Faculty of Science and Engineering

School of Geography, Earth and Environmental Sciences

2023-07

# Occurrence and characteristics of fibreglass-reinforced plastics and microplastics on a beach impacted by abandoned fishing boats: A case study from Chellanam, India

## Lekshmi, NM

https://pearl.plymouth.ac.uk/handle/10026.1/21165

10.1016/j.marpolbul.2023.114980 Marine Pollution Bulletin Elsevier BV

All content in PEARL is protected by copyright law. Author manuscripts are made available in accordance with publisher policies. Please cite only the published version using the details provided on the item record or document. In the absence of an open licence (e.g. Creative Commons), permissions for further reuse of content should be sought from the publisher or author.

1	Occurrence and	characteristics (	of fibreglass-re	einforced pl	lastics and
---	----------------	-------------------	------------------	--------------	-------------

- 2 microplastics on a beach impacted by abandoned fishing boats: A
- **3** case study from Chellanam, India
- 4
- N. Manju Lekshmi<sup>1</sup>, Sreejith S Kumar<sup>1</sup>, P. Muhamed Ashraf<sup>1</sup>. S.P. Nehala<sup>1</sup> and Leela Edwin<sup>1</sup>,
   Andrew Turner<sup>2</sup>
- ICAR-Central Institute of Fisheries Technology, P. O. Matsyapuri, Willingdon Island,
   Cochin 682029, India
- 9 2) School of Geography, Earth and Environmental Sciences, University of Plymouth,
  10 Plymouth PL4 8AA, UK
- 11 \*Correspondence: manjulekshmi.n@icar.gov.in / manjuaem@gmail.com (Manju Lekshmi N.)
- 12
- 13 https://doi.org/10.1016/j.marpolbul.2023.114980
- 14
- 15 Accepted 20 April 2023

## 16 Abstract

Plastics and microplastics have been quantified and characterised at boat disposal sites and 17 along the high-water line (HWL) of a fish landing centre beach in Chellanam, India. Fibreglass-18 reinforced plastic (FRP) made a greater contribution to the plastic pool at the disposal sites (~ 19 4.5  $n \text{ m}^{-2}$  and 18 g m<sup>-2</sup>) than at the HWL (~ 0.25  $n \text{ m}^{-2}$  and < 1 g m<sup>-2</sup>) and was an abundant 20 component of the microplastic pool at the former. Infrared analysis of microplastic-sized FRPs 21 revealed various resins (e.g., alkyd, polyester, epoxy), while X-ray fluorescence analysis of 22 the painted surfaces of meso-sized FRPs returned variable concentrations of copper and lead. 23 24 Concentrations of Pb were high enough to contaminate sand up to ~400 mg kg<sup>-1</sup>. The relatively 25 high density of FRP and its association with glass fibres and metal-bearing paints results in particles with potentially very different fates and toxicities to more "conventional" (non-26 27 composite) thermoplastics.

28

## 29

## 30 Keywords

- 31 Debris; fragments; paint; copper; lead; disposal
- 32

## 33 Highlights

- 34 Little information exists on fibreglass-reinforced plastic (FRP) as marine litter
- 35 Different sizes of FRP were found on a beach impacted by fishing boat abandonment
- 36 FRP can make an important contribution to the microplastic pool
- 37 Lead in painted surfaces of FRP are sufficient to significantly contaminate sand
- 38 FRP requires further study because of its association with glass fibres and metals

## 39 1. Introduction

40 Plastic is a versatile, lightweight, chemically stable and inexpensive material with multiple

41 uses across a wide range of sectors. Because of these properties, however, and despite the

42 recyclability of many polymers, the use of plastics, and in particular in the consumer sector,

43 results in the generation of large quantities of poorly-degradable waste. Inadequate

44 management of this waste then leads to contamination of the environment, with the marine

45 setting an ultimate receptor of much plastic (Auta et al., 2017; Woods et al., 2021).

46 In the marine environment, the focus of most research has been on plastic litter along

47 shorelines, and microplastics (with sizes < 5 mm) within intertidal sediments and suspended

48 in the water column. Here, most commonly documented are thermoplastics like polyethene,

49 polypropylene, polyethylene terephthalate and polystyrene (Erni-Cassola et al., 2019).

50 Interest in these polymers is driven by their ubiquity in consumer goods and, for

51 microplastics, constraints on plastic separation from sediments by flotation. The latter

52 normally involves separation of microplastics from other particulate matter in a saturated salt

53 solution whose density typically ranges from 1.2 to 1.8 g cm<sup>-3</sup> (Mattsson et al., 2022). As a

54 consequence of this approach, however, composite materials whose densities exceed this

55 range, including fibreglass reinforced plastic (FRP), are overlooked.

FRP consists of a polymeric matrix or binding agent, such as epoxy or polyester thermoset, 56 57 that is reinforced with filaments or fibres of textile-grade glass to create a relatively strong and elastic composite. Because of its low cost, low weight and resistance to corrosion, FRP 58 has extensive usage in components and structures in the maritime sector. Of particular 59 importance here is the manufacture of small boats (e.g., fishing boats and recreational craft), 60 with about 80% of vessel hulls up to 20 m in length constructed of FRP (Rubino et al., 2020). 61 In India, FRP is used to construct boat hulls or as a sheathing material over a wooden 62 63 substrate. Due to the high costs involved in their proper disposal and difficulties in recycling thermosetting composites, however, end-of-life boats constructed of or sheathed by FRP are 64

often abandoned in the coastal zone (Eklund et al., 2013; IMO, 2019).

Despite the importance of FRP as a material and contaminant, it has received little attention
in the marine environmental literature, with reports limited to its detection in surface trawls
(Song et al., 2014; Higgins and Turner, 2023) and the potential toxicity of ground composites
to two aquatic organisms (Ciocan et al., 2020). In this study, therefore, we quantified the
occurrence within different size fractions of plastic litter on a sandy beach impacted by the

71 FRP-sheathed boats used in the artisanal fishing industry of Chellanam (India). Here, the

52 beach is used as a landing stage and boats are stored, maintained and abandoned without

73 any regulation or guidelines (Lekshmi et al., 2023). FRP particles are characterised

- 74 microscopically and by infrared spectroscopy, and the content of potentially toxic metals on
- 75 any painted surfaces is quantified by X-ray fluorescence spectrometry. Local metal
- contamination by painted FRP is also evaluated by chemical analysis of beach sand.
- 77

## 79 2. Materials and methods

## 80 2.1. Study site and boat abandonment

Chellanam is one of the major small-scale fish landing centres in the Ernakulum district of Kerala. Sampling was undertaken at the landing stage during November 2021 (Figure 1). Most fishing boats operating from Chellanam are motorised and below 14 m in length and are FRPsheathed over plywood/wood. The life span of these boats is up to ten years but may be significantly shortened by bad weather and rough seas. With no formal decommissioning or disposal process, end-of-life boats are commonly abandoned above the high water line (HWL) on local beaches (Figure 2).

88

- 89 Figure 1: The location of Chellanam in India and an aerial view of the landing centre.
- 90 Sampling was undertaen along the HWL from (H1 to H10) and at three disposal sites (D1 to
- 91 D3).





With aid of Google maps, the HWL was divided into 5 m sections and ten sections were
randomly selected according to Lee et al. (2015) (and shown in Figure 1). Just after high water,
these sections were visited and used to define 5 m x 5 m quadrats, centred on the HWL (Opfer

131 et al., 2012). Plastic visible at the surface was manually collected from each quadrat and

classified as FRP (relatively brittle and flat fragments, often painted on one side and with visible 132 fibres and remains of a wooden substrate on the other side), fishing waste (including 133 polyurethane foam used as an insulating material, netting, floats and sinkers) and other (e.g., 134 135 bottles and bottle tops, food packaging, shoes, toys and other fragments). A 0.5 m x 0.5 m quadrat was then defined towards the centre of each main quadrat and sand to a depth of 136 about 2 cm was collected with a stainless-steel trowel and stored in a stainless-steel container 137 in order to determine microplastics and metals. At each disposal site, three 5 m x 5 m quadrats 138 were sampled for plastic debris before three 0.5 m x 0.5 m quadrats were defined within each 139 140 larger quadrat and sampled for sand as above.

141

## 142 2.3. Sample processing

In the laboratory, classes of plastic were counted and weighed and FRP debris was further 143 classified by size as mega-debris: >10 cm; macro-debris: 2 - 10 cm; and meso-debris: 5 - 20 144 145 mm (Jayasiri et al., 2013). About 1 kg of sand from each small quadrat along the HWL (n = 10) and within the disposal sites (n = 9) was weighed out on a Sartorius electronic balance 146 147 before the contents were passed through a 5-mm stainless steel sieve. Material passing through the sieve was dried at 40 °C for 24 h before being subject to density separation. Thus, 148 30 g of material was added to a 100 mL solution of saturated ZnCl<sub>2</sub> (CDH Analytical Grade in 149 150 Milli-Q water) of density 1.6 to 1.7 g cm<sup>-3</sup> in a glass beaker and the contents were agitated with a magnetic stirrer for 20 min before being allowed to settle for 2 h. Supernatants were 151 filtered through individual Whatman 41 filter papers (20 µm pore size) before retained particles 152 153 were passed through a sequence of stainless steel sieves (mesh sizes 300  $\mu$ m and 63  $\mu$ m) to separate coarse (300  $\mu$ m to 5 mm), medium (63  $\mu$ m to 300  $\mu$ m) and fine (20  $\mu$ m to 63  $\mu$ m) 154 155 material and microplastics. Fractionated particles were transferred to watch glasses and plastic particles identified, counted and categorised (by shape, size and colour) under a LEICA 156 157 MZ16A stereomicroscope at up to 230 X magnification.

## 158 2.4. Microplastic identification by FTIR

The polymer content of selected, fractionated microplastics of various shapes, colours and sizes from the HWL (n = 13) and disposal sites (n = 33) and ten fragments cut or plucked from abandoned boat hulls was determined by attenuated total reflectance Fourier Transform infrared (ATR-FTIR) spectroscopy. Specifically, small offcuts with no visible extraneous contamination or paint were placed on the diamond compression cell of a Thermo Scientific ATR sampling accessory and analysed using a Thermo Scientific Nicolet iS 10 spectrometer. Spectra were acquired between 650 and 4000 cm<sup>-1</sup>, with 50 scans per sample, and were compared with a Hummel polymer library embedded in Omnic software.

167

## 168 2.5. Sample digestion and copper and lead analysis

From six sand samples along the HTL and all samples from the disposal sites (n = 9), about 169 170 10 g of < 5 mm material was ground in a pestle and mortar. In triplicate, 1 g portions were 171 weighed into a series of acid-cleaned, 50 mL Pyrex beakers to which 8 mL of aqua-regia (3:1 172 HCI:HNO3: Merck AR) was added. The contents of each beaker were covered with a watch 173 glass and gently boiled on a hotplate for 1 h before the cooled digests were vacuum-filtered 174 through 0.45  $\mu m$  Whatman Millipore filters. Filtrates were transferred to volumetric flasks and made up to 50 mL with Milli-Q water pending analysis. Triplicate controls were subject to the 175 same protocol but in the absence of sand. 176

177 Copper and Pb, as indicators of contamination from boat paints, were analysed in the digests

by inductively coupled plasma-optical emission spectroscopy (ICP-OES) using a Perkin Elmer

179 Optima 2000 DV. The instrument was calibrated with blanks and standards of 0.1, 0.2, 0.5

and 1 mg  $L^{-1}$  prepared from dilutions of a NIST multi-element standard.

The Cu and Pb content of the painted surfaces of a range of randomly selected mesofragments of FRP that had been retrieved from the HWL (n = 12) and disposal sites (n = 17) were determined by portable X-ray fluorescence (XRF) spectrometry. Specifically, we used a Niton XL3t He GOLDD+ spectrometer housed in a laboratory test and operated in a plastics mode (counting time 30 s) according to procedures detailed elsewhere (Turner and Solman, 2016). Precise limits of detection varied depending on sample thickness but were typically around 30 mg kg<sup>-1</sup> for Cu and 10 mg kg<sup>-1</sup> for Pb.

188

#### 190 3. Results and Discussion

#### 191 3.1. Abandoned boats and plastic debris

Across the entire length of the landing centre, we observed 27 abandoned boats, of which 21 192 193 were within disposal sites and six were encountered around the HWL. All abandoned boats were constructed of plywood sheathed with approximately 6 mm of FRP, and ranged in length 194 from 6 m to 8 m. 195

196

197 Table 1: Summary of plastic debris (on a number basis and by size category for FRP) recovered from the quadrats along the HWL and within the disposal sites. Also shown are the 198 199 average numbers of plastic per m<sup>2</sup> based on totals for each category divided by the total area of quadrats surveyed. 200

201

202

	FRP			_	
	mega	macro	meso	fishing	other
HWL					
H1	0	0	7	2	5
H2	0	2	0	7	1
H3	1	12	5	5	5
H4	0	0	0	7	7
H5	1	3	0	3	2
H6	0	0	12	3	3
H7	1	0	3	1	4
H8	0	0	7	1	3
H9	0	2	4	6	3
H10	0	1	0	3	2
total	3	20	38	38	35
<i>n</i> m <sup>-2</sup>	0.01	0.08	0.15	0.15	0.14
disposal sites					
D1	8	25	225	37	20
D2	4	101	257	37	16
D3	20	92	289	12	36
total	32	218	771	86	72
<i>n</i> m <sup>-2</sup>	0.14	0.97	3.43	0.38	0.32

203	Table 1 summarises the number of pieces of plastic debris retrieved from the quadrats along
204	the HWL (H1 to H10) and within the three disposal sites (D1, D2 and D3). Along the HWL, 134
205	pieces of plastic debris were recovered, of which about 46% were visually identified as FRP.
206	At the disposal sites 1179 pieces of plastic debris were recovered of which about 87% were

FRP. When normalised on an area basis, debris in each category, and in particular for the different size classes of FRP, was greater at the disposal sites than along the HWL.

Table 2 summarises the plastic debris data on a mass basis. Along the HWL, over 2 kg of plastic was retrieved, with fishing waste contributing more than 70% and FRP contributing about 5%. By comparison, at the disposal sites more than 11 kg was retrieved, with fishing

212 waste contributing 45% and FRP contributing about 36%.

213

Table 2: Summary of plastic debris (on a mass basis and by size category for FRP) recovered

from the quadrats along the HWL and within the disposal sites. Also shown are the masses of

216 plastic per  $m^2$  for each category.

217

		FRP			
	mega	macro	meso	fishing	other
HWL					
g	60	30	27	1530	505
g m <sup>-2</sup>	0.24	0.12	0.11	6.12	2.02
disposal sites					
g	2160	1305	480	4925	2180
g m <sup>-2</sup>	9.60	5.80	2.13	21.89	9.69

218 219

#### 221 3.2. Microplastic abundance and visual characteristics



Figure 3: Examples of FRP and other microplastic fragments observed under the 223

- 224 microscope. (a) and (b) were taken directly from abandoned boats and (c) to (f) were
- 225 sampled from disposal sites.
- 226

222

Microscopic images of six microplastic fragments retrieved in the study are shown in Figure 227 3. Fragments were either (i) relatively stiff and flat irregular shapes, with (often) one or more 228 229 straight edge and straight fibres in parallel bundles and usually with one painted surface (Figures 3a, 3b and 3c), or (ii) irregular shapes with angular or curved edges and no 230 231 evidence of fibrous additives or painted surfaces (Figures 3d, 3e and 3f). Microplastic fibres (not illustrated) consisted of single, twisted threads or filaments that were sometimes coiled 232 or single or multiple threads that appeared to be intertwined, while microplastic pellets (not 233 234 illustrated) were distinctly rounded but with occasional irregularities (e.g., pits, protrusions, 235 overlapping layers). Table 3 shows the number of MPs identified in the quadrats at each location, normalised to a 236

sediment mass of 30 g, along with their distributions by size and shape. Along the HWL, 237 238 numbers range from 34 to 155 with a median of 82. At the disposal sites, numbers range 239 from 54 to 253 with a median of 127. Overall, and at both the HWL and disposal sites, numbers in the coarse fraction ranged from 21 to 52, while in the medium and fine fractions 240 241 numbers were more heterogenous between locations, ranging from 2 to 182 and from 5 to 72, respectively. Fragments were the most abundant shape in the study and pellets were the 242

least abundant. 243

- 244 On a size basis, however, and as illustrated in Figure 4, the percentage of fragments
- 245 decreased with decreasing size at the HWL (from about 72% in coarse to about 26% in fine)
- 246 whereas pellets (about 50%) and fibres (about 65%) were most important in the medium and
- 247 fine fractions, respectively. By contrast, at the disposal sites, fragments dominated the
- 248 medium size fraction (nearly 90%) whereas pellets and fibres were most important in the
- 249 fines.
- 250 Figure 5 compares the percentages of each microplastic shape by colour along the HWL and
- at the disposal sites. More abundant along the HWL, and lying below unit slope, are all red
- 252 microplastics, blue fragments and fibres, yellow and green pellets and fragments, and brown
- 253 fibres. More abundant at the disposal sites, and above the line of unit slope, are white
- 254 microplastics, blue pellets, yellow fibres and brown fragments and pellets.
- 255 In summary, therefore, there were no clear differences in microplastic numbers among the
- sites and location types, but differences were more evident in size, shape and colourdistribution.
- 258



Figure 4: Percentage distribution of microplastics by size and shape across the HWL and atthe disposal sites.

- 261
- 262
- 263

264 Table 3: Number of MPs identified in the quadrats at each location and categorised by size

and shape. Note that MPs along the HWL are per 30 g of sediment and those at the disposal

sites are totals for three sites each but normalised to a mass of 30 g.

267

		coarse				medium				fine			
location	fibres f	fragments	beads	sum	fibres	ragment	s beads	sum	fibresf	ragment	s beads	sum	total
HWL													
H1	0	42	10	52	4	11	16	31	31	15	5	51	134
H2	13	17	0	30	1	5	7	13	20	7	1	28	71
H3	4	16	1	21	2	8	10	20	24	14	5	43	84
H4	9	30	0	39	4	18	22	44	56	15	1	72	155
H5	8	36	2	46	0	11	11	22	23	4	0	27	95
H6	9	19	3	31	0	4	4	8	11	12	7	30	69
H7	3	18	0	21	1	0	1	2	4	6	1	11	34
H8	0	18	3	21	4	8	12	24	25	4	3	32	77
H9	14	10	4	28	5	5	10	20	20	7	5	32	80
H10	11	41	0	52	2	2	6	10	14	7	0	21	83
disposal	sites												
D1	13	18	2	33	3	7	4	15	3	1	1	5	54
D2	18	25	1	44	4	175	3	182	4	8	14	26	253
D3	31	13	5	49	13	22	2	37	25	9	8	42	127



Figure 4: Scatter plot of the percentages of fibres (squares), fragments (diamonds) and

pellets (circles) by colour at the disposal sites and along the HWL (note that transparent is

represented by grey). The line represents unit slope.

## 275 3.3. Polymeric composition of microplastics

276 Results of the FTIR analysis of selected samples (n = 56) are summarised in Table 4. The ten fragments obtained from abandoned boats were identified as resinous, with alkyd resin 277 most abundant and polyester-, epoxy-, polystyrene- and piperylene-based resins also 278 279 present. In seven cases, fragments contained glass fibres that were visible under the microscope (e.g., Figure 3) and that were most evident in the FTIR spectra as absorbance 280 peaks in the region 1000-1200 cm<sup>-1</sup> through asymmetric stretches associated with silicate 281 282 glass (Hopkinson et al., 2021). At the disposal sites, there was a combination of resin-based 283 fragments with or without visible glass fibres, and non-resinous fragments, fibres and beads that were dominated by polyethylene and polypropylene. Along the HWL, resinous materials 284 were not identified and polyethylene was the most abundant polymer among the various 285 286 fragments and fibres tested.

287 Table 4: Distribution by polymer type of samples positively identified from abandoned boats,

disposal sites and HWL (*n* represents the total number of samples analysed in each

289 category). Note that polyester includes polyethylene terephthalate.

		resin	polyolefin	polyester	polyamide
	boats ( <i>n</i> = 10)	10			
	disposal sites (n = 33)	10	18	1	
290	HWL ( <i>n</i> = 13)		9	2	1

291

## 292 3.4. Copper and lead in sand and mesoplastic FRP

The concentrations of Cu and Pb in sand samples are shown in Table 5. Mean 293 concentrations of Cu range from 1.1 mg kg<sup>-1</sup> to 11.0 mg kg<sup>-1</sup>, with relative standard 294 deviations for replicate analyses ranging from about 5% to 35%. Median concentrations are 295 1.5 mg kg<sup>-1</sup> and 3.6 mg kg<sup>-1</sup> at the HWL and disposal sites, respectively, with a Mann-296 297 Whitney U-test (Minitab,  $\sqrt{19}$ ) revealing a significantly higher median (p < 0.05) at the latter. 298 Mean concentrations of Pb are greater than Cu in each sample and are more variable (ranging from about 3 mg kg<sup>-1</sup> to 400 mg kg<sup>-1</sup>). As with Cu, median concentrations are 299 significantly higher at the disposal site than the HWL. 300

301

302	Concentrations of Cu and Pb in the mesoplastic FRP fragments analysed by XRF are
303	summarised in Table 6. Where detected, Cu concentrations are greater than those found

in

304 sand by up to two orders of magnitude but, according to a Mann-Whitney U-test, median

- 305 concentrations are not significantly different between (p < 0.05) the HWL and disposal sites.
- 306 Lead was detected in fewer mesoplastics but median concentrations are significantly higher
- 307 than Cu in both the HWL and disposal sites.

308

## 310 Table 5: Concentrations of Cu and Pb (in mg kg<sup>-1</sup>) in samples of sand from the HWL and

311 disposal sites. Note that here, individual data are shown for each quadrat (a, b, c) of the

disposal sites. Errors are one standard deviation about the mean of three determinations.

	Cu	Pb
HWL		
H2	1.1 <u>+</u> 0.3	7.8 <u>+</u> 1.0
H3	1.6 <u>+</u> 0.5	2.8 <u>+</u> 0.6
H4	1.5 <u>+</u> 0.2	3.9 <u>+</u> 0.2
H6	2.9 <u>+</u> 0.2	6.3 <u>+</u> 0.1
H9	1.5 <u>+</u> 0.1	3.4 <u>+</u> 0.6
H10	7.5 <u>+</u> 1.1	26.5 <u>+</u> 2.0
median	1.5	5.1
disposal si	tes	
D1a	3.1 <u>+</u> 0.3	12.8 <u>+</u> 2.5
D1b	6.2 <u>+</u> 0.5	8.0 <u>+</u> 0.3
D1c	11.0 <u>+</u> 1.1	50.3 <u>+</u> 0.8
D2a	3.3 <u>+</u> 0.5	5.4 <u>+</u> 1.0
D2b	3.5 <u>+</u> 1.3	10.5 <u>+</u> 1.2
D2c	3.7 <u>+</u> 0.7	58.3 <u>+</u> 3.5
D3a	6.2 <u>+</u> 1.0	391.8 <u>+</u> 35.0
D3b	3.5 <u>+</u> 0.4	9.7 <u>+</u> 0.3
D3c	2.8 <u>+</u> 0.3	7.8 <u>+</u> 0.1
median	3.6	30.4

313 314

317

315	Table 6: Summary	/ statistics for the	e concentrations	of Cu and	Pb (in	mg kg <sup>-1</sup> ) in	the
-----	------------------	----------------------	------------------	-----------	--------	--------------------------	-----

316 mesoplastics sampled from the HWL and disposal sites.

	Cu	Pb
HWL (n = 10)		
no. detected	8	5
median	304.5	752.2
min	35.7	82.6
max	1047.0	1858.0
disposal sites ( $n = 17$ )		
no. detected	16	13
median	118.6	345.3
min	17.7	15.0
max	985.2	33606.6

## 318 4. Discussion

The maintenance and abandonment of fishing boats on the beach at Chellanam is associated with high levels of various forms of visible (> 5 mm) plastic waste, both on a number basis and a mass basis and in particular for FRP, compared with the neighbouring HWL. The waste may be generated by the decay and weathering of the boats themselves, or the disposal of other, more general waste at locations of abandonment. As pointed out by Turner and Rees (2016), the presence of discarded boats may be perceived as justification for the deliberate dumping of other forms of litter.

Microplastics (< 5 mm) were dominated by similar quantities of fragments at both the 326 327 disposal sites and along the HWL, but differences were evident between the colour and size 328 distributions at the different location types. Analysis of a selection of microplastics by FTIR 329 revealed a variety of polymers at the disposal sites, including various resins used in FRP, but a dominance of polyolefins at the HWL. This discrepancy maybe related to the lower 330 densities of polyethylene and polypropylene than FRP. Thus, while the density of polyolefins 331 332 is below 1 g cm<sup>-3</sup>, the density of plastic resins reinforced by glass fibres (often visible under 333 the microscope and confirmed by FTIR) is typically between 1.25 to 2.5 g cm<sup>-3</sup> (Abbood et al., 2021), with slight modifications possible when the surface is painted. The lower density 334 plastics may be more readily transported from the disposal sites to the HWL and, with 335 buoyant plastics derived from offshore, are subject to redistribution, recirculation and 336 accumulation in the intertidal zone (Graca et al., 2017). Conversely, higher density FRP is 337 less readily transported to the HWL (Ciocan et al., 2020), and any material reaching this area 338 is more likely to be subject to transportation with subsurface offshore currents and sinking 339 and burial. 340

Analysis of the painted surfaces of larger fragments of meso-sized FRP from the disposal 341 342 sites and, where available, the HWL, indicate variable concentrations of Cu, but at levels insufficient to act as an antifoulant (Singh and Turner, 2009). More significant from an 343 environmental perspective, however, are variable but higher (median) concentrations of Pb. 344 Quantitatively, our results are similar to those reported by Hopkinson et al. (2021) for 14 345 samples of FRP sourced from various breakers yards in southern England (Cu ~ 150 to 1600 346 347 mg kg<sup>-1</sup>; Pb ~ 95 to 10,400 mg kg<sup>-1</sup>). This suggests that, despite extensive restrictions on the Pb content of consumer paints, leaded paint is still used or has been applied relatively 348 recently on FRP more generally. With regard to India, a maximum Pb concentration in 349 consumer paints of 90 mg kg<sup>-1</sup> has been required since 2017. However, we note that in a 350 chemical assessment of Indian paints undertaken since the restrictions, Arora et al. (2018) 351 found that paints with very high levels of Pb are still widely sold across the country and that 352

consumer awareness of the problem is extremely low. In the present setting at least, a 353 354 consequence of the use and removal of leaded paint is that beach sand is heterogeneously contamination by the metal, both within and between disposal sites. Contamination may 355 356 arise directly from the presence of Pb-rich paint-FRP particulates in the sand, or indirectly via the dissolution of particulate Pb and its adsorption to neighbouring sand grains. Significantly, 357 the Pb concentration in one sample at disposal site D3 exceeded the CCME marine 358 sediment quality guideline for the protection of aquatic life (112 mg kg<sup>-1</sup>; Canadian Council of 359 360 Ministers of the Environment, 2012).

There is very little information on the occurrence, fate and effects of FRP in the marine 361 science literature, and especially regarding microplastics in sediment, from which to draw 362 comparisons with the present study. The main reason for this is, likely, that conventional 363 364 means of isolating microplastics from sediments involves flotation in a saturated salt solution, 365 and common solutions employed, like NaCl, Nal and ZnCl<sub>2</sub> (Cutroneo et al., 2021), have a density range (~1.2 to of 1.8 cm<sup>-3</sup>) that would preclude many glass-reinforced resins. 366 Nevertheless, given the importance of FRP in the leisure and commercial boating sectors 367 (Ciocan et al., 2020) and with regard to other maritime structures (Summerscales et al., 368 369 2016), and the documentation of other boat-derived debris (mainly paint particles) in the 370 vicinity of boat maintenance facilities or near to abandoned boats (Singh and Turner, 2009; Eklund et al., 2014; Rees et al., 2014; Turner and Rees, 2016; Soroldoni et al., 2018), FRP 371 contamination is predicted to be a more general problem. 372 373 Unlike sediment, microscopic particles of FRP or the common resins used therein have been

reported at the surface of seawater. Specifically, Song et al. (2014) identified polymers originating from paints and FRP used on ships off the southern coast of Korea, while Higgins and Turner (2023) found particles of polyester and epoxy resins in coastal waters around Plymouth, UK. It was suggested that particles were shed from transiting vessels and, despite their relatively high densities, could be maintained temporarily at the sea surface through surface tension or when associated with other floating debris.

Despite limited information on the abundance and properties of microscopic FRP particles, 380 they are potentially more hazardous than "conventional" MPs that are commonly described 381 382 and studied. Thus, in addition to a resinous matrix, FRP is a source of glass fibres that have similar physical and chemical properties to asbestos (Galimany et al., 2009), and harmful 383 leaded pigments in associated paints. In the only study of the ecotoxicological impacts of 384 FRP that we are aware of, Ciocan et al. (2020) exposed fine particles ground from a 385 laminated sheet sourced from a boatyard in southern England (< 4 mm and up to 120 mg L<sup>-1</sup>) 386 to the mussel, Mytilus edulis, and water flea, Daphnia magna. In M. edulis, FRP was 387

detected in the digestive tubules and gills and caused a range of inflammatory responses in
all examined organs. In *D. magna*, particles adhered to the filament hairs of appendages and
impaired swimming.

## 391

## 392 5. Conclusions

393 High concentrations of plastic debris and microplastics have been found on and within the 394 sand in the vicinity of boat disposal sites on the landing stage at Chellanam. Plastic was 395 heterogeneously distributed amongst different sites and between size classifications and colours, but fragments of FRP made a significant and persistent contribution to the litter pool 396 397 in all cases. By comparison, along the HWL, plastic was less abundant and the contribution from FRP was lower. Paint associated with FRP had variable contents of Cu and Pb, with 398 concentrations of the latter being sufficient to contaminate sediments from a few mg kg-1 up 399 to about 400 mg kg<sup>-1</sup>. Because of its relatively high density, FRP is generally overlooked in 400 401 studies of plastics, and in particular microplastics. However, given its association with asbestos-like glass fibres and metal-bearing paints, further environmental studies are 402 403 recommended.

404

## 405 Acknowledgments

The authors thank ICAR for funding the study and the Director of the ICAR-Central Institute
of Fisheries Technology for providing facilities and guidance. We are grateful to the
fishermen of Chellanam for their time, cooperation and hospitality during the investigation,
and the technical staff of the Fishing Technology Division of ICAR-CIFT for their support.

## 416 References

Abbood, I.S., Odaa, S.A., Hasan, K.F., Jasim, M.A., 2021. Properties evaluation of fiber
reinforced polymers and their constituent materials used in structures – A review. Materials
Today Proceedings 43, 100301008.

- Arora, T., Rajankar, P., Chetry, B., 2018. Lead in paints in India: Concerns and Challenges.
  Toxics Link, New Delhi, 48pp.
- 422 Auta, H.S., Emenike, C.U., Fuziah, S.H., 2017. Distribution and importance of microplastics in
- 423 the marine environment: A review of the sources, fate, effects, and potential solutions.
- 424 Environmental International 102, 165-176.
- Canadian Council of Ministers of the Environment (2012) Canadian Environmental Quality
   Guidelines and Summary Table: http://www.ccme.ca/ accessed 12/2022.
- 427 Ciocan, C., Kristova, P., Annels, C., Derjean, M., & Hopkinson, L. (2020). Glass reinforced
  428 plastic (GRP) a new emerging contaminant First evidence of GRP impact on aquatic
  429 organisms. Marine Pollution Bulletin, 160, 111559.
- Cutroneo, L., Reboa, A., Geneselli, I., Capello, M., 2021. Considerations on salts used for
  density separation in the extraction of microplastics from sediments. Marine Pollution Bulletin
  16, 112216.
- 433 Eklund, B., Haaksi, H., Syversen, F., Elsted, R., 2013. Norden, (2013). Disposal of End-of-
- 434 life Plastic Boats. TemaNord https://www.diva-portal.org/smash/get/
- diva2:741961/FULLTEXT01.pdf. Accessed 12/2022.
- Eklund, B., Johansson, L., Ytreberg, E., 2014. Contamination of a boatyard for maintenanceof pleasure boats. Journal of Soils and Sediments 14, 955-967.
- 438 Erni-Cassola, G., Zadjelovic, V., Gibson, M.I., Christie-Oleza, J.A., 2019. Distribution of plastic
- 439 polymer types in the marine environment; a meta-analysis. J. Hazard. Mater. 369, 691-698.
- 440 GESAMP, 2015. Sources, fate and effects of microplastics in the marine environment: A global
- assessment. In: Kershaw, P.J. (Ed.), GESAMP Reports & Studies 93, International Maritime
  Organization, London.
- Galimany, E., Ramón, M., Delgado, M., 2009. First evidence of fiberglass ingestion by a marine invertebrate (*Mytilus galloprovincialis* L.) in a NW Mediterranean estuary. Mar. Pollut.
- 445 Bull. 58, 1334–1338.

- Graca, B., Szewc, K., Zakrzewska, D., Dolega, A., Szczerbowska-Boruchowska, M., 2017.
  Sources and fate of microplastics in marine and beach sediments of the Southern Baltic Sea—
  a preliminary study. Environmental Science and Pollution Research 24, 7650-7661.
- Higgins, C., Turner, A., 2023. Microplastics in surface waters around Plymouth, UK:
  Differential sources and transport of fibres and fragments. Science of the Total Environment
  (in review).
- IMO, 2019. End-of-Life Management of Fibre Reinforced Plastic Vessels: Alternatives to at
   Sea Disposal. International Maritime Organisation, CPY Group UK.
- 454 <u>https://wwwcdn.imo.org/localresources/en/OurWork/Environment/Documents/Fibre%20Reinf</u>
   455 <u>orced%20Plastics%20final%20report.pdf</u>.
- Jayasiri, H. B., Purushothaman, C. S., & Vennila, A., 2013. Quantitative analysis of plastic
  debris on recreational beaches in Mumbai, India. Marine Pollution Bulletin 77, 107–112.
- 458 Lee, J., Lee, J. S., Jang, Y. C., Hong, S. Y., Shim, W. J., Song, Y. K., Hong, S. H., Jang, M.,
- 459 Han, G. M., Kang, D., 2015. Distribution and size relationships of plastic marine debris on
- beaches in South Korea. Archives of Environmental Contamination and Toxicology 69, 288–298.
- Lekshmi, N.M., Kumar, S.S., Ashraf, P.M., Xavier, K.A.M., Prathish, K.P., Ajay, S.V., Edwin,
  L., Turner, A., 2023. Abandonment of fibreglass reinforced plastic fishing boats in Kerala, India,
  and chemical emissions arising from their burning. Estuarine, Coastal and Shelf Science
- 465 (submitted).
- Mattsson, K., Ekstrand, E., Granberg, M., Hassellöv, M., Magnusson, K., 2022. Comparison
  of pre-treatment methods and heavy density liquids to optimize microplastic extraction from
  natural marine sediments. Scientific Reports 12, 15459.
- 469 Opfer, S., Arthur, C., Lippiatt, S., 2012. NOAA Marine Debris Shoreline Survey Field Guide.
- 470 U.S. Department of Commerce National Oceanic and Atmospheric Administration.
- Rees, A. B., Turner, A., Comber, S., 2014. Metal contamination of sediment by paint peeling
  from abandoned boats, with particular reference to lead. Science of The Total Environment,
  494–495, 313–319.
- Rubino, F., Nisticó, A., Tucci, F., Carlone, P., 2020. Marine application of fiber reinforced
  composites: A review. Journal of Marine Science and Engineering 8, 26.
- 476 Singh, N., Turner, A., 2009. Trace metals in antifouling paint particles and their heterogeneous
- 477 contamination of coastal sediments. Mar. Pollut. Bull. 58, 559-564.

- Song, Y.K., Hong, S.H., 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.
  Environmental Science and Technology 48, 9014-9021.
- Soroldoni, S., Castro, Í.B., Abreu, F., Duarte, F.A., Choueri, R.B., Möller, O.O., Fillmann, G.,
  Pinho, G.L.L., 2018. Antifouling paint particles: Sources, occurrence, composition and
  dynamics. Water Research 137, 47-56.
- Summerscales, J., Singh, M.M., Wittamore, K., 2016. Disposal of composite boats and other
  marine composites. In: Marine Applications of Advanced Fibre-Reinforced Composites,
  Woodhead Publishing Series in Composite Science and Engineering pp. 185-213.
- 487 Turner, A., Rees, A., 2016. The environmental impacts and health hazards of abandoned
- boats in estuaries. Regional Studies in Marine Science, 6, 75–82.
- Turner, A., Solman, K.R., 2016. Lead in exterior paints from the urban and suburban environs
  of Plymouth, south west England. Science of the Total Environment 547, 132-136.
- Woods, J.S., Verones, F., Jolliet, O., Vázquez-Rowe, I., Boulay, A.M., 2021. A framework for
  the assessment of marine litter impacts in life cycle impact assessment. Ecological Indicators
  12, 107918.
- 494