex endocrine changes (Kühn et al., 2015; Verla et al., 2019). Marine plastic debris is known to accumulate toxic compounds from the surrounding water onto its surface. Several experimental and field studies demonstrated that, upon ingestion, these toxins can detach from the plastics and accumulate in animal tissues, depending on the equilibrium (e.g. Gassel et al., 2013; Rochman et al., 2014; Tanaka et al., 2020). As a result, great concern has been on the rise about the impacts of plastic pollution on human health (Menéndez-Pedriza and Jaumot, 2020; Davison et al., 2021; Yee et al., 2021).

Being a pervasive global phenomenon, plastic pollution has not bypassed the seemingly pristine South Pacific Islands. Owens et al. (2011) reported that 57 % of all solid waste generated on Kayangel Island, Palau, in fact, originates from the ocean and that, considering solid waste management already is an immense challenge for these small islands, it presents a '*significant burden of marine litter on a SIDS community*' (p. 941). Generally, plastic pollution research in this region, though scarce and fragmented, indicates high contamination levels and the need for more in-depth and systematic investigation. The pioneering

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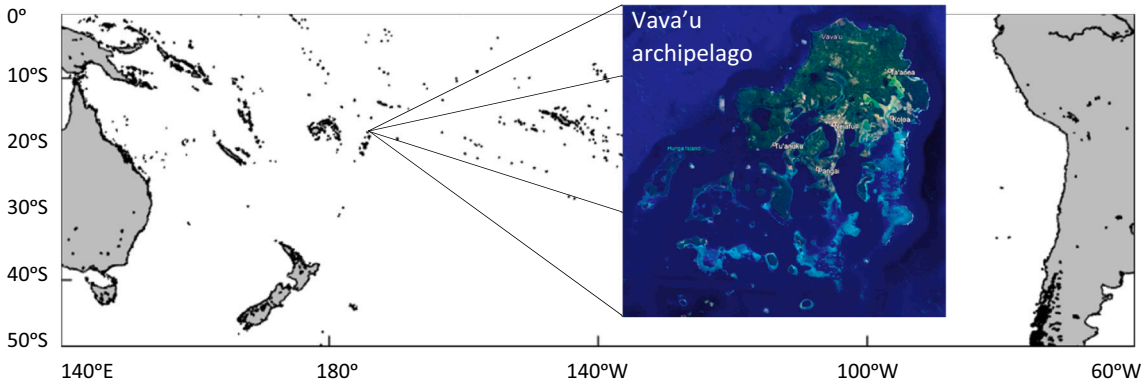
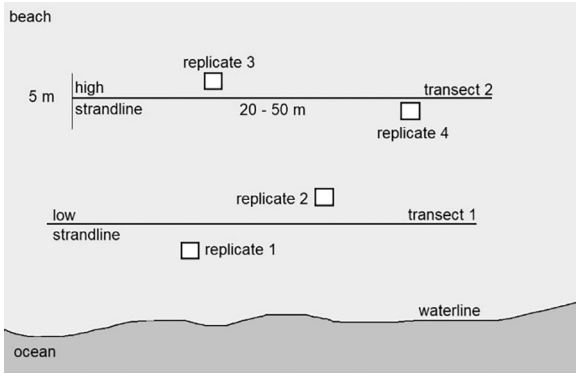
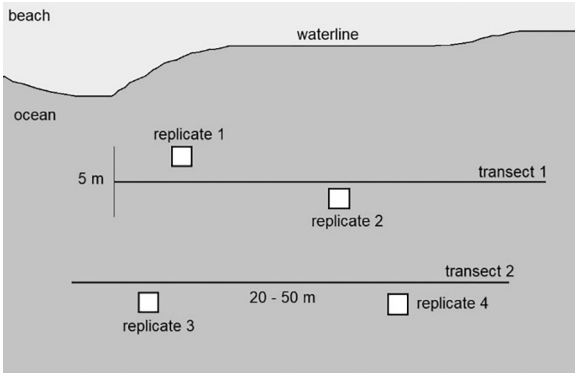


Fig. 1. Study location showing Vava'u archipelago, Tonga, and its position in the South Pacific.

Table 1
Description and scheme of sampling design for intertidal and subtidal samples.

Intertidal samples	Subtidal samples
Two belt transects were placed along the beach, parallel to the waterline. One transect was at the low and the other at the high strandline. The transects were 5-m wide with varying lengths (20–50 m), depending on the terrain. At four random points along the transects (two points at each transect), sediment samples were scooped with a metal collection container to a depth of 4 cm to collect 1 L of sediment.	Two belt transects were placed subtidally randomly in the general direction parallel to the shore, where sediment was visible. The depth of transects was between 3 and 15 m. The transects were 5-m wide with varying lengths (20–50 m), depending on the terrain. At four random points along the transects, sediment samples were scooped with a metal collection container to a depth of 4 cm to collect 1 L of sediment.
	

and most notable work in the South Pacific Island states was carried out by Gregory (1990) (Table 4), who reported high concentrations of plastic pellets on Tongatapu (the main island of the Kingdom of Tonga) with over 1000 plastic pellets m^{-1} of Laulea beach.

In some distant locations, such as Easter Island or Henderson Island, plastic debris accumulates in exceptionally high concentrations, as much as 805 and 672 items m^{-2} of the beach, respectively (Hidalgo-Ruz and Thiel, 2013; Lavers and Bond, 2017). The concentration of micro and nanoplastics in the sediments of the East Beach on Henderson Island collected in 2019 was, on average, 1960 ± 356 pieces kg^{-1} of sediment (Nichols et al., 2021). Eriksen et al. (2013) uncovered the South Pacific 'garbage patch' with floating plastic concentrations reaching 400,000 microplastic particles km^{-2}.

Considering the Pacific Ocean is the largest ocean, research efforts on plastic pollution in the Pacific region have been disproportionally small. The most likely reason for this gap is the lack of researchers and access to suitable facilities. Quantification of microplastics by the most common approach requires specialist laboratory facilities, equipment and chemicals, making it challenging for many SIDS. In this study, we primarily aimed to determine, for the first time, the level of microplastic contamination in the Vava'u archipelago, Tonga, and secondarily, to develop a low-budget method for microplastics research that is suitable for low and middle-income countries. Some of the requirements for low-cost and simple methods suitable for the SIDSs are: easily obtained and cheap equipment and chemicals, reusable equipment, the minimal requirement for lab facilities and easy to perform for untrained volunteers.

2. Methods

2.1. Sample collection

Plastic pollution assessment was carried out in the Vava'u archipelago, Tonga (Fig. 1), from August to October 2017, onboard the 120-foot sailing vessel Infinity, on multiple sites within the archipelago with a research permit from the Ministry of Education and Training of the Kingdom of Tonga. Two types of assessment were done: (i) macroscopic, or in situ visual quantification of debris larger than 1 mm, and (ii) microscopic, quantification of debris over 63 μm in field-collected seawater and sediment samples using a dissecting microscope. In this paper, we will present and discuss only the methodology and results concerning **microplastics** in **intertidal** and **subtidal sediments**. We divided the study into three papers to keep the three different methods (visual macroscopic assessment, surface water assessment and sediment assessment) separate.

Sediment sample collection was done using a nested sampling design with four replicates per sampling site. There were seven intertidal and eight subtidal sites. At each site, two transects were placed parallel to the

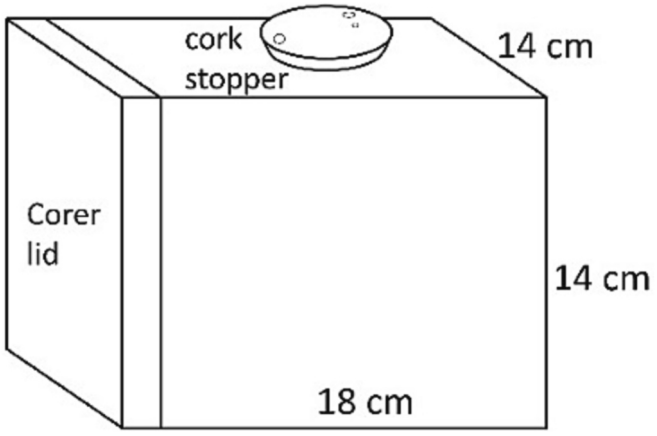


Fig. 2. Metal box corer used for sediment sample collection.

Table 2
Density of common plastics (extracted from Mohanty et al., 2005 and Shackelford et al., 2016).

Polymer type	Abbreviation	Density (g cm^{-3})
Polypropylene	PP	0.9–0.91
Polyethylene	PE	0.91–0.96
Polystyrene	PS	1.04–1.07
Polyamide (or nylon)	PA	1.12–1.15
Acrylics	ACR	1.17–1.20
Thermoplastic polyester	PES, PET	1.31–1.39
Polyvinyl chloride	PVC	1.20–1.44
Rayon (or viscose)	RAY	1.52

waterline, explained in more detail in Table 1.

Metal collection containers, used for sediment collection, were made from modified biscuit containers (Fig. 2). The size of the containers was 18 × 14 × 14 cm. On one side of the container, a hole was drilled and a large cork stopper was placed in it. This was done to ensure that the underwater samples can be collected with minimal disturbance. The sediment from both compartments (intertidal and subtidal) was collected by scooping the surface 4 cm of the sediment. Underwater sediment was scooped very slowly to keep the surface of the sediment undisturbed, allowing the water to pass through the hole on the top as the sediment was filling the container. Once the 4 cm of sediment were in the container, the container was closed by placing the lid and the cork stopper firmly in position. Sediment samples, both intertidal and subtidal, were rinsed out of the collection container with a squeeze bottle filled with filtered seawater into a rinsed zip-lock bag for further laboratory analysis. Most commonly, mass (kg) is used as a measure for the amount of sampled sediment, but we chose volume (L) to avoid weighing in the field and to give equal representation to each sample space-wise, regardless of the sediment type and particle size (for example, 1 kg of fine sediment is smaller in volume than 1 kg of coarser sediment).

2.2. Sample processing

Sediment samples were analysed through the following three steps: density separation, vacuum filtration and microscopic analysis. To separate the plastics from the sediment, we used a high-density solution ($\rho = 1.40–1.45$ g cm^{-3}) of anhydrous calcium chloride ($CaCl_2$) (supplier: Shouguang Hengyi Chemical Technology Co., a free sample). Density separation of microplastics from the sediment is a common method; however, the most commonly used high-density solution in the microplastics studies has been a hypersaline (NaCl) solution of 1.2 g cm^{-3} density (see reviews: Bellasi et al., 2021; Cutroneo et al., 2021; Phuong et al., 2021; Tirkey and Upadhyay, 2021), which theoretically excludes some of the common denser plastics, such as PVC and PET (Table 2). To avoid the potential underestimate of denser plastics, we used a $CaCl_2$ solution. Other high-density solutions such as NaI were cost-prohibitive with added health and safety risks.

2.2.1. Step 1: density separation

Between 1000 and 1300 g of $CaCl_2$ salt was dissolved in 1 L of tap water at ambient temperatures ranging from 25 to 35 °C. This equals to 53.3–56.6 % $CaCl_2$, which according to OxyChem (2012) guide, gives densities over 1.435 g cm^{-3}. We also measured the density of the solution using the weighing method, and 1 L of the $CaCl_2$ solution in the measuring cylinder weighed between 1400 and 1450 g, giving the density between 1.40 and 1.45 g cm^{-3}. Prior to density separation, $CaCl_2$ solution was filtered on a 5-μm stainless steel filter. Before processing the samples, since there was no adequate space and equipment to dry the sediments, excess water was left to slowly drain through tiny holes made on the plastic bag of each replicate sample to prevent dilution of $CaCl_2$ solution. The sediment was put into a ceramic bowl with 1 L of $CaCl_2$ solution. It was then stirred with a metal whisk for 20–30 s to allow the plastic particles to surface, covered with aluminium foil to avoid airborne contamination, and left to settle for 15–45 min until the solution was clear. Settling time depended on the sediment grain size, with longer settlement time needed for finer sediments.

2.2.2. Step 2: vacuum filtration

After settling, the supernatant was decanted into a large glass beaker and the density separation process was repeated twice for each sample (three times altogether). The glass beaker was also covered with aluminium foil. The decanted solution was then filtered on a stainless-steel filter (mesh size 63 μm), which was visually analysed under a dissecting microscope. It should be noted that the mesh size of the filter determined our particle lower size limit due to potential loss of particles smaller than 63 μm. During decantation and vacuum filtration, the inner sides of the bowls and beakers were continuously rinsed with a filtered $CaCl_2$ solution in a squeeze bottle to ensure all plastic particles were collected. The remaining $CaCl_2$ solution was re-filtered over a 5-μm filter and reused for other samples. Its density was checked by weighing the solution in a glass measuring cylinder and adjusted if needed.

2.2.3. Step 3: microscopic analysis

Microscopic analysis of the filters was done by visual identification, where all particles resembling synthetic materials were isolated, separated into categories by morphology (fibre, fragment, film), colour (all colours) and size (<100 μm, 100–200 μm, 200–300 μm, 300–400 μm, 400–500 μm, 500–1000 μm, 1–2 mm, 2–5 mm, >5 mm), counted and stored into 2-mL glass vials for further analysis (i.e. polymer characterisation).

2.3. Method testing and contamination control

We tested the density separation method in two separate trials, using 10 blue polyethylene beads extracted from Neutrogena face wash (100–500 μm). In both trials, 100 % of the plastic beads were recovered.

We designed the method to have as few steps as possible to minimise contamination. To prevent airborne contamination, all samples were covered with aluminium foil when not working with them. All instruments were rinsed with clean water before use and all liquids used in the analysis were filtered (5-μm filter). We also used blank tests every few samples and kept them uncovered only while processing a sample, which was very short. We did not find microplastic contamination on them.

Other potential sources of contamination were metal corers, squeeze bottles and plastic bags. The outside of the metal corers was painted in light yellow colour and any paint particles of that colour were excluded. Squeeze bottles were regularly rinsed and particles suspected to originate from the squeeze bottles were excluded. Plastic bags used for

storing sediment samples were rinsed thoroughly before use in the surrounding seawater. Since the analysis of the surface waters of Vava'u showed that the microplastics concentration was generally very low ($10^{-3} \pm 10^{-4}$ pcs L^{-1} of water) (Markic et al., 2022), we were confident that the bags would not get contaminated from the seawater. Additionally, all fresh transparent film resembling the sample bags were also excluded.

2.4. Polymer characterisation – chemical analysis of retrieved plastics

To confirm identity of counted microplastics, Fourier transform infrared (FTIR) spectroscopy was performed for polymer identification. Since FTIR spectroscopy is an expensive procedure due to high personnel and time cost, the analyses were done in collaboration with a research partner Scion (New Zealand). FTIR spectra were obtained for a random subset of sediment microplastics (70 particles). Before analysis, all samples were dried at 70 °C for 4 h. Larger microplastics (>300 μm) were analysed using a Bruker Tensor 27 Instrument with a diamond attenuated total reflectance (ATR) cell acquiring 32 background and sample scans from 725 to 4000 cm^{-1} at 4 cm^{-1} resolution. Smaller microplastics (<300 μm) were analysed using a Bruker Tensor 27 Instrument connected to a Bruker IRScope II equipped with a mercury cadmium telluride (MCT) detector. Samples were placed in a diamond compression cell and analysed in transmission with 32 background and sample scans from 725 to 4000 cm^{-1} at 4 cm^{-1} resolution. All spectra were baseline-corrected using Bruker OPUS 7.2 software.

Following a workflow adapted from Kroon et al. (2018), spectra were searched against a selection of Bio-Rad FTIR spectral databases using an Euclidean distance algorithm with Bio-Rad KnowItAll® software. The databases included the following polymer types and naturally occurring materials: polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyester (PES); nylon (PA), ethylene vinyl acetate (EVA), polyurethane (PUR), styrene acrylonitrile copolymer (SAN), rubber (RUB), rayon (RAY), acrylics (ACRY), organosilicone (OS), chitin, keratin, quartz, calcium carbonate, calcium phosphate, hydroxyapatite, and magnesium silicate. A percent match between the sample spectra and database reference spectra were obtained to establish the material type. A match of ≥70 % was classified as positive identification, 60–70 % required user interpretation and <60 % was classified as inconclusive.

2.5. Data analysis

The data were presented in several different measurement units to provide information comparable to a wider range of studies, and they included the number of particles per area and per volume. Pieces (pcs) and particles are used interchangeably, as they indicate the count or the number of micro-debris items. The variability of data was presented with standard errors (SE), and to express the measure of central tendency we used arithmetic mean ($\bar{x}$). The most common measurement units are used in the main text, graphs, images and tables, while the less common units are provided in the Results table and Discussion where they are used for comparison with other studies. To express the precision of estimated plastics concentrations, we provided confidence intervals for 95 % confidence level following the formula provided by Milton (1999):

$$CI = \bar{x} \pm z \frac{SD}{\sqrt{N}}$$

where CI is confidence interval, $\bar{x}$ is arithmetic mean, z is the standard z-score extracted from the z-table for standard normal probabilities (for 95 % confidence level z = 1.96), SD is standard deviation and N is the sample size (i.e. the number of measurements or data points).

The concentrations of microplastics were presented on a bubble map made in an online mapping software Maply, that is partially free of charge (previous Geolytics, currently https://maply.com/).

Table 3
Microplastics concentrations in intertidal and subtidal sediments of Vava'u (SE – standard error, pcs – particles, *LL* – lower limit, *UL* – upper limit).

	Intertidal sediments	Subtidal sediments
Number of samples (cores)	28	32
Sample area (m^2)	0.025	0.025
Sample volume (L = dm^3)	1	1
Total number of particles before FTIR	739	480
Total number of particles after FTIR	659[a]	480
Average concentration (pcs $m^{-2} \pm$ SE)	**942 ± 78**	**600 ± 75**
Minimum concentration (pcs m^{-2})	615	350
Maximum concentration (pcs m^{-2})	1142	850
95 % confidence intervals (*LL*, *UL*)	779, 1095	453, 747
Average concentration (pcs $L^{-1} \pm$ SE)	**23.5 ± 1.9**	**15.0 ± 1.9**
Minimum concentration (pcs L^{-1})	15.4	8.8
Maximum concentration (pcs L^{-1})	28.5	21.3
95 % confidence intervals (*LL*, *UL*)	19.7, 27.3	11.3, 18.7

Bold values are the average microplastics concentration in two different units.
[a] Value adjusted for 10.8 % misidentification error.

3. Results

3.1. Quantitative assessment of microplastics in the sediments

The quantification of plastic debris in sediments was done on 28 cores from seven intertidal sites and 32 cores from eight subtidal sites. Microplastics were found in all cores. In total, there were 739 and 480 plastic particles extracted from intertidal and subtidal sediments, respectively; however, the concentration of microplastics in the intertidal sediments was corrected for the identification error of 10.8 % confirmed by the FTIR analysis. This resulted in an average concentration of 23.5 ± 1.9 for intertidal sediments and 15.0 ± 1.9 pcs L^{-1} for subtidal sediments (Table 3).

The greatest concentrations of microplastics were found in the intertidal sediments of Ovaka Island, with an average of 28.5 pcs L^{-1} (Fig. 3a). The most contaminated subtidal sediments were at the Mystic Sands Resort (Utungake), with an average of 21.3 pcs L^{-1} (Fig. 3b).

3.2. Qualitative assessment of microplastics in the sediments

3.2.1. Size distribution

The size of the microplastics from intertidal and subtidal sediments was measured for 221 and 195 particles, respectively. Intertidal and subtidal microplastics were first pooled across all locations, after which approximately one quarter and one half of each pooled sample, respectively, was measured. The majority of microplastics in the intertidal sediments was larger than 1 mm, with only 3 % being smaller than 300 μm (Fig. 4). Subtidal microplastics were mainly smaller than 1 mm (60 %), but with only 6 % of particles smaller than 300 μm.

3.2.2. Form

The dominant type of microplastics in both intertidal and subtidal samples of Vava'u were fibres (>60 %) (Figs. 5, 6). Intertidal microplastics are composed of as much as 85 % fibres.

3.2.3. Colour composition

The most common colours in both types of sediments were blue, black and colourless (Fig. 7a). The majority of microplastics were blue and black fibres (Fig. 7b).

3.2.4. Polymer characterisation

FTIR analysis was performed on a random selection of 37 particles from the intertidal sediments and 33 particles from the subtidal sediments, 5 % and 7 % of the total count, respectively. Out of the 37 intertidal particles, five had poor quality spectra (HQI (hit quality

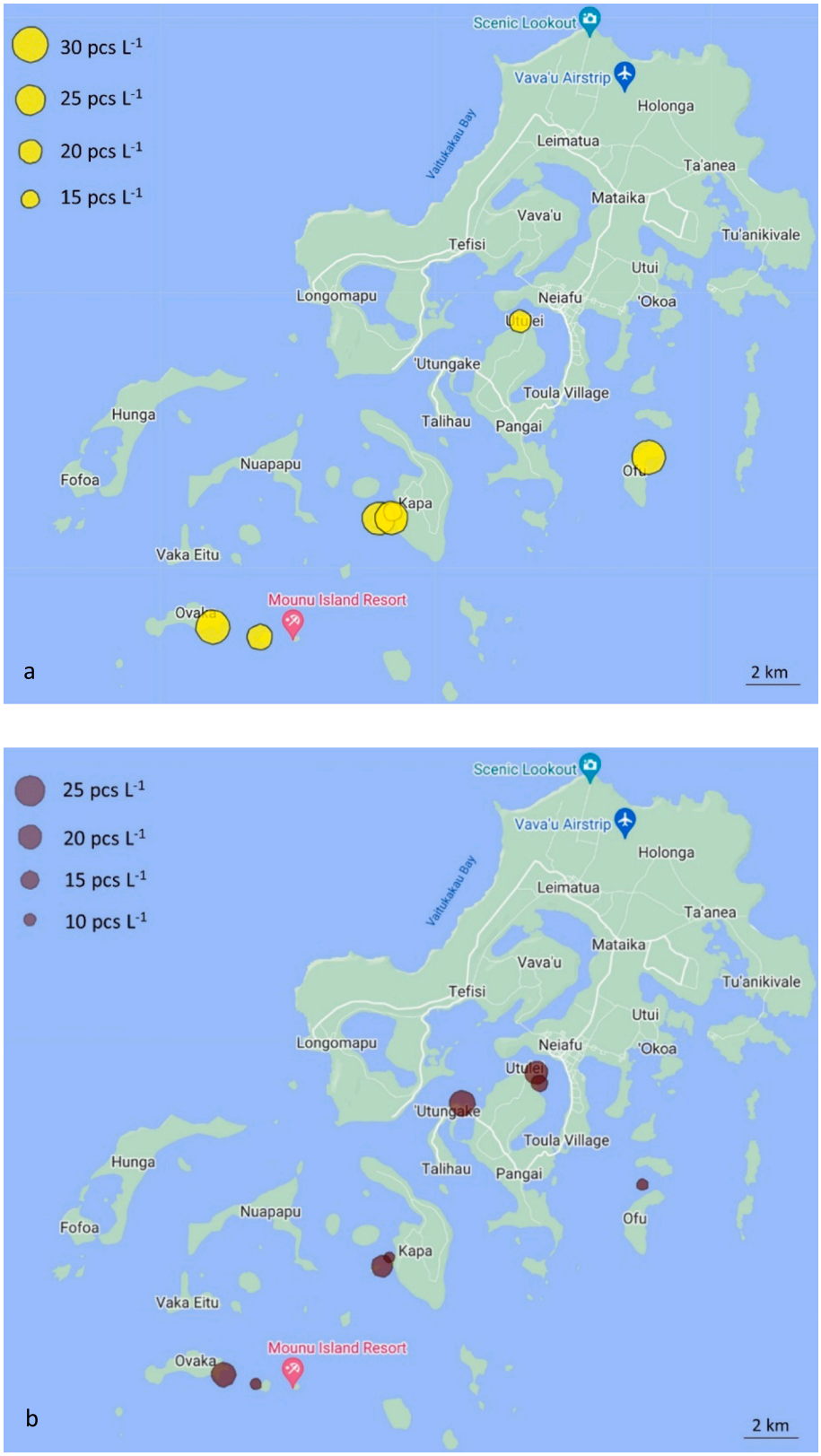


Fig. 3. Concentrations of microplastics in the a) intertidal sediments and b) subtidal sediments of Vava'u archipelago (pcs – pieces/particles of microplastics).

index) < 60 %), four were misidentified ($CaCO_3$) and two were inconclusive. Out of 33 subtidal particles, one had HQI < 60 % and another one was inconclusive. Due to the misidentification of some particles from the intertidal sediments, the overall microplastics quantity was reduced by 10.8 %. Other unsuccessfully analysed particles were believed to be plastic but too degraded for the FTIR analysis. The remaining 57 particles were identified as PE, PP, PA, PES and RAY (Fig. 8). Surprisingly, as much as 42 % of analysed subtidal microplastics is polyethylene (PE), mainly in the form of fragments. Positively and negatively buoyant plastics with respect to fresh and sea water ($\rho \approx 1$ g

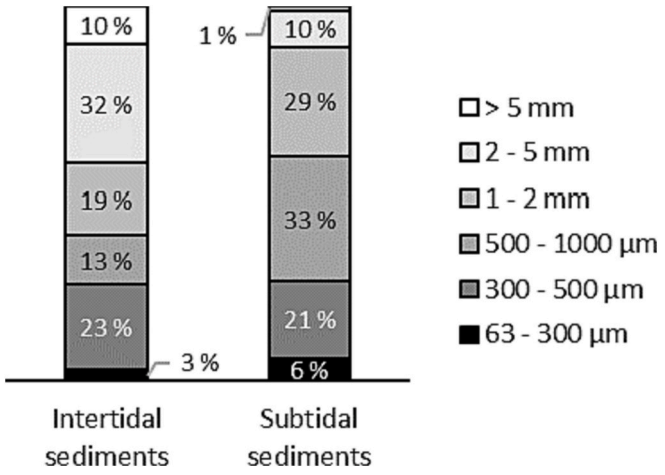


Fig. 4. Size distribution of microplastics in intertidal and subtidal sediments.

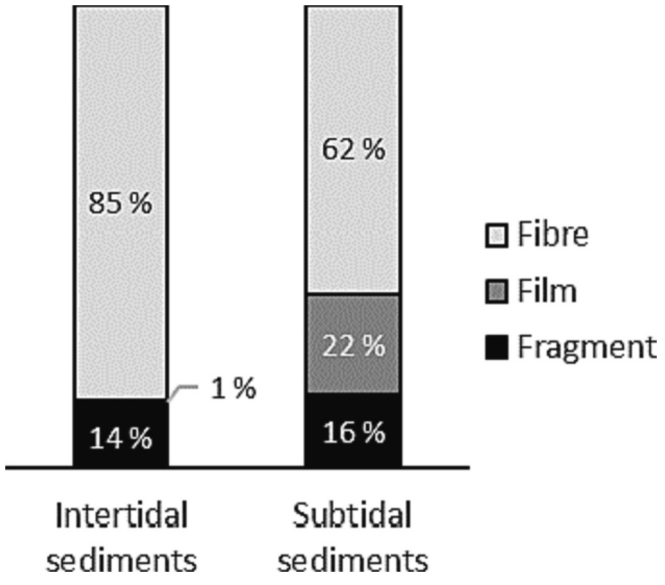


Fig. 5. Composition of microplastics by form in intertidal and subtidal sediments.

cm^{-3}) were found in similar proportions in both compartments (Fig. 9a). Regarding the polymers denser than 1.2 g cm^{-3}, out of the 26 microplastic particles analysed from the intertidal sediments, only three were rayon fibres, while out of the 31 analysed particles from the subtidal sediments, six were rayon fibres, four were polyester fibres and one polyester fragment (Fig. 9b).

4. Discussion

4.1. Quantitative comparison to other regional studies

We found the concentration of microplastics in the intertidal and subtidal sediments of Vava'u to be 23.5 ± 1.9 and 15.0 ± 1.9 pcs L^{-1} of sediment, respectively. Our study is not entirely comparable to any of the previous studies of sediments in the South Pacific mainly because of different measurement units but also due to different methodology (Table 4). Most studies provided the concentration as the number of particles per sediment mass, while we specifically decided on reporting in volumes of sediment to have consistency in the space taken by the sediment, regardless of its gain size. The studies that provided concentration in particle per volume units, either used a different high-density solution for separation or used a different lower-size limit of the filters. However, the concentrations of microplastics in beach sediments in several studies most comparable to ours were lower than concentrations obtained in Vava'u. In two recent studies conducted in New Zealand, microplastic concentration in beach sediments was, on average, 9.1 particles L^{-1} in Auckland region (Bridson et al., 2020) and 4.6 particles L^{-1} in the Northland region (de Lena et al., 2021). More than a decade before our study, Browne et al. (2011) carried out a worldwide assessment of fibres in shore sediments of ten countries, including the Pacific coast of Australia and Chile. The concentration in the Australian beach sediments was eight fibres L^{-1}. Concentration in the Chilean sediments was not given in detail; however, their figure suggests it was greater than in Australian sediments, but <40 fibres L^{-1}, similar to our results.

In recent years, several studies in the South Pacific region looked at microplastics in the subtidal sediments, only one of which provided results as particles per volume (Ling et al., 2017). Ling et al. reported an average microplastics concentration in coastal sediments of South Australia and Tasmania to be 3400 pcs L^{-1} (or 3,4 pcs mL^{-1}), two orders of magnitude greater than in Vava'u subtidal sediments (av. 15 pcs L^{-1}). Possibly, the reason for this could be a much higher population and urbanisation in Australia than in Vava'u. There is also a difference in methodology, where Ling et al. used a NaI solution of density between 1.6 and 1.8 g cm^{-3} for density separation of microplastics from the sediments, but it is unlikely that this would result in such a great difference in concentration.

In another study on microplastic contamination in the subtidal sediments of the northern part of the New Zealand South Island (Ribó et al., 2021), the concentration was an order of magnitude higher than in Vava'u. The concentration was not directly provided in the paper. The number of microplastics along the entire length of the core that takes up a volume of 264 cm^3 ranged from 91 to 120 particles, which recalculated gives the concentration of 344.7 to 454.5 pcs L^{-1}.

4.2. Microplastics size, morphology and colour

4.2.1. Size and morphology

Sizes of retrieved microplastics largely depend on the mesh size of the sieves and filters used in the lab analysis, which makes it difficult to compare the findings among studies. However, the form and the colour mainly do not depend on the methods used. The dominant form of microplastics contained in the intertidal and subtidal sediments of Vava'u are fibres (Figs. 5 and 6), which was also the case with several other studies from the region, in intertidal sediments (Bridson et al., 2020; de Lena et al., 2021) as well as subtidal sediments (Ling et al., 2017; Willis et al., 2017; Ferreira et al., 2020). The discharge from washing machines, entering the oceans via sewage, is believed to be the main source of marine fibre pollution (Habib et al., 1998; Thompson et al., 2004; Browne et al., 2011). Some vessels, such as cruise ships, superyachts, research vessels, and similar, that have onboard washing machines, could be an important source of fibres too (pers. obs. T. Maes). Ribó et al. (2021) found more fibres near Picton and more fragments in the marine reserve area, which is most likely owing to the marine reserve being further from the source of microfibres. Barrett et al. (2020) reported only 10 % of microplastics being fibres. Clunies-Ross et al. (2016) excluded synthetic fibres from their measurements to avoid false positives from airborne contamination. As a result, they mainly recovered fragments (86.3 %) and pellets (11 %) from the shore sediments in New Zealand.

4.2.2. Colour

With respect to colour, not many studies from Table 4 provided colour composition of microplastics. In the ones that did, white, colourless, black and blue were the dominant colours (Clunies-Ross et al., 2016; Jahan et al., 2019; Bridson et al., 2020; Ribó et al., 2021), while Vava'u sediments were mainly contaminated with black and blue fibres (Fig. 7b).

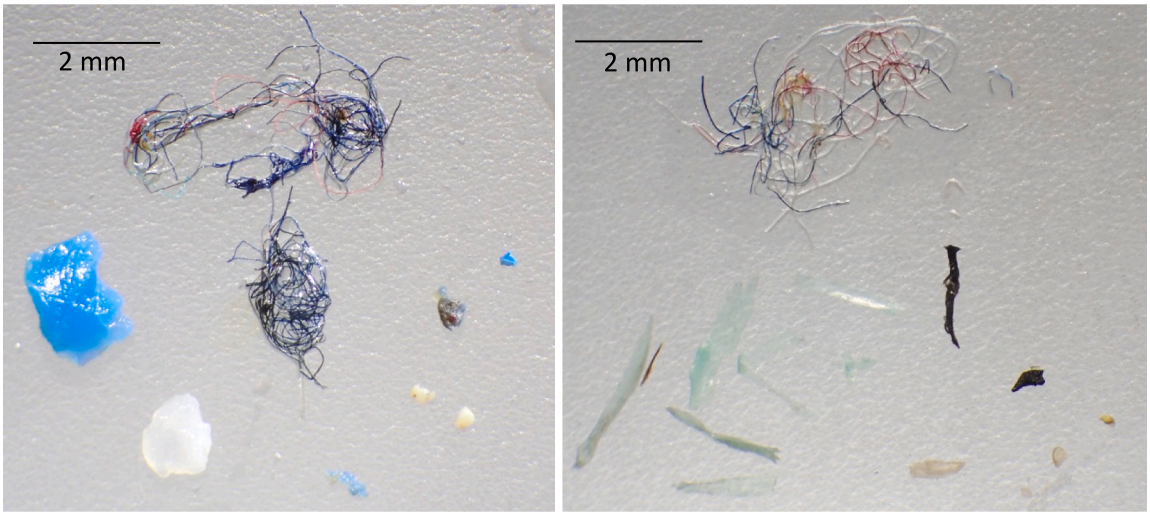


Fig. 6. Fibres as a predominant form of microplastics in the shore (left) and benthic sediments (right). The shore sample was collected from the Nuku high strandline, and the benthic sample was from the seafloor below the Ovaka southern lagoon. Both images show microplastics extracted from 1 L of sediment.

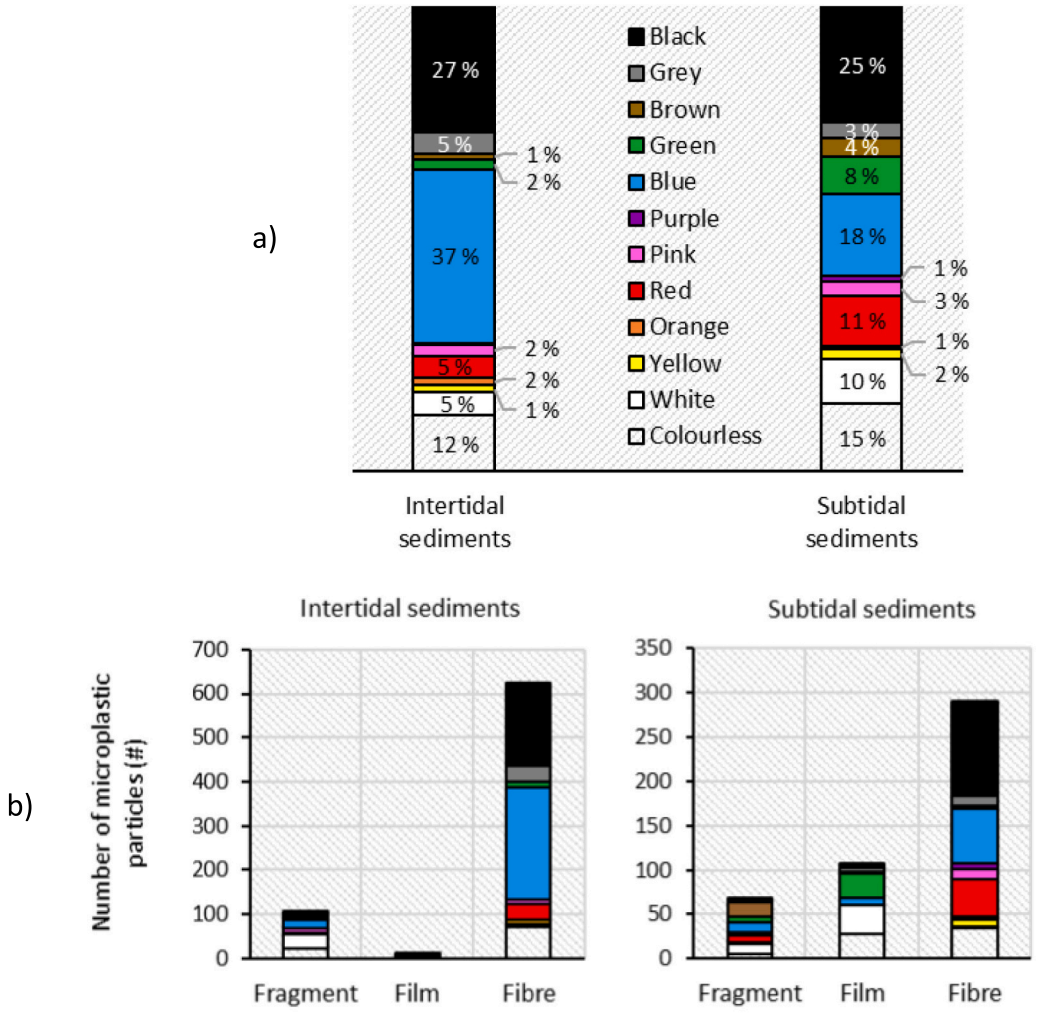


Fig. 7. Colour composition of microplastics in intertidal and subtidal sediments a) overall and b) broken down by plastic type.

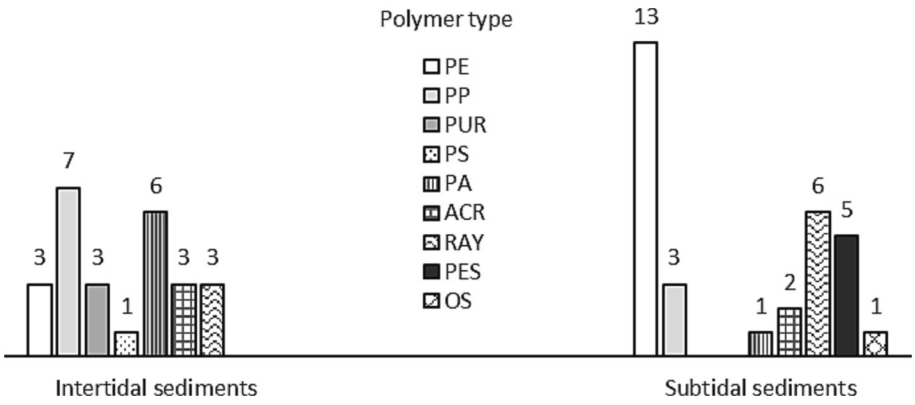


Fig. 8. Analysed sediment microplastics by polymer type. The labels present the number of particles for each polymer.

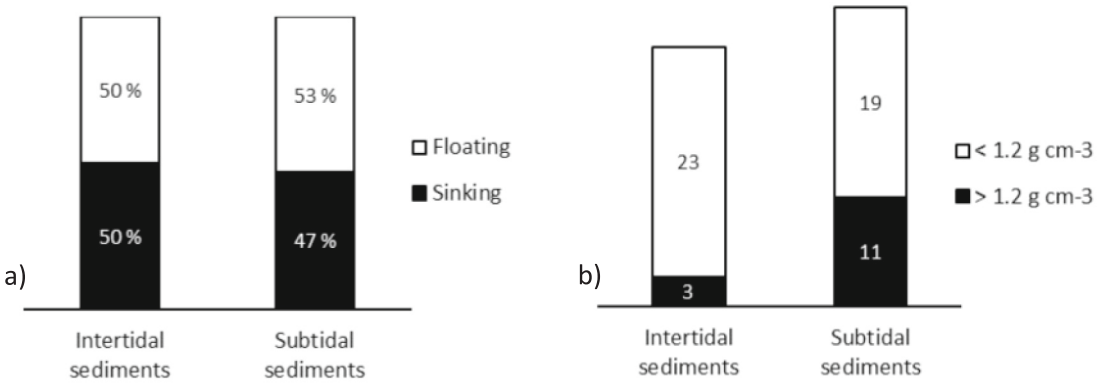


Fig. 9. Proportions of a) positively and negatively buoyant (< and >1 g cm^{-3}) microplastics, and b) microplastics less dense and denser than 1.2 g cm^{-3} (the number labels present the number of chemically analysed plastic particles), in the intertidal and subtidal sediments. Note that positive and negative buoyancy refers to fresh and sea water density of around 1 g cm^{-3}.

4.3. Polymer type, buoyancy and density separation

4.3.1. Polymer type

Intertidal sediments of Vava'u comprised mainly PP (27 %), PA (23 %), PE (12 %) and PES (12 %) microplastics. Clunies-Ross et al. (2016) found considerably more PS (55 %) in New Zealand beach sediments than was present in our study (4 %) (Table 5). We anticipated more PS microplastics in our samples because PS takeaway containers and styrofoam are common street and beach litter in Vava'u (A. Markic, pers. obs.). In other studies that analysed intertidal sediments, PE and PP were common as well (Hayes et al., 2021; de Lena et al., 2021). Interestingly, Bridson et al. (2020) recovered predominantly regenerated cellulose (34 %) and PET (22 %), with only 15 % of PE.

Subtidal sediments in Vava'u contained a surprisingly high percentage of PE (43 %), followed by rayon (20 %), PES (17 %) and PP (10 %). Ferreira et al. (2020) also reported PE (24 %) as the dominant polymer in the sediments of Fiji, together with PA (9.5 %), PET (9.5 %), PP (9.5 %) and polytetrafluoroethylene (PTFE) (9.5 %) (Table 5). Jahan et al. (2019) found PET and PA to be the most common polymers in the subtidal sediments of New South Wales ports, Australia, while Barrett et al. (2020) reported cis-polyisoprene (latex), PU, PES, PP as the predominant polymers in the deep-sea sediments of the Great Australian Bight.

4.3.2. Buoyancy

With respect to buoyancy, contrary to our expectations, over half of the microplastics extracted from Vava'u benthic sediments were made of materials that are positively buoyant in their virgin form (Fig. 8) (PE 43 %, PP 10 % and PUR 3 %, see Table 2) and should theoretically stay afloat in the surface waters. The negatively buoyant polymers common in our benthic sediments were RAY (20 %), PES (17 %), PA (3 %), mainly present in the form of fibres and filaments.

There are a number of factors that may lead to expanded PS (EPS) occurring in abundance in benthic sediments. Sagawa et al. (2018) detailed potential factors that changed the buoyancy of EPS from positive to negative. They describe extensive morphological changes in benthic EPS particles, compared to beach EPS. Sunken EPS was flat with collapsed pores and due to the loss of air from the pockets in the polymer structure, EPS buoyancy approached the negative buoyancy of PS (Table 2). Furthermore, the authors found that the surfaces of benthic EPS were fragmented and complex, with many cracks and cavities, some of which contained diatoms which could have also contributed to the decreased buoyancy. The diatoms were found in both benthic and beached EPS, which implies they were acquired while the particles were in the water column. Long et al. (2015) performed experiments on the effects of phytoplankton, such as diatoms and cryptophytes, on the sinking rates of microplastics, and found they indeed increased. Phytoplankton produces sticky algal aggregates which entrap microplastic debris and enables it to sink faster. Similarly, Porter et al. (2018) described incorporation of microplastics into marine snow, aggregates composed of organic and inorganic particulate matter dispersed in the water and merged by Brownian forces. The sinking rates of marine snow are much higher than those of the individual components, including microplastics. Cole et al. (2016) proposed the vertical transfer of microplastics via ingestion by zooplankton and their excretion in the form of faecal pellets, which subsequently sink. Another route, widely accepted, is change in buoyancy due to biofouling. In their experimental study, Kaiser et al. (2017) confirmed that the positively buoyant PE sinks

Table 4
Studies on microplastic pollution of the intertidal and subtidal sediments conducted in the South Pacific region, including islands and the continental coasts (n/a – information not available, LSL – lower size limit, pcs – pieces or particles, L – large microplastics, S – small microplastics, Ch – Chile, EI – Easter Island, A – Australia, V – Vanuatu, S – Solomon Islands).

Location/s	Year	Survey type	Methods	LSL (μm)	Units	Microplastics quantity	Reference
Intertidal							
New Zealand, Auckland	2018	Quantity	0.5 × 0.5 m quadrats, 5 cores per quadrat (D = 6 cm, 5 cm deep), NaCl solution	300	pcs kg^{-1}, m^{-2}, m^{-3}	6 pcs kg^{-1} 459 pcs m^{-2} 9188 pcs m^{-3}	Bridson et al., 2020
American Samoa	2017–18	Quantity	Hand shovel, top 10 cm (200 g), NaCl solution	0.7	n/a	n/a	Polidoro et al. (2022)
Tonga, Vava'u	2017	Quantity	Collection container, 1 L of sed, $CaCl_2$ solution	63	pcs L^{-1}, pcs m^{-2}	23.5 ± 1.9 pcs L^{-1}	This study
Pitcairn Islands, Henderson Is.	2015–19	Trends	1 × 1 m quadrats, top 1 cm, corer, $ZnCl_2$ solution	0.2	pcs kg^{-1}	1960 ± 356	Nichols et al., 2021
New Zealand, Northland	2020	Trends	0.5 × 0.5 m quadrats, cores 5 × 5 cm top 5 cm, NaI	32	pcs kg^{-1}, m^{-2}, m^{-3}	3 pcs kg^{-1} (L), 405 pcs kg^{-1} (S)	de Lena et al., 2021[a]
South Australia	n/a	Quantity	0.2 × 0.2 m quadrats, top 5 cm, NaCl solution	25	pcs kg^{-1}	0.5–2.2 ± 1.2	Hayes et al., 2021
New Zealand, Canterbury region	2013	Quantity	0.25 × 0.25 m quadrats, top 2 cm, NaCl solution	32	pcs kg^{-1}	0–45.4	Clunies-Ross et al., 2016
Chile, Pacific coast, Easter Island	2011	Quantity	0.5 × 0.5 m quadrats	1000	pcs m^{-2}	27 (Ch), 805 (EI)	Hidalgo-Ruz and Thiel, 2013
Australia & Chile (global study)	2004–07	Quantity	0.5 × 0.5 m quadrats, NaCl solution	Paper filter	pcs 250 mL^{-1}	2 (A), <10 (Ch)	Browne et al., 2011
SW Pacific Islands, Australia	n/a	Quantity	n/a	Pellets	pellet m^{-1}	0–≫2000	Gregory, 1990
New Zealand	1972–76	Quantity	Transect II to strandline, only pellets	Pellets	pellet m^{-1}	>100,000	Gregory, 1978
New Zealand	n/a	Quantity	n/a, only pellets collected	Pellets	pellet m^{-1}	0–>40,000	Gregory, 1977
Subtidal							
Chile	n/a	Quantity	Van Veen grab, NaCl solution	200	pcs kg^{-1}	72.2 ± 32.4	Jorquera et al., 2022
New Zealand, Marlborough Sounds	2020	Quantity	KC Denmark & Gravity corers, NaI solution	0.45	pcs cm^{-3}	0.3447–0.4545	Ribó et al., 2021
Vanuatu & Solomon Islands	2017–18	Quantity, trends	Van Veen grab, NaCl solution	0.2	pcs kg^{-1}	833–19,167 (V) 450–15,167 (S)	Bakir et al., 2020
Tonga, Vava'u	2017	Quantity	Collection container, 1 L of sed, $CaCl_2$ solution		pcs L^{-1}, pcs m^{-2}	15.0 ± 1.9 pcs L^{-1}	This study
Australia, East Coast	2017	Quantity	Ekman grab, NaI solution	1	pcs kg^{-1}	205–350	Jahan et al., 2019
Australia, Great Australian Bight	2017	Quantity	Deep-sea sediment cores, $ZnCl_2$, Red Nile	0.22	pcs g^{-1}	1.3 ± 0.7	Barrett et al., 2020
Fiji	2016–18	Quantity	Van Veen grabs, metal scoop (scuba), KO_3P, $Li_2O_{13}W_4^{-24}$ and NaCl solutions	300	pcs g^{-1}	0.008–0.034	Ferreira et al., 2020
South Australia	2015	Quantity	Van Veen grab, NaI solution	38	pcs mL^{-1}	3.4	Ling et al., 2017
Australia, Tasmania	2004	Trends	1-m deep cores, NaI solution	1.2	pcs g^{-1}	2.43–4.2	Willis et al., 2017

[a] In this study, large microplastics were defined as being between 300 μm and 5 mm in size. Small microplastics were smaller than 300 μm.

when exposed to biofouling in the marine environment. Fazey and Ryan (2016) demonstrated that the smaller the PE particle, the faster the biofouling-induced change in buoyancy. However, Amaral-Zettler et al. (2021) proposed that there is a tipping point (surface area to volume of particle equals 100), above which the smaller particles become too small for microbial colonisation substantial enough to cause the particle to sink. More generally, Kowalski et al. (2016) pointed out that the sinking rates of microplastics depend on multiple factors, which include particle density, size and shape, fluid density and temperature, as well as biofouling and weathering of the particle.

4.3.3. Density separation method

Floatation is the most common method to separate microplastics from sediments and has been proposed in several standardised methods, including NOAA (Masura et al., 2015) and JPI Oceans (Frias et al., 2018). Floatation can be achieved in several different ways, including: (i) mixing of sediment with salt solution in a vessel, (ii) elutriation apparatus, (iii) air-induced floatation, (iv) Munich Plastic Sediment Separator, or (v) pressed fluid extraction (Mai et al., 2018). For this study, floatation with salt solution was selected due to simplicity, affordability and no requirement for specialised equipment.

We used several criteria for the choice of salt for density separation and these included: low cost, availability in the Pacific region, high solubility to densities over 1.2 g cm^{-3} and non-toxicity. $CaCl_2$ fit all the criteria. Thus, we aimed to examine the efficacy of high-density $CaCl_2$ solution (ρ = 1.40–1.45 g cm^{-3}) and to determine the proportion of sediment microplastics that are denser than the hypersaline NaCl solution (1.2 g cm^{-3}) – the most commonly used solution for density separation of microplastics from the sediments (Bellasi et al., 2021; Cutroneo et al., 2021). The use of $CaCl_2$ theoretically enables isolation of common synthetic (and semi-synthetic, i.e. rayon) materials denser than 1.2 g cm^{-3} are PES, PVC, and RAY (Table 2).

Unlike many other calcium compounds, a typical ionic halide calcium chloride ($CaCl_2$) is very soluble. It is also referred to as one of the most versatile of the basic chemicals that have been produced commercially for over 100 years that has commonly applied in: concrete to speed up the initial setting, ice and refrigerating controls due to a highly exothermic dissolving process, but most importantly, due to its hygroscopic nature, as a desiccant capable of moist absorption to the point of turning into brine (Ropp, 2013). Its solubility rapidly increases with temperature, making its use particularly suitable for studies in the lower latitudes. $CaCl_2$ is produced directly from limestone or as a by-product of generating soda ash from brine. It is applied in medicine, listed as a permitted food additive in the European Union (Younes et al., 2019) and is generally considered safe by the U.S. Food and Drug Administration (FDA, 2022). In the Pacific region, $CaCl_2$ is a readily available and cheap chemical, since it is used in

Table 5
Studies in the South Pacific region that provided information on the polymer types in the analysed sediments (RC – regenerated cellulose, HDPE – high-density polyethylene, PTFE – polytetrafluoroethylene, KO_3P - potassium metaphosphate, $Li_2O_{13}W_4^{-24}$ - lithium metatungstate).

Study	Most common polymers found	High-density solution	Density (g cm^{-3})
Intertidal			
Hayes et al., 2021	PP, HDPE, PS	NaCl	1.2
Bridson et al., 2020	RC (cellulose) (34 %), PET (22 %), PE (15 %)	NaCl	1.2
Clunies-Ross et al., 2016	PS (55 %), PE (21 %), PP (11 %)	NaCl	1.2
Browne et al., 2011	PES (56 %), ACR (23 %), PP (7 %), PE (6 %)	NaCl	n/a
de Lena et al., 2021	PE, PP, PET	NaI	1.6
Subtidal			
Jahan et al., 2019	PET, PA, PE, PS, PES	NaI	1.2
Jorquera et al., 2022	Cellulose-like (45 %), cotton-like (20 %), PET (11 %), Acrylics (2 %), PP (1.5 %), PUR (0.5 %)	NaCl	1.23
Barrett et al., 2020	Cis-polyisoprene (latex), PU, PES, PP	$ZnCl_2$	1.37
Ferreira et al., 2020	PE (24 %), PA, PET, PP and PTFE (each 9.5 %)	KO_3P[a], $Li_2O_{13}W_4^{-24}$, NaCl[a]	1.6

[a] It is unclear what the function of KO_3P was, nor the density of NaCl solution used at the end.

swimming pool maintenance. In pool shops, it can be purchased for less than $15 NZ/kg (e.g. Perfect Pools and Pool Doctor in Auckland). Another positive of $CaCl_2$ is that waste disposal is easier and safer, which is especially important for the SIDS where presumably hazardous waste disposal facilities may be limited.

The downside of the $CaCl_2$ high-density solution is its viscosity. It can be challenging to handle the equipment when it becomes slippery from the solution. Additionally, its solubility, highly dependent on the temperature, can prove disadvantageous if the temperature drops. Our labwork involving $CaCl_2$ was not done in early morning because it was too cold (<25 °C) to keep the solution from crystallising.

Regarding the dense plastics in our sediments, PES (ρ = 1.31–1.39 g cm^{-3}) and RAY (ρ = 1.52 g cm^{-3}) were present in both intertidal and subtidal sediments. The polymers were mainly in the form of fibres, with only two PES particles being fragments, most likely PET, the most common type of non-fibrous PES. Surprisingly, polymer characterisation did not detect any PVC particles (ρ = 1.2–1.44 g cm^{-3}), and interestingly, it was reported in only one of the studies from Table 5, but in minute concentrations (de Lena et al., 2021, p. 13). A possible reason that we did not find any PVC is that the sample size (N = 70) of the particles analysed by FTIR spectroscopy was too small and by chance, none of the particles were made of PVC.

In two studies where $CaCl_2$ solution was used for density separation, more of the less dense polymers were reported, such as PE, PP and PS (Li et al., 2018; Schröder et al., 2021). This could potentially be because the examined sediments were beach sediments, and in our study, we found more of the denser microplastics in the subtidal sediments. In one study on Arctic seafloor sediments, cellulose fibres were predominantly found (57 %), while only 11 % were synthetic polymers identified as PES, polyacrylonitrile and PU (Adams et al., 2021). In another two studies that used $CaCl_2$ solution, polymer characterisation was not performed (Stolte et al., 2015; Collicutt et al., 2019).

A recent experimental methodological study compared the efficacy of four commonly used salts in microplastics density separation procedure: NaCl, NaI, $ZnCl_2$ and $CaCl_2$ (Duong et al., 2022). The most successful recovery rate of the test microplastics was obtained with $ZnCl_2$ (84 %, 1.5 g cm^{-3}), followed by NaI (82 %, 1.6 g cm^{-3}) and NaCl (80 %, 1.2 g cm^{-3}), while $CaCl_2$ exhibited the poorest performance (68 %, 1.4 g cm^{-3}). The authors attributed this low recovery rate of $CaCl_2$ to its high viscosity and '*the remaining flocculation of calcium ions (Ca^{2+}) with organic matter after the oxidation process*'. Our method testing resulted in 100 % recovery of all test beads, in two separate trials. However, our test beads from a cosmetic product were most likely PE, the polymer that was also recovered quite successfully in the Duong et al. study (~80 %). The authors tested the recovery on six different polymers (PE, PP, HDPE, PS, PVC, PET) and $CaCl_2$ demonstrated poor recovery for PVC (<30 %) and PET (~55 %). Thus, this might be another potential reason for not finding any PVC particles in our samples.

Some studies focused on both method development and extraction of microplastics from environmental samples (Nuelle et al., 2014; Coppock et al., 2017). Findings of the studies, which used denser solutions than NaCl, demonstrated that sometimes predominantly denser polymers were extracted (e.g. Coppock et al., 2017; Martin et al., 2017; Naji et al., 2017), while in other studies, mainly lighter polymers (Dekiff et al., 2014). Similarly, some studies in which NaCl solution was used, recovered mainly dense polymers (e.g. Woodall et al., 2014; Frias et al., 2016), and others mainly lighter polymers (e.g. Vianello et al., 2013). However, Duong et al. (2022) demonstrated that NaCl solution is not so effective in extracting heavy polymers, with recovery rates of ~35 % for PVC and 70 % for PET. It is not clear how the hypersaline NaCl solution could extract denser polymers, unless these were very small particles that remained dispersed in the solution at the time of decantation, due to short settlement time.

In conclusion, it is not clear whether $CaCl_2$ in our study did not manage to isolate PVC and other dense polymers or the chemically analysed polymers simply did not include any of the PVC particles. Furthermore, ideally, we would have carried out an experimental methodological study to compare the efficacy of these two salts with different polymer types; however, due to time limitation, it was not performed.

4.4. Recommendations for future work

Even though $CaCl_2$ might not be an ideal salt for a high-density solution, we still recommend it for studies with limited resources. With respect to high-density solutions other than $CaCl_2$, according to Duong et al., 2022, $ZnCl_2$ and NaI seem to be the most suitable options owing to their high solubility. However, due to their high cost, perhaps the higher densities could be achieved in two steps, first creating a 1.2 or 1.4 g cm^{-3} density solution using NaCl or $CaCl_2$, respectively, and then increasing to 1.5 g cm^{-3} or more by adding $ZnCl_2$ or NaI. It should be noted we have not tried it and do not know how these chemicals react with each other. Mixing of chemicals could be avoided by having two separate steps of density separation (e.g. Nava and Leoni, 2021). In the first step, the density separation is done with NaCl and in the second with NaI. However, this does not decrease the amount of NaI used. Nevertheless, the chemicals could and should be recycled when possible (e.g. Kedzierski et al., 2017) to avoid unnecessary disposal of hazardous chemicals. Perhaps, the high-density solution could be desiccated and returned back to solid crystal form.

We do not recommend omission of synthetic fibres as this could lead to serious underestimation of microplastics quantities. We found fibres in amounts that could not be airborne contamination, including other studies as well (e.g. Ling et al., 2017; Bridson et al., 2020). Instead, all measures should be taken to minimise airborne contamination and blank lab tests should be used for reference. Additionally, if filtered water is not available for rinsing equipment (i.e. nets, filters, tweezers), we believe seawater could be used, as we found a maximum of one particle in 333 L of water (Markic et al., 2022). Even if a microplastic particle gets in contact with equipment, it would likely slip off with excess water. Similarly, if there are any microplastics in tap water, it is unlikely that a particle would remain on the equipment after rinsing. Thus, the likelihood of contamination with microplastics from the ocean or tap water during rinsing is highly unlikely.

Recipe for low-budget research

1. Use cheap and readily available chemicals
2. Borrow equipment
3. Custom-make equipment
4. Buy equipment in regular shops (bowls, dishes, stirrers)
5. Use alternatives (e.g. standup paddleboard pump instead of a vacuum pump)
6. Use test microbeads from cosmetic products or use plastic objects to make test microplastics
7. Collaborate with institutions to have expensive analyses done
8. Use free or cheap online software for analysis
9. Work with volunteers
10. In general, be resourceful.

CRediT authorship contribution statement

Ana Markic: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Writing – Original draft, Writing – Review & editing, Project administration
James H. Bridson: Methodology (FTIR), Formal analysis, Investigation, Writing – Review & editing
Peta Morton: Methodology, Formal analysis, Investigation, Writing – Review & editing
Lucy Hersey: Methodology, Formal analysis, Investigation, Writing – Review & editing
Andrea Budiša: Validation, Writing – Review & editing (advising)
Thomas Maes: Validation, Writing – Review & editing (advising)
Melissa Bowen: Validation, Writing – Review & editing (advising).

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

We submitted supplementary data.

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Appendix A. Supplementary data

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