



Microplastic pollution in the surface waters of Vava'u, Tonga

Ana Markic^{a,*}, James H. Bridson^b, Peta Morton^c, Lucy Hersey^d, Thomas Maes^e,
Melissa Bowen^f

^a Blue Spark Put za Marleru 20, 52204 Ližnjan, Croatia

^b Scion, Titokorangi Drive, Private Bag 3020, Rotorua 3046, New Zealand

^c University of Sydney, Camperdown, NSW 2006, Australia

^d Monash University, Wellington Road, Clayton 3800, Victoria, Australia

^e Grid-Arendal, Teaterplassen 3, 4836 Arendal, Norway

^f School of Environment, University of Auckland, Auckland 1010, New Zealand

ARTICLE INFO

Keywords:

Microplastics
Plastic pollution
Surface waters
Plankton net trawls
Tonga
Pacific

ABSTRACT

Marine plastic pollution, particularly microplastics, has been recognised as a global issue in the recent years, but research efforts in the Pacific are lagging. We carried out research on microplastics contamination of surface waters of the Vava'u archipelago, Tonga. Since microplastics smaller than the standard mesh size (333–335 µm) are readily reported in the literature on microplastics, we used a finer plankton net (100 µm) to determine the proportion of captured microplastics smaller than 300 µm. Isolated microplastics were counted and measured using stereomicroscope with polymer identification performed by FTIR spectroscopy. The analysis revealed high microplastics concentrations ($329,299.7 \pm 40,994.2$ pcs km⁻² or 1.05 ± 0.13 pcs m⁻³). The proportion of particles smaller than 300 µm was 40 %. The predominant type of microplastics in surface waters were small bits of white film, which we associated with cement-filled white bags used to construct docks throughout Vava'u, often heavily eroded.

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 several decades ago, marine plastic pollution continues to prompt questions about human and environmental health as the annual global production and consumption of plastics increases exponentially (PlasticsEurope, 2021). In response, research on the levels and impacts of plastic pollution in world's oceans has noticeably intensified in the recent years. However, research effort in the South Pacific islands and Oceania seems to be lagging and most likely due to lack of researchers and access to suitable infrastructure. Quantification of microplastics by the most common approaches requires specialist laboratory facilities, equipment and chemicals, making it challenging for many SIDS.

Plastic debris and microplastics have been documented in the digestive system of marine organisms, regularly consumed as seafood in the Pacific, such as fish and shellfish (Van Cauwenbergh and Janssen,

2014; Rochman, 2015; Forrest and Hindell, 2018; Markic et al., 2018; Bakir et al., 2020; Alfaro-Núñez et al., 2021). As a result, concerns are rising about the indirect impacts of plastic pollution on human health (Menéndez-Pedriz and Jaumot, 2020; Davison et al., 2021; Yee et al., 2021). The SIDS of the South Pacific are located in the South Pacific subtropical gyre, a region of converging surface currents, that traps floating particles such as plastics (Eriksen et al., 2013; Cózar et al., 2014; Lebreton et al., 2012), and will be accumulating this material for many decades. Establishing a baseline measurement of plastic pollution in these regions now is essential for understanding change and mitigation needs in future.

As research on plastic pollution in the South Pacific islands, particularly on microplastics, is scarce, the primary aim of our study was to carry out the first investigation of microplastic pollution in the Vava'u archipelago, Tonga, to obtain baseline concentrations and to describe the characteristics of the collected microplastics. A second aim was to determine the proportion of surface water microplastics smaller than the standard mesh size of common sampling nets.

* Corresponding author.

E-mail address: sve.se.more@gmail.com (A. Markic).

<https://doi.org/10.1016/j.marpolbul.2022.114243>

Received 26 May 2022; Received in revised form 7 October 2022; Accepted 8 October 2022

Available online 29 October 2022

0025-326X/© 2022 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

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 a 120-foot vessel Infinity, on multiple sites within the archipelago. A research permit was obtained from the Ministry of Education and Training of the Kingdom of Tonga for the activity. Two types of assessment were done: macroscopic, or in situ naked-eye quantification of debris larger than 1 mm, and microscopic, quantification of debris over 63 μm in field-collected sea water and sediment samples using a dissecting microscope. However, in this paper, we will present and discuss only the methodology and results concerning microplastics in the surface waters.

To assess the quantities of floating plastic debris, we collected volume-reduced samples of the surface water using a phytoplankton net (0.8 m diameter mouth, 100 μm mesh size), by dragging the net next to the dinghy (Fig. 2) along a 500 m transect at the speed of 2–3 knots. Each transect covered a surface area of 400 m^2 or 0.0004 km^2 , occupying a volume of 125,600 L or 125.6 m^3 of water. Since previous work found 19.1 % (67 out 351) of microplastics extracted from South Pacific fish (Markic et al., 2018) is $\leq 300 \mu\text{m}$, we used a 100- μm mesh. We aimed to assess the proportion of surface plastics smaller than the standard mesh size ($\sim 333 \mu\text{m}$) of neuston nets commonly used for collecting surface water microplastics (Cutroneo et al., 2020).

The direction of the boat was always perpendicular to the main current to avoid sampling variable volumes of water with variable flow. If there was wind, the net was always placed on the leeward side of the boat. Only half of the net opening was immersed (Fig. 3), so the surface of the water coincided with the net's diameter. After each tow, debris that was left in the cod end was washed out into a freshly rinsed plastic zip-lock bag using a squeeze bottle, transported to the main vessel and frozen.

2.2. Sample processing and contamination control

At the onboard lab, the water samples were defrosted and vacuum filtered over a stainless-steel filter (63 μm). The filter was then visually analysed under a dissecting microscope. All particles resembling synthetic materials (i.e. no cellular structure, uniform colour) were isolated, separated by form (fibre, fragment, film), colour (all colours) and size ($<100 \mu\text{m}$, 100–200 μm , 200–300 μm , 300–400 μm , 400–500 μm , 500–1000 μm , 1–2 mm, 2–5 mm, $>5 \text{ mm}$), counted and stored into 2-mL glass vials for further analysis (i.e. polymer characterisation). Various verification methods were used during the microscopic identification to confirm the synthetic polymer origin and to eliminate minerals, rocks, shells, organic particles and other natural material. These methods include the floating test, Nile red dying test and poking test. For the

floating test, we used high density CaCl_2 solution ($\rho = 1.40\text{--}1.45 \text{ g cm}^{-3}$), a medium in which the most common plastic polymers float, but minerals, rocks and shells sink. Nile red dyes organic matter in pink, leaving plastic undyed. Poking a piece of plastic with a needle is more of a sensory test and, unlike rocks and minerals, we feel that the plastic is soft or rubbery when poked. To avoid air-borne contamination, all samples were always covered with aluminium foil when not working with them. Blank tests were used every few samples and were kept uncovered only while processing a sample, which was very short. We did not find any microplastic contamination on them.

2.3. Polymer characterisation – chemical analysis of retrieved plastics

Fourier transform infrared (FTIR) spectroscopy was used for polymer identification. The analysis was done in collaboration with Scion (New Zealand Crown Research Institute). FTIR spectra were obtained for a random subset of 37 surface water microplastic particles. Before analysis, all samples were dried at 70 $^\circ\text{C}$ for 4 h. Larger microplastics ($>300 \mu\text{m}$) 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 ($<300 \mu\text{m}$) were analysed using a Bruker Tensor 27 Instrument connected to a Bruker IIRscope 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), 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 hit quality index (HQI) $\geq 70 \%$ was classified as positive identification, HQI = 60–70 % required user interpretation and HQI $< 60 \%$ was classified as inconclusive.

2.4. 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 items or particles per area and per volume. Pieces (pcs) and particles are used interchangeably, as they indicate the

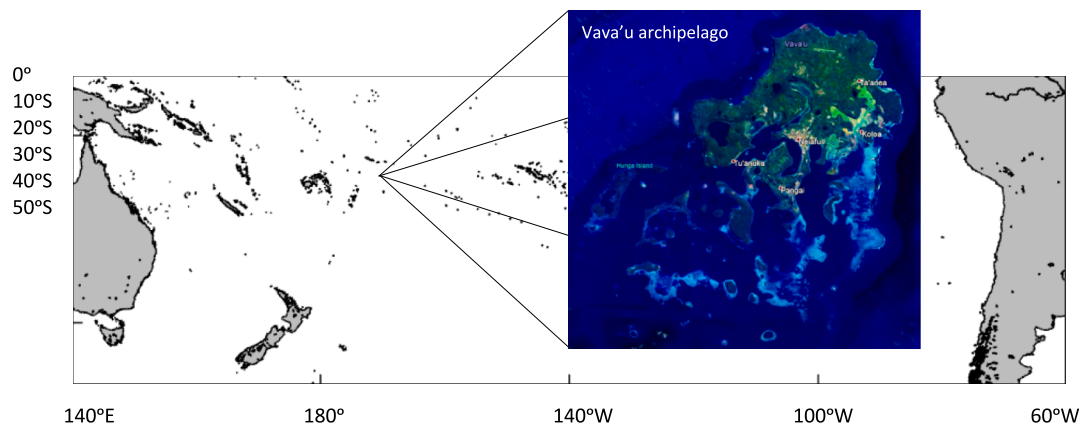


Fig. 1. Study location showing Vava'u archipelago, Tonga, and its position in the South Pacific.

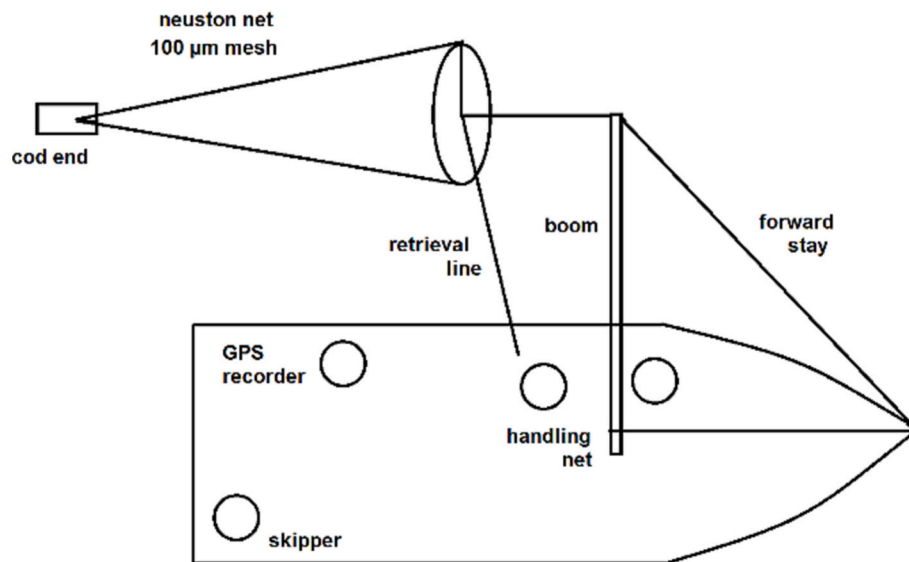


Fig. 2. Surface water sampling using a phytoplankton net dragged on the side of the dinghy.



Fig. 3. The phytoplankton net position in the water during sampling.

count or the number of micro-debris plastic 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 only in the tables, excluding the discussion where the less common units were used for comparison with other studies. In some studies, the mass of debris is also reported but we did not have the opportunity to record mass for each debris item or microscopic particle. Additionally, since the FTIR analysis showed a misidentification error of 2.7 % of the analysed particles, for a more accurate and conservative result, we adjusted the number of particles in the concentration calculation for this error.

To express the precision of estimated plastics concentrations, we provided 95 % confidence intervals 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/>).

3. Results

3.1. Quantitative assessment of surface water microplastics

The concentration of microplastics in the surface waters was assessed on 32 trawl transects. We did not encounter problems with clogging of the net by plankton and other organic material, and generally, there was very little organic content in the samples. Microplastic debris was found in all surface trawls with the average of 132 particles per trawl. The estimated average concentration across the 32 transects is $329,299.7 \pm 40,994.2$ pcs km^{-2} , or 1.05 ± 0.13 pcs m^{-3} (Table 1), with a maximum of 975,432.5 pcs km^{-2} southeast from Neiafu, the largest village in the archipelago (Fig. 4).

Table 1

Quantification of microplastics in the surface waters of Vava'u (SE – standard error, pcs – pieces or particles, LL – lower limit, UL – upper limit).

Number of samples (trawls)	32
Sample area (m ²)	400
Sample area (km ²)	0.0004
Sample volume (L = dm ³)	125,600
Sample volume (m ³)	125.6
Total number of particles before FTIR	4332
Total number of particles with 2.7 % error applied	4215
Average number of particles per trawl	132
Average concentration (pcs m ⁻² ± SE)	0.33 ± 0.04^a
Minimum concentration (pcs m ⁻²)	0.01
Maximum concentration (pcs m ⁻²)	0.98
95 % confidence intervals (LL, UL)	0.25, 0.41
Average concentration (pcs km ⁻² ± SE)	329,299.7 ± 40,994.2
Minimum concentration (pcs km ⁻²)	12,162.5
Maximum concentration (pcs km ⁻²)	975,432.5
95 % confidence intervals (LL, UL)	248,951.1, 409,648.3
Average concentration (pcs L ⁻¹ ± SE)	0.001 ± 0.0001
Minimum concentration (pcs L ⁻¹)	0.00004
Maximum concentration (pcs L ⁻¹)	0.003
95 % confidence intervals (LL, UL)	0.0008, 0.0013
Average concentration (pcs m ⁻³ ± SE)	1.05 ± 0.13
Minimum concentration (pcs m ⁻³)	0.04
Maximum concentration (pcs m ⁻³)	3.11
95 % confidence intervals (LL, UL)	0.79, 1.30

^a Bold values are the average microplastics concentration in four different units.

3.2. Qualitative assessment of surface water microplastics

Size distribution – The size of the microplastics from the surface waters was measured using 783 particles from six randomly chosen samples. These six trawls were spread out in the Vava'u archipelago, covering all sampled areas of the archipelago. Particles smaller than 300 µm were the most prevalent (40 %) (Fig. 5a).

Form – The form of the microplastic particles was determined for all collected particles. Surface waters in Vava'u are mainly

contaminated by small bits of film (Fig. 5b), similar in appearance to a shredded plastic bag, followed by synthetic fibres.

Colour composition – The colour of the microplastic particles was determined for all collected particles. The shredded fragments of plastic film were mainly white (Figs. 6a, 7), resulting in over 60 % of surface microplastics being white (Fig. 6b).

Polymer characterisation – FTIR analysis was performed on 37 random surface water particles to validate the visual identification of microplastics and to determine the common polymer types. Out of these 37 particles, only one particle of CaCO₃ was misidentified. Another six particles had a poor quality spectra and one was inconclusive (where the percent match with database spectra was <60 %). Poor quality spectra could be a result of material degradation, biofouling, or very small particle size leading to weak signal intensity. Microplastic abundance results were adjusted based on a misidentification rate of 1 out of 37 (2.7 %), but not for particles with poor or inconclusive spectra, for which visual identification was given precedence. The most common polymer types identified in the subset of samples was PES, PP and PE (Fig. 8a). Examples of FTIR spectra for PP and PES are shown on Fig. 9. Positively and negatively buoyant plastics were found in similar proportions (Fig. 8b).

4. Discussion

4.1. Quantitative comparison to other regional studies

The average concentration of microplastics in the surface waters of Vava'u expressed as the number of particles in surface area ($329,299.7 \pm 40,994.2$ pcs km⁻²) is greater than the concentrations reported in most other studies from the South Pacific region (Table 2). In fact, Vava'u concentrations are comparable to those obtained in the North Pacific subtropical gyre in 1999 ($334,271$ pcs km⁻²), also known as the 'garbage patch' (Moore et al., 2001). Subtropical gyres are oceanic accumulation zones containing exceptionally high concentrations of plastic debris. Eriksen et al. (2013) sampled the surface waters of the South Pacific subtropical gyre and found on average 26,898 particles km⁻² and a maximum of 396,342 particles km⁻², while the maximum

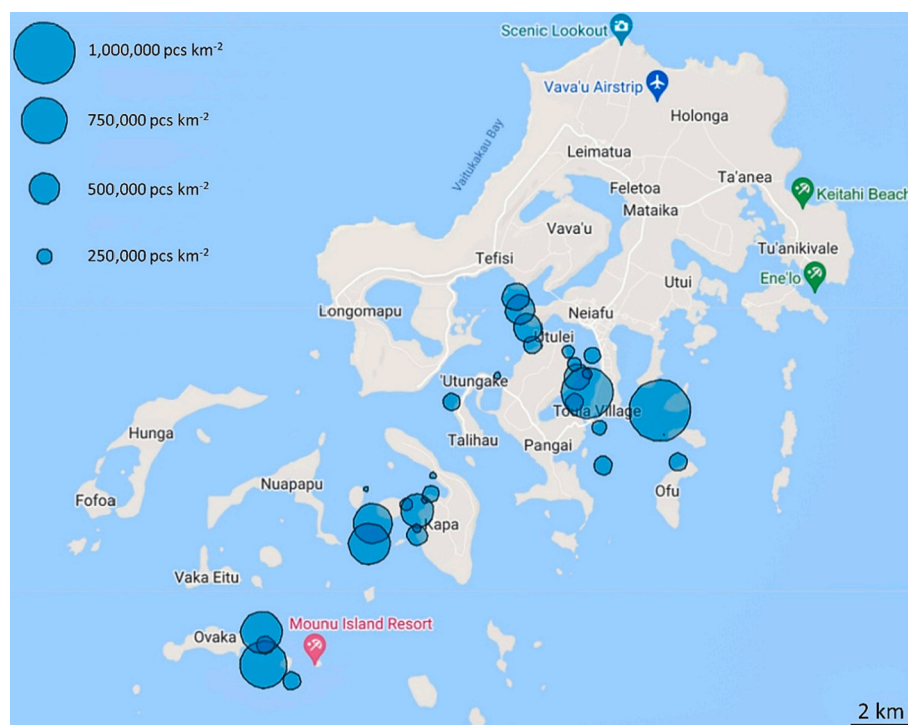


Fig. 4. Concentrations of microplastics in the surface waters of Vava'u archipelago (pcs – pieces of microplastics).

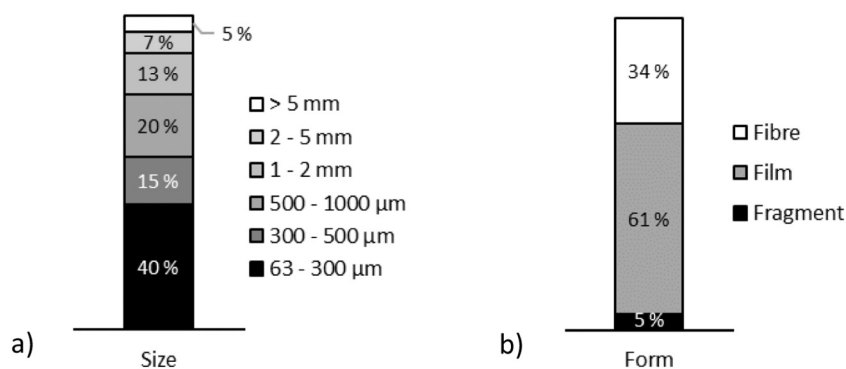


Fig. 5. Distribution of microplastics in the surface waters by: a) size and b) form.

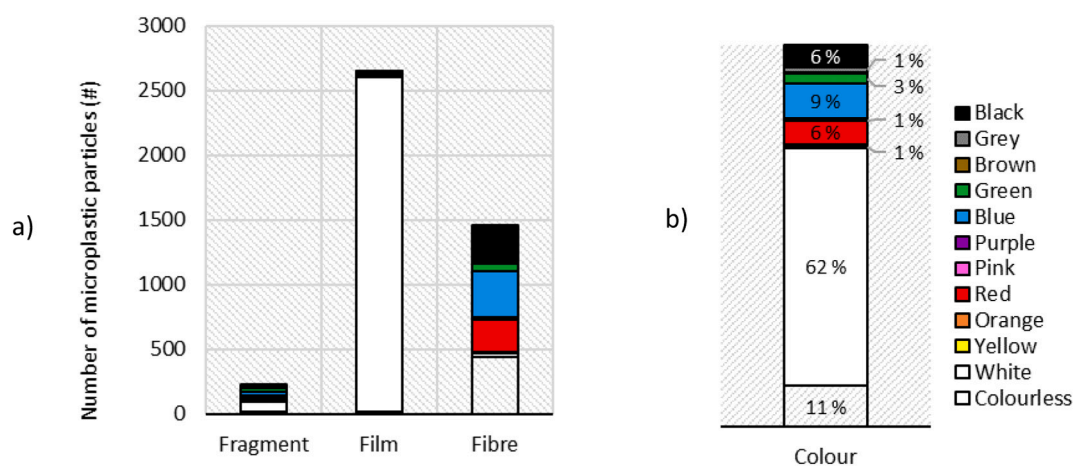


Fig. 6. Colour composition of microplastics a) broken down to plastic type and b) overall.

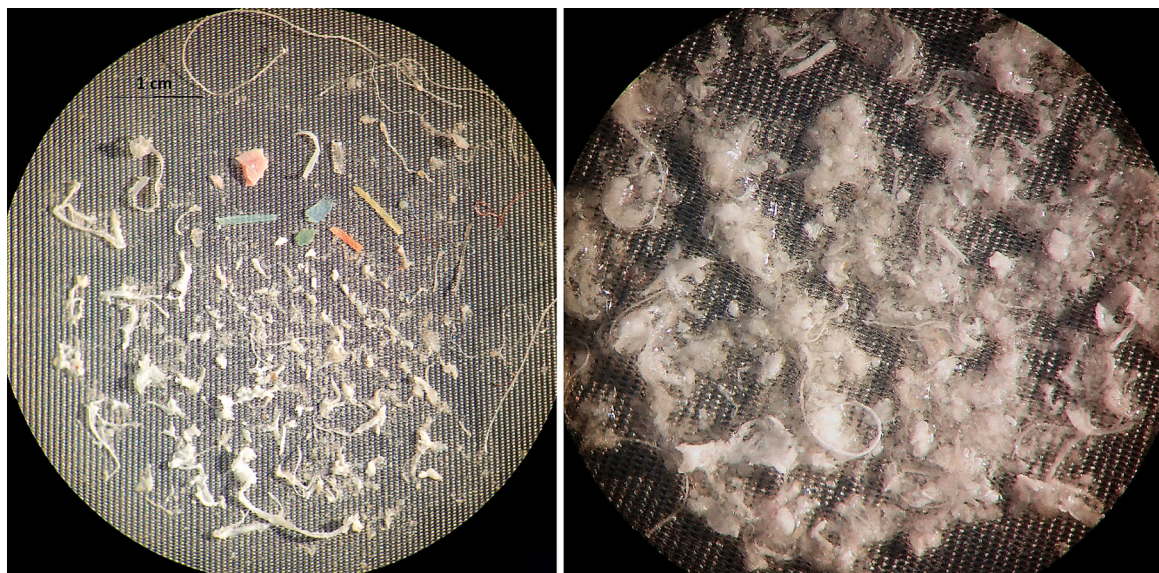


Fig. 7. Microplastics from Vava'u surface waters, near Neiafu (left) – 179 pieces extracted from one sample 1 transect, 125 m³ of water (station Mt. Talau, coordinates: S 18°38.578' W 174°00.404'). The image on the right shows white bits of plastic film or bag, the most common type of microplastic extracted from the surface water.

concentration in Vava'u reached 975,432.5 particles km⁻². Low microplastics concentrations were reported in the western South Pacific, reaching a maximum of 23,611 particles km⁻² between the east coast of Australia and the islands of Fiji (Reisser et al., 2013). Bakir et al. (2020)

reported, on average, microplastics concentrations of 51,144 pcs km⁻² in Vanuatu.

Some Pacific studies report microplastics concentrations greater than the ones found in Vava'u. For example, Rudduck et al. (2017) reported

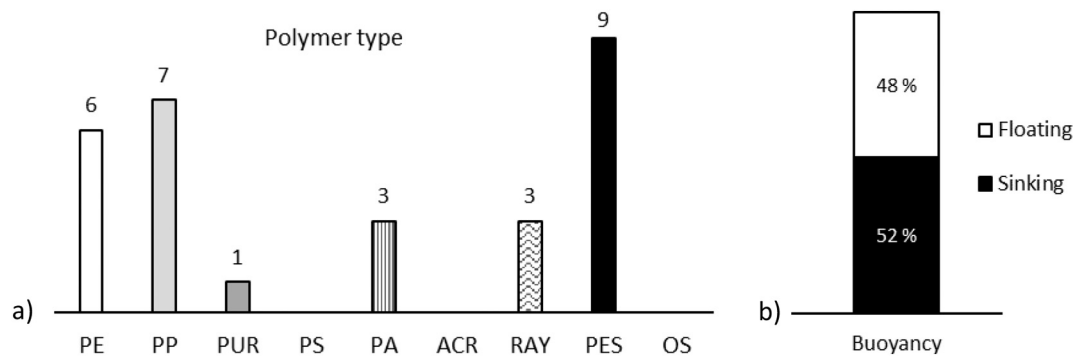


Fig. 8. Surface water microplastics by a) polymer type and b) buoyancy.

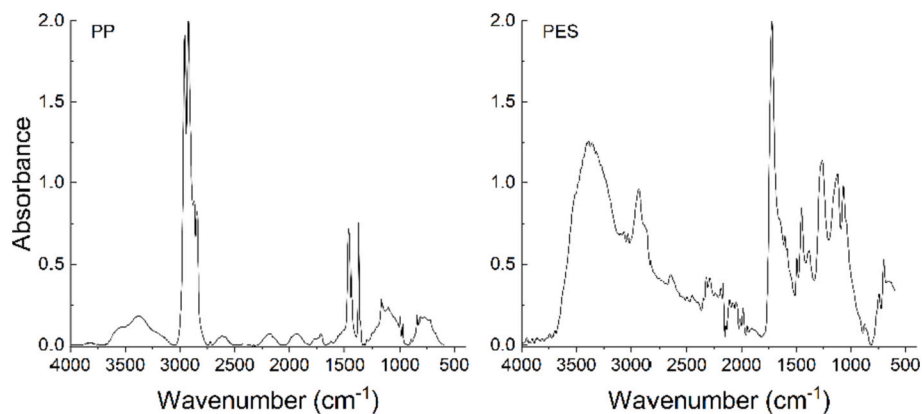


Fig. 9. Examples of PP and PES FTIR spectra.

Table 2

Studies on microplastics in surface waters conducted in the South Pacific region, including islands and the continental coasts (n/a – information not available, LSL – lower size limit; pcs – particles or pieces; S – surface, V – vertical).

Locations	Year	Survey type	Methods	LSL (μm)	Units	Debris quantity	Reference
Viti Levu, Fiji	2018	Quantity	Niskin bottle, 1 L samples	0.45	pcs L ⁻¹	1–2.9	Dehm et al., 2020
Vanuatu	2018	Quantity	Manta trawl (5 Gyres)	335	pcs km ⁻²	51,144	Bakir et al., 2020
Galapagos Islands, East Pacific	2018	Quantity	Surface trawls, net	150	μp m ⁻³ (i.e. pcs m ⁻³)	0.22–0.36	Alfaro-Núñez et al., 2021
Tuamotu Archipelago, French Polynesia	2017–18	Quantity	Surface samples and vertical samples, net	335 & 40	pcs m ⁻³	3.3 (0.2–8.4) (S) 14–716.2 (V)	Gardon et al., 2021
Vava'u, Tonga	2017	Quantity	Surface trawls, net	100	pcs km⁻² m⁻², m⁻³, L⁻¹	329,299.7 ± 40,994.2 pcs km⁻²	This study
Viti Levu, Fiji	2016–18	Quantity	Surface trawls, net	125	pcs m ⁻³	0.09–0.24	Ferreira et al., 2020
French Polynesia, Moorea	2016	Quantity	Surface trawls, net	500	pcs m ⁻²	0.74	Connors, 2017
Southeast Pacific	2015	Quantity	Surface trawls, net	333	pcs km ⁻²	~10,000	Eriksen et al., 2018
Australia, Tasmania	2013–14	Trends	Surface trawls, net	333	pcs km ⁻²	10,720 (2013) 571,932 (2014)	Rudduck et al., 2017
Australia, all around	n/a	Quantity	Surface trawls, net	333–335	pcs km ⁻²	4256.4	Reisser et al., 2013
Southeast Pacific	2011	Quantity	Surface trawls	333	pcs km ⁻²	26,898	Eriksen et al., 2013
Ross Sea, Antarctica	2010	Quantity	Pump	1	pcs m ⁻³	0.17 ± 0.34	Cincinelli et al., 2017
New Zealand, Hauraki Gulf	2008	Quantity	Surface trawls, net	250	pcs trawl ⁻¹	2317–16,626	Young and Adams, 2010
Pacific, Western Pacific (North & South)	2000–01	Quantity	Surface trawls, net	1000–1640		~263.5 (S Pacific trawls)	Uchida et al., 2016
New Zealand & Southern Ocean	n/a	Quantity	Surface trawls, net	n/a	n/a	n/a	Gregory, 1987

great concentrations in Tasmanian waters, with a maximum average of 571,931.8 particles km⁻² in the harbour sites in 2014, and a maximum as high as 2,258,665 pieces km⁻² per single trawl. The authors also found substantial temporal (inter-annual) and spatial variations (harbour vs. offshore) in surface microplastics concentrations with low average concentrations in the same harbour in 2013 (10,719.6 pieces

km⁻²). Connors (2017) found microplastics concentration in Moorea, French Polynesia (0.74 pcs m⁻² or 740,000 pcs km⁻²), double than that of Vava'u. However, the author collected samples in shallow intertidal waters of a public beach and estimated the concentrations based on six 3-m trawls which yielded 4 plastic particles in total. This estimate does not deem statistically sound or representative of surface microplastics

concentrations in the waters of Moorea.

With respect to concentrations expressed as the number of particles per volume of water (m^{-3} or L^{-1}), Dehm et al. (2020) reported greater concentrations in Fiji (Table 2). However, the methodology used in their study was different to ours, as we used volume-reduced samples, while Dehm et al. collected bulk samples (1 L samples) in the coastal waters shallower than 1 m, which makes our results difficult to compare. Interestingly, one would expect bulk samples to be quite ineffective (e.g. Tamminga et al., 2018), yet the authors reported more than one particle per litre. Furthermore, Alfaro-Núñez et al. (2021) and Ferreira et al. (2020) reported concentrations in Galapagos Islands and Fiji, respectively, lower than ours in Vava'u, using a similar methodology (including the net mesh size, 150 and 125 μm), while Gardon et al. (2021) report greater concentrations in Tuamotu Archipelago, particularly in the water column (Table 2).

4.2. Microplastics size and methodological variations

Size distribution of the surface water microplastics in Vava'u showed that as much as 40 % of all surface microplastic particles were smaller than 300 μm (Fig. 5). The most common sampling nets used to collect surface water microplastics are the standard 330, 333 and 335- μm nets (Prata et al., 2019; Stock et al., 2019; Cutroneo et al., 2020). Accordingly, most studies that used these mesh sizes reported concentrations based on microplastics larger than 300 μm (Table 3). If we exclude 40 % of particles from the total of surface microplastics, average surface concentrations in Vava'u decrease from 329,299.7 to 197,579.8 pcs km^{-2} . However, even these decreased concentrations are greater than concentrations in many studies from Table 2.

To examine the effect of different mesh sizes on microplastics concentrations, Lindeque et al. (2020) verified that, as expected, the smaller the mesh size of the sampling nets, the greater the resultant concentration. In their study, microplastics concentrations obtained with 100- μm mesh were 2.5-fold and 10-fold greater than concentrations obtained with 333 and 500- μm mesh, respectively. Similar findings were obtained in other studies (e.g. Frias et al., 2014; Gardon et al., 2021). Gardon et al. (2021) reported much lower microplastics concentrations in the surface waters (0.2–8.4 pcs m^{-3}) using a 335- μm mesh, than in the water column (14–716.2 pcs m^{-3}) using a 40- μm mesh. However, in contrast, Schmidt et al. (2018) assessed microplastic concentrations using nets with two mesh sizes – 330 μm and 780 μm , with subsequent filtration of the samples with 125- μm mesh filters and found no particles smaller than 400 μm , and no difference in size distribution of microplastics collected with the two different nets, which presumably means the mesh size did not affect the quantities either.

The difference in microplastics concentration between different methods can range across several orders of magnitude. Apart from neuston nets being used to collect surface water samples, other sampling techniques have been reported as well, for example the intake pump system of the vessel (e.g. Desforges et al., 2014; Lusher et al., 2015; Cincinelli et al., 2017; Cai et al., 2018) and other types of pumps (Zhao et al., 2014; Preston-Whyte et al., 2021), or bulk water samples (Song et al., 2014; Karlsson et al., 2017). The pumps allow the use of filters with particularly small mesh size, some as small as 1 μm (e.g. Cincinelli et al., 2017), which is probably why some of these studies reported exceptionally high concentrations. When the efficacy of the pump system is compared to the standard nets, the concentrations obtained with pumps are much greater (e.g. Zhao et al., 2014; Lusher et al., 2015; Cai et al., 2018). For example, Cai et al. (2018) recorded concentrations in South China Sea almost 60,000 times greater with a pump system (44- μm mesh, 2569 particles m^{-3}) than the standard net (333- μm mesh, 0.045 particles m^{-3}). The authors also found that 92 % of microplastics they collected were smaller than 300 μm . Lusher et al. (2015) reported average concentrations of 2.7 particles m^{-3} in the Arctic waters using the pump system, and only 0.34 particles m^{-3} when using the nets. Authors generally suggest that the pump method is considerably more

Table 3

A list of studies on surface microplastics in which various mesh-size nets and pumps were used for sampling. 'Trawl net' here refers to various sampling nets that are dragged on the surface or subsurface to collect samples (e.g. neuston net, manta net, plankton net, bongo net).

Mesh size (μm)	Minimum reported particle size (μm)	Method	Reference
1	60	Pump	Cincinelli et al., 2017
50	n/a	Hand net	Connors, 2017
62.5	62	Pump	Desforges et al., 2014
80	80	Small net	Nel and Froneman, 2015
100	63	Trawl net	This study
180, 280, 335	n/a	Trawl net	Frias et al., 2014
200	200	Trawl net	Cózar et al., 2015
200	200	Trawl net	Panti et al., 2015
300	250	Trawl net	Naidoo et al., 2015
300	n/a	Trawl net	Ivar do Sul et al., 2013, 2014
300	300	Trawl net	Faure et al., 2015
300	100	Trawl net	Gallagher et al., 2016
300	n/a	Trawl net	Lima et al., 2014
330	n/a	Trawl net	Carpenter and Smith, 1972
330	<1000	Trawl net	Li et al., 2021
32, 333	500	Pump, trawl net	Zhao et al., 2014
44, 333	20, 333	Pump, trawl net	Cai et al., 2018
50, 333	n/a	Hand net, trawl net	Song et al., 2014
250, 333	250	Pump, trawl net	Lusher et al., 2014, 2015
333	<333	Trawl net	Zhang et al., 2020
333	400	Trawl net	Reisser et al., 2013
333	355	Trawl net	Eriksen et al., 2013
333	330	Trawl net	Rudduck et al., 2017
333	355	Trawl net	Maes et al., 2017
333	300	Trawl net	Zhang et al., 2017
333	355	Trawl net	Moore et al., 2001, 2002
333	330	Trawl net	Kang et al., 2015
333	100	Trawl net	Carpenter et al., 1972
333	333	Trawl net	Collignon et al., 2012
335	290	Trawl net	Gewert et al., 2017
335	410	Trawl net	Morét-Ferguson et al., 2010
335	n/a	Trawl net	Bakir et al., 2020
350	350	Trawl net	Isobe et al., 2015
350	355	Trawl net	Sagawa et al., 2018
350	200	Trawl net	Isobe et al., 2017
100, 333, 500	5	Trawl net	Lindeque et al., 2020
330, 780	400	Trawl net	Schmidt et al., 2018
900	n/a	Trawl net	Ryan, 1988
947	200	Trawl net	Colton et al., 1974

effective.

Furthermore, some studies used nets of the standard mesh size, but reported microplastics sizes lower than 300 μm (e.g. Lusher et al., 2015; Naidoo et al., 2015; Isobe et al., 2017). In our study, we filtered surface water samples through a 63- μm filter and also recovered particles smaller than the mesh (100 μm) of the sampling net. In facts, as much as 25 % of the measured surface microplastics were smaller than 100 μm , which suggests that some smaller particles do not pass through the net, but either adhere to the inside of the cod end or the netting itself and remain in the sample.

Although the comparative studies clearly showed that surface trawling with standard mesh size greatly underestimates the concentration of microplastics in the surface waters, it is still the most widely used method and is largely standardised. It can be used to detect patterns and trends, because a consistent method allows the results to be compared. Even the oldest studies of surface microplastics (e.g.

Carpenter et al., 1972) used the standard surface nets and reported results in measurement units comparable to current reports. However, if the aim of the study is to determine the concentrations of microplastics as accurately as possible, Tamminga et al. (2019) suggest a combination of a fine-mesh pump and a standard surface trawling net. The authors found that these two methods, in fact, complement each other, as the large microplastics are mainly collected with a surface net, while the smaller microplastics are more effectively captured by a pump (Fig. 10).

4.3. Microplastics form, colour and polymer types

Surface waters of Vava'u archipelago are predominantly contaminated with small bits of white shredded film (Fig. 7), which we assume originate from the same source. We noticed that some docks in Vava'u are built with white cement bags, or similar heavy-duty big white bags, which are filled with concrete and used as 'bricks' or building blocks. When we examined one of these docks closely, we found the white bags are completely eroded in many areas exposing the cement. The white material likely disintegrates into microplastics from a combination of photodegradation, wave abrasion and bioerosion by crustaceans, mollusks and other grazing and boring marine organisms. We do not have a definite proof that the 'plastic' docks are the source of these microplastics, since we were unable to collect the plastic from the docks to compare it to the white bits from our samples, but it is highly likely. A final examination and confirmation are planned for future work.

Bioerosion and biomechanical degradation of plastic in the marine environment have been reported in several experimental and field studies and are an important, but often overlooked, source of microplastics. More than 40 years ago, Holmström (1975) described 'eating traces' on polyethylene sheets (irregularly shaped holes on the material) collected from 180 to 400 m deep seafloor. The author suggested the marks most likely belong to grazing mollusks, as the sheets were covered in bryozoans and algae, which is also a reasonable explanation as to why the positively buoyant plastic sank. The formation of microplastics by other animals has also been documented; more specifically, by boring crustaceans (Davidson, 2012), fish (Carson, 2013), amphipods (Hodgson et al., 2018) and polychaetes (Jang et al., 2018).

White colour is seldom reported as the most common colour of surface water microplastics in the South Pacific (Young and Adams, 2010; Reisser et al., 2013; Rudduck et al., 2017). The film type of microplastics has not been documented in such prevalence in the South Pacific waters either. Instead, more commonly found are fragments, or broken pieces of hard plastic (Eriksen et al., 2013; Reisser et al., 2013; Rudduck et al., 2017). The pervasiveness of white shredded film in the surface waters is most probably specific to Tonga, but possibly in other areas as well where the plastics, which are not designed for marine environments, are used as construction material in or near the marine environment. Additionally, water exchange in the Vava'u archipelago with the

surrounding ocean is limited, causing the white microplastic bits to accumulate in the surface waters within the archipelago.

Our polymer analysis was done on a random and small subset of surface microplastics to provide an indication of the most common polymers and does not represent the overall composition of all microplastics found. Of the particles analysed, only one was misidentified, indicating highly accurate visual identification of plastic particles. The most common polymer types found were PES, PP and PE. Only two white bits of film were analysed by FTIR spectroscopy and they were PP and PU. The Studies in the Pacific region reported PE (Reisser et al., 2013; Rudduck et al., 2017; Bakir et al., 2020; Dehm et al., 2020; Gardon et al., 2021), PP (Reisser et al., 2013; Rudduck et al., 2017; Bakir et al., 2020; Dehm et al., 2020), PS (Rudduck et al., 2017; Bakir et al., 2020) and PET (Dehm et al., 2020) as the most common polymer types. Apart from PES fibres, PE and PP polymers were also more numerous than others (Fig. 8).

5. Conclusion

The concentration of microplastics in the surface waters of the Vava'u archipelago obtained in our study is high, considering the remote location and the low population nearby. We assume that the most likely reason for this is the mesh size of the sampling nets, which was 200 μm smaller than the standard mesh size (333 μm) commonly used in surface water microplastics research. We would recommend a combination of sampling nets and pumps for a more accurate result.

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

Thomas Maes: Validation, Writing – Review & editing (advising)

Melissa Bowen: Methodology, 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.

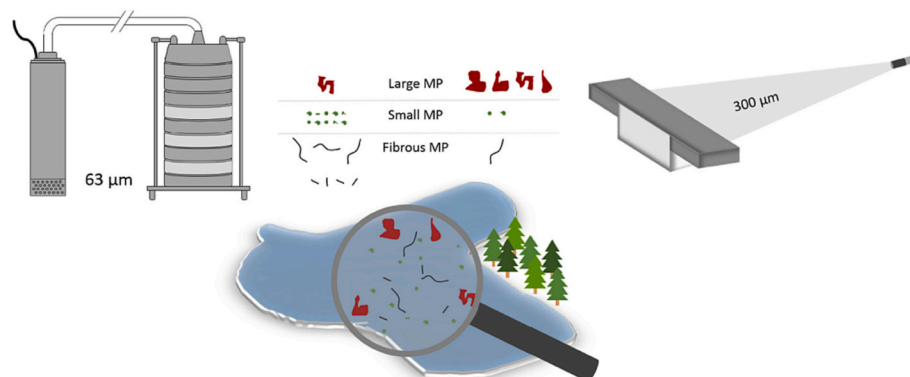


Fig. 10. Compatibility of two methods for sampling floating microplastics, pump system on the left and manta trawl on the right. (Adopted from Tamminga et al. (2019).)

Data availability

Data will be made available on request.

Acknowledgements

This paper was made Open Access with support from GRID-Arendal. We would also like to thank the Secretariat of the Pacific Regional Environment Programme for the financial support. A big thank you goes to Fred Faiva'ilo from the University of South Pacific in Tonga, and Errol Murray, Peter Browne, Richard Taylor and Boyd Taylor from the Institute of Marine Science, New Zealand. This study would not be possible, nor special, without Infinity and Captain Clemens, Sage, Rueben, Rhian and Chloe, Gary, Lindsay, Ayla, Squeegie, Scale, Petri, Pipetta, Epi, Pump and Beaker, Antonia, Kristy and Ella.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2022.114243>.

References

- Alfaro-Núñez, A., Astorga, D., Cáceres-Farías, L., Bastidas, L., Soto Villegas, C., Macay, K., Christensen, J.H., 2021. Microplastic pollution in seawater and marine organisms across the tropical eastern Pacific and Galápagos. *Sci. Rep.* 11, 6424.
- Bakir, A., Desender, M., Wilkinson, T., Van Hoytema, N., Amos, R., Airahui, S., Graham, J., Maes, T., 2020. Occurrence and abundance of meso and microplastics in sediment, surface waters, and marine biota from the South Pacific region. *Mar. Pollut. Bull.* 160, 111572.
- Cai, M., He, H., Liu, M., et al., 2018. Lost but can't be neglected: huge quantities of small microplastics hide in the South China Sea. *Sci. Total Environ.* 633, 1206–1216.
- Carpenter, E.J., Smith Jr., K.L., 1972. Plastics on the Sargasso Sea surface. *Science* 175, 1240–1241.
- Carpenter, E.J., Anderson, S.J., Harvey, G.R., Miklas, H.P., Peck, B.B., 1972. Polystyrene spherules in coastal waters. *Science* 173, 749–750.
- Carson, H.S., 2013. The incidence of plastic ingestion by fishes: from the prey's perspective. *Mar. Pollut. Bull.* 74, 170–174.
- Cincinelli, A., Scopetani, C., Chelazzi, D., et al., 2017. Microplastic in the surface waters of the Ross Sea (Antarctica): occurrence, distribution and characterization by FTIR. *Chemosphere* 175, 391–400.
- Collignon, A., Hecq, J.-H., Glagani, F., Voisin, P., Collard, F., Goffart, A., 2012. Neustonic microplastic and zooplankton in the North Western Mediterranean Sea. *Mar. Pollut. Bull.* 64, 861–864.
- Colton Jr., J.B., Knapp, F.D., Burns, B.R., 1974. Plastic particles in surface waters of the northwestern Atlantic. *Science* 185, 491–497.
- Connors, E.J., 2017. Distribution and biological implications of plastic pollution of the fringing reef of Mo'orea, French Polynesia. *PeerJ* 5, e3733. <https://doi.org/10.7717/peerj.3733>.
- Cózar, A., Echevarria, F., González-Gordillo, J.I., et al., 2014. Plastic Debris in the Open Ocean. *PNAS*. <https://doi.org/10.1073/pnas.1314705111>.
- Cózar, A., Sanz-Martín, M., Martí, E., et al., 2015. Plastic accumulation in the Mediterranean Sea. *PLoS ONE* 10 (4), e0121762. <https://doi.org/10.1371/journal.pone.0121762>.
- Cutroneo, L., Reboa, A., Besio, G., Borgogno, F., Ganesi, L., Canuto, S., et al., 2020. Microplastics in seawater: sampling strategies, laboratory methodologies, and identification techniques applied to port environment. *Environ. Sci. Pollut. Res.* 27, 8938–8952.
- Davidson, T.M., 2012. Boring crustaceans damage polystyrene floats under docsk polluting marine waters with microplastics. *Mar. Pollut. Bull.* 64, 1821–1828.
- Davison, S.M.C., White, M.P., Pahl, S., Taylor, T., Fielding, K., Roberts, B.R., Economou, T., McMeel, O., Kellett, P., Felming, L.E., 2021. Public concern about, and desire for research into, the human health effects of marine plastic pollution: results from a 15-country survey across Europe and Australia. *Glob. Environ. Chang.* 69, 102309.
- Dehm, J., Singh, S., Ferreira, M., Piovano, S., 2020. Microplastics in subsurface coastal waters along the southern coast of Viti Levu in Fiji, South Pacific. *Marine Pollution Bulletin* 156, 111239.
- Desforges, J.-P.W., Galbraith, M., Dangerfield, N., Ross, P.S., 2014. Widespread distribution of microplastics in subsurface seawater in the NE Pacific Ocean. *Mar. Pollut. Bull.* 79, 94–99.
- Eriksen, M., Maximenko, N., Thiel, M., et al., 2013. Plastic pollution in the South Pacific subtropical gyre. *Mar. Pollut. Bull.* 68, 71–76.
- Eriksen, M., Liboiron, M., Kiesslong, T., et al., 2018. Microplastic sampling with the AVANI trawl compared to two neuston trawls in the bay of Bengal and South Pacific. *Environ. Pollut.* 232, 430–439.
- Faure, F., Saini, C., Potter, G., Galgani, F., de Alencastro, L.F., Hagmann, P., 2015. An evaluation of surface micro- and mesoplastic pollution in pelagic ecosystems of the Western Mediterranean Sea. *Environ. Sci. Pollut. Res.* 22, 12190–12197.
- Ferreira, M., Thompson, J., Paris, A., Rohindra, D., Rico, C., 2020. Presence of microplastics in water, sediments and fish species in an urban coastal environment of Fiji, a Pacific small island developing state. *Mar. Pollut. Bull.* 153, 110991.
- Forrest, A.K., Hindell, M., 2018. Ingestion of plastic by fish destined for human consumption in remote South Pacific Islands. *Aust. J. Mar. Ocean Affairs* 10 (2), 81–97.
- Frias, J.P.G.L., Otero, V., Sobral, P., 2014. Evidence of microplastics in samples of zooplankton from Portuguese coastal waters. *Mar. Environ. Res.* 95, 89–95.
- Gallagher, A., Rees, A., Rowe, R., Stevens, J., Wright, P., 2016. Microplastics in the Solent estuarine complex, UK: an initial assessment. *Mar. Pollut. Bull.* 102, 243–249.
- Gardon, T., El Rakwe, M.E., Paul-Pont, I., et al., 2021. Microplastics contamination in pearl-farming lagoons of French Polynesia. *J. Hazard. Mater.* 419, 126396.
- Gewert, B., Ogonowski, M., Barth, A., MacLeod, M., 2017. Abundance and composition of near surface microplastics and plastic debris in the Stockholm archipelago, Baltic Sea. *Mar. Pollut. Bull.* 120, 292–302.
- Gregory, M.R., 1987. Plastics and other seaborne litter on the shores of New Zealand's Subantarctic Islands. *N. Z. Antarctic Record* 7 (3), 32–47.
- Hodgson, D.J., Bréchon, A.L., Thompson, R.C., 2018. Ingestion and fragmentation of plastic carrier bags by the amphipod *Orchestoidea gammarellus*: effects of plastic type and fouling load. *Mar. Pollut. Bull.* 127, 154–159.
- Holmström, A., 1975. Plastic films on the bottom of the Skagerrack. *Nature* 255, 622–623.
- Isobe, A., Uchida, K., Tokai, T., Iwasaki, S., 2015. East Asian seas: a hot spot of pelagic microplastics. *Mar. Pollut. Bull.* 101, 618–623.
- Isobe, A., Uchiyama-Matsumoto, K., Uchida, K., Tokai, T., 2017. Microplastics in the Southern Ocean. *Mar. Pollut. Bull.* 114, 623–626.
- Ivar do Sul, J.A., Costa, M.F., Barletta, M., Cysneiros, F.J.A., 2013. Pelagic microplastics around an archipelago of the Equatorial Atlantic. *Marine Pollution Bulletin* 75, 305–309.
- Ivar do Sul, J.A., Costa, M.F., Fillman, G., 2014. Microplastics in the pelagic environment around oceanic islands of the Western Tropical Atlantic Ocean. *Water, Air & Soil Pollution* 225 (2004), 1–13.
- Jang, M., Shim, W.J., Han, G.M., Song, Y.K., Hong, S.H., 2018. Formation of microplastics by polychaetes (*Marphysa sanguinea*) inhabiting expanded polystyrene marine debris. *Mar. Pollut. Bull.* 131, 365–369.
- Kang, J.-H., Kwon, O.-Y., Shim, W.J., 2015. Potential threat of microplastics to zooplanktivores in the surface waters of the Southern Sea of Korea. *Arch. Environ. Contam. Toxicol.* 69, 340–351.
- Karlsson, T.M., Vethaak, A.D., Almroth, B.C., Ariese, F., van Velzen, M., Hasselöv, M., Leslie, H.A., 2017. Screening for microplastics in sediment, water, marine invertebrates and fish: method development and microplastic accumulation. *Mar. Pollut. Bull.* 122, 403–408.
- Kroon, F., Motti, C., Talbot, S., Sobral, P., Puotinen, M., 2018. A workflow for improving estimates of microplastic contamination in marine waters: a case study from North-Western Australia. *Environ. Pollut.* 238, 26–38.
- Lebreton, L.C.-M., Greer, S.D., Borrero, J.C., 2012. Numerical modelling of floating debris in the world's oceans. *Mar. Pollut. Bull.* 653–661.
- Li, C., Wang, X., Liu, K., Shu, L., Wei, N., Zong, C., Li, D., 2021. Pelagic microplastics in surface water of the eastern Indian Ocean during monsoon transition period: abundance, distribution, and characteristics. *Sci. Total Environ.* 755, 142629.
- Lima, A.R.A., Costa, M.F., Barletta, M., 2014. Distribution patterns of microplastics within the plankton of a tropical estuary. *Environ. Res.* 132, 146–155.
- Lindeque, P.K., Cole, M., Coppock, R.L., et al., 2020. Are we underestimating microplastic abundance in the marine environment? A comparison of microplastic capture with nets of different mesh-size. *Environ. Pollut.* 265, 114721.
- Lusher, A.L., Tirelli, V., O'Connor, I., Officer, R., 2015. Microplastics in Arctic polar waters: the first reported values of particles in surface and sub-surface samples. *Sci. Rep.* 5, 14947. <https://doi.org/10.1038/srep14947>.
- Lusher, A.L., Burke, A., O'Connor, I., Officer, R., 2014. Microplastic pollution in the Northeast Atlantic Ocean: validated and opportunistic sampling. *Mar. Pollut. Bull.* 88, 325–333.
- Maes, T., Van der Muelen, M.D., Devriese, L.I., et al., 2017. Microplastics baseline surveys at the water surface and in sediments of the north-east Atlantic. *Front. Mar. Sci.* 4, 1–13.
- Markic, A., Niemand, C., Bridson, J.H., Mazouni-Gaertner, N., Gaertner, J., Eriksen, M., Bowen, M., 2018. Double trouble in the South Pacific subtropical gyre: increased plastic ingestion by fish in the oceanic accumulation zone. *Mar. Pollut. Bull.* 136, 547–564.
- Menéndez-Pedriz, A., Jaumot, J., 2020. Interaction of environmental pollutants with microplastics: a critical review of sorption factors, bioaccumulation and ecotoxicological effects. *Toxics* 8, 40.
- Milton, J.S., 1999. In: *Statistical Methods in the Biological and Health Sciences*. The McGraw-Hill Companies Inc, US, pp. 1–588.
- Moore, C.J., Moore, S.L., Leecaster, M.K., Weisberg, S.B., 2001. A comparison of plastic and plankton in the North Pacific Central Gyre. *Mar. Pollut. Bull.* 42 (12), 1297–1300.
- Moore, C.J., Moore, S.L., Weisberg, S.B., Lattin, G.L., Zellers, A.F., 2002. A comparison of neustonic plastic and zooplankton abundance in southern California's coastal waters. *Mar. Pollut. Bull.* 44, 1035–1038.
- Moré-Ferguson, S., Law, K.L., Proskurowski, G., Murphy, E.K., Peacock, E.E., Reddy, C. M., 2010. The size, mass, and composition of plastic debris in the western North Atlantic Ocean. *Mar. Pollut. Bull.* 60, 1873–1878.
- Naidoo, T., Glassom, D., Smit, A.J., 2015. Plastic pollution in five urban estuaries of KwaZulu-Natal, South Africa. *Mar. Pollut. Bull.* 101, 473–480.
- Nel, H.A., Froneman, P.W., 2015. A quantitative analysis of microplastic pollution along the south-eastern coastline of South Africa. *Mar. Pollut. Bull.* 101, 274–279.

- Panti, C., Giannetti, M., Bani, M., Rubegni, F., Minutoli, R., Fossi, M.C., 2015. Occurrence, relative abundance and spatial distribution of microplastics and zooplankton NW of Sardinia in the Pelagos sanctuary protected area, Mediterranean Sea. *Environ. Chem.* 12, 618–626.
- PlasticsEurope, 2021. An Analysis of European Plastics Production, Demand and Waste Data. PlasticsEurope, Belgium.
- Prata, J.C., da Costa, J.P., Duarte, A.C., Rocha-Santos, T., 2019. Methods for sampling and detection of microplastics in water and sediment: a critical review. *Trends Anal. Chem.* 110, 150–159.
- Preston-Whyte, F., Silburn, B., Meakins, B., et al., 2021. Meso- and microplastics monitoring in harbour environments: A case study for the Port of Durban, South Africa. *Marine Pollution Bulletin* 163, 111948.
- Rawlins, B.G., Ferguson, A.J., Chilton, P.J., Arthurton, R.S., Rees, J.G., Baldock, J.W., 1998. Review of agricultural pollution in the Caribbean with particular emphasis on Small Island developing states. *Mar. Pollut. Bull.* 36 (9), 658–668.
- Reisser, J., Shaw, J., Wilcox, C., Hardesty, B.D., Proietti, M., Thums, M., Pattiaratchi, C., 2013. Marine plastic pollution in waters around Australia: characteristics, concentrations and pathways. *PLoS ONE* 8 (11), e80466.
- Rochman, C.M., 2015. The complex mixture, fate and toxicity of chemicals associated with plastic debris in the marine environment. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), *Marine Anthropogenic Litter*. Springer Open, Heidelberg, pp. 117–140.
- Rudduck, O., Lavers, J.L., Fischer, A.M., Stuckenbrock, S., Sharp, P.B., Banati, R.B., 2017. Inter-annual variation in the density of anthropogenic debris in the Tasman Sea. *Mar. Pollut. Bull.* 124, 51–55.
- Ryan, P.G., 1988. The characteristics and distribution of plastic particles at the sea-surface off the Southwestern Cape Province, South Africa. *Marine Environmental Research* 25 (4), 249.
- Sagawa, N., Kawaai, K., Hinata, H., 2018. Abundance and size of microplastics in a coastal sea: comparison among bottom sediment, beach sediment, and surface water. *Mar. Pollut. Bull.* 133, 532–542.
- Sareer, A., 2017. Protecting Small Island developing states from pollution and the effects of climate change. *UN Chron.* 1 & 2, 17–18.
- Schmidt, N., Thibault, D., Galgani, F., Paluselli, A., Sempéré, R., 2018. Occurrence of microplastics in surface waters of the Gulf of lion (NW Mediterranean Sea). *Prog. Oceanogr.* 163, 214–220.
- Song, Y.K., Hong, S.E., Jang, M., Kang, J.-H., Kwon, O.Y., Han, G.M., Shim, W.J., 2014. Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. *Environ. Sci. Technol.* 48, 9014–9021.
- Stock, F., Kochleus, C., Bansch-Baltruschat, B., Brennholt, N., Reifferscheid, G., 2019. Sampling techniques and preparation methods for microplastic analyses in the aquatic environment - a review. *Trends Anal. Chem.* 113, 84–92.
- Tamminga, M., Hengstmann, E., Fischer, E.K., 2018. Microplastic analysis in the south funen archipelago, Baltic Sea, implementing manta trawling and bulk sampling. *Mar. Pollut. Bull.* 128, 601–608.
- Tamminga, M., Stoewer, S.-C., Fischer, E.K., 2019. On the representativeness of pump water samples versus manta sampling in microplastic analysis. *Environ. Pollut.* <https://doi.org/10.1016/j.envpol.2019.112970>.
- Uchida, K., Hagita, R., Hayashi, T., Tokai, T., 2016. Distribution of small plastic fragments floating in the western Pacific Ocean from 2000 to 2001. *Fish. Sci.* 82, 969–974.
- Van Cauwenberghe, L., Janssen, C.R., 2014. Microplastics in bivalves cultured for human consumption. *Environ. Pollut.* 193, 65–70.
- Yee, M.S., Hii, L., Looi, C.K., Lim, W., Wong, S., Kok, W., Tan, B., Wong, C., Leong, C., 2021. Impact of microplastics and nanoplastics on human health. *Nanomaterials* 11, 496.
- Young, M., Adams, N.J., 2010. Plastic debris and seabird presence in the Hauraki Gulf, New Zealand. *N. Z. J. Mar. Freshw. Res.* 44 (3), 167–175.
- Zhang, W., Zhang, S., Wang, J., et al., 2017. Microplastic pollution in the surface waters of the Bohai Sea, China. *Environ. Pollut.* 231, 541–548.
- Zhang, W., Zhang, S., Zhao, Q., Qu, L., Ma, D., Wang, J., 2020. Spatio-temporal distribution of plastic and microplastic debris in the surface water of the Bohai Sea, China. *Marine Pollution Bulletin* 158, 111343.
- Zhao, S., Zhu, L., Wang, T., Li, D., 2014. Suspended microplastics in the surface water of Yangtze estuary system, China: first observations on occurrence, distribution. *Mar. Pollut. Bull.* 86, 532–568.