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

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- **microplastics on a beach impacted by abandoned fishing boats: A**
- **case study from Chellanam, India**
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Abstract

 Plastics and microplastics have been quantified and characterised at boat disposal sites and along the high-water line (HWL) of a fish landing centre beach in Chellanam, India. Fibreglass- reinforced plastic (FRP) made a greater contribution to the plastic pool at the disposal sites (~ $4.5 n m²$ and 18 g m⁻²) than at the HWL ($\sim 0.25 n m⁻²$ and $\lt 1$ g m⁻²) and was an abundant component of the microplastic pool at the former. Infrared analysis of microplastic-sized FRPs revealed various resins (e.g., alkyd, polyester, epoxy), while X-ray fluorescence analysis of the painted surfaces of meso-sized FRPs returned variable concentrations of copper and lead. 24 Concentrations of Pb were high enough to contaminate sand up to \sim 400 mg kg⁻¹. The relatively 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-composite) thermoplastics.

Keywords

- Debris; fragments; paint; copper; lead; disposal
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Highlights

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

1. Introduction

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

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

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

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

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

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

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

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

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

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

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

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

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

solution whose density typically ranges from 1.2 to 1.8 g $cm⁻³$ (Mattsson et al., 2022). As a

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

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

 FRP consists of a polymeric matrix or binding agent, such as epoxy or polyester thermoset, 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 has extensive usage in components and structures in the maritime sector. Of particular importance here is the manufacture of small boats (e.g., fishing boats and recreational craft), with about 80% of vessel hulls up to 20 m in length constructed of FRP (Rubino et al., 2020). In India, FRP is used to construct boat hulls or as a sheathing material over a wooden 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 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 FRP-sheathed boats used in the artisanal fishing industry of Chellanam (India). Here, the beach is used as a landing stage and boats are stored, maintained and abandoned without

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

- microscopically and by infrared spectroscopy, and the content of potentially toxic metals on
- 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.
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2. Materials and methods

2.1. Study site and boat abandonment

81 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

83 fishing boats operating from Chellanam are motorised and below 14 m in length and are FRP-sheathed 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).

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

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 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

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 fibres and remains of a wooden substrate on the other side), fishing waste (including polyurethane foam used as an insulating material, netting, floats and sinkers) and other (e.g., 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 about 2 cm was collected with a stainless-steel trowel and stored in a stainless-steel container in order to determine microplastics and metals. At each disposal site, three 5 m x 5 m quadrats were sampled for plastic debris before three 0.5 m x 0.5 m quadrats were defined within each larger quadrat and sampled for sand as above.

2.3. Sample processing

 In the laboratory, classes of plastic were counted and weighed and FRP debris was further classified by size as mega-debris: >10 cm; macro-debris: 2 - 10 cm; and meso-debris: 5 - 20 mm (Jayasiri et al., 2013). About 1 kg of sand from each small quadrat along the HWL (*n* = 146 10) and within the disposal sites $(n = 9)$ was weighed out on a Sartorius electronic balance before the contents were passed through a 5-mm stainless steel sieve. Material passing 148 through the sieve was dried at 40 $^{\circ}$ C for 24 h before being subject to density separation. Thus, 149 30 g of material was added to a 100 mL solution of saturated ZnCl₂ (CDH Analytical Grade in 150 Milli-Q water) of density 1.6 to 1.7 g cm⁻³ 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 152 filtered through individual Whatman 41 filter papers (20 μ m pore size) before retained particles 153 were passed through a sequence of stainless steel sieves (mesh sizes 300 μ m and 63 μ m) to 154 separate coarse (300 μ m to 5 mm), medium (63 μ m to 300 μ m) and fine (20 μ m to 63 μ m) 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 MZ16A stereomicroscope at up to 230 X magnification.

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 infra- red (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.

165 Spectra were acquired between 650 and 4000 cm^{-1} , with 50 scans per sample, and were compared with a Hummel polymer library embedded in Omnic software.

2.5. Sample digestion and copper and lead analysis

169 From six sand samples along the HTL and all samples from the disposal sites $(n = 9)$, about 10 g of < 5 mm material was ground in a pestle and mortar. In triplicate, 1 g portions were weighed into a series of acid-cleaned, 50 mL Pyrex beakers to which 8 mL of aqua-regia (3:1 172 HCl: HNO₃; Merck AR) was added. The contents of each beaker were covered with a watch glass and gently boiled on a hotplate for 1 h before the cooled digests were vacuum-filtered 174 through 0.45 µm 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 176 same protocol but in the absence of sand.

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

180 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 meso- fragments 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 187 around 30 mg kg $^{-1}$ for Cu and 10 mg kg $^{-1}$ for Pb.

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 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 from 6 m to 8 m.

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 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 199 average numbers of plastic per $m²$ based on totals for each category divided by the total area of quadrats surveyed.

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 Table 1 summarises the number of pieces of plastic debris retrieved from the quadrats along the HWL (H1 to H10) and within the three disposal sites (D1, D2 and D3). Along the HWL, 134 pieces of plastic debris were recovered, of which about 46% were visually identified as FRP. At the disposal sites 1179 pieces of plastic debris were recovered of which about 87% were 207 FRP. When normalised on an area basis, debris in each category, and in particular for the 208 different size classes of FRP, was greater at the disposal sites than along the HWL.

209 Table 2 summarises the plastic debris data on a mass basis. Along the HWL, over 2 kg of 210 plastic was retrieved, with fishing waste contributing more than 70% and FRP contributing 211 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

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

215 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

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3.2. Microplastic abundance and visual characteristics

 Figure 3: Examples of FRP and other microplastic fragments observed under the microscope. (a) and (b) were taken directly from abandoned boats and (c) to (f) were sampled from disposal sites.

 Microscopic images of six microplastic fragments retrieved in the study are shown in Figure 3. Fragments were either (i) relatively stiff and flat irregular shapes, with (often) one or more 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 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 or single or multiple threads that appeared to be intertwined, while microplastic pellets (not illustrated) were distinctly rounded but with occasional irregularities (e.g., pits, protrusions, overlapping layers). Table 3 shows the number of MPs identified in the quadrats at each location, normalised to a

 sediment mass of 30 g, along with their distributions by size and shape. Along the HWL, numbers range from 34 to 155 with a median of 82. At the disposal sites, numbers range 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 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 least abundant.

- On a size basis, however, and as illustrated in Figure 4, the percentage of fragments
- decreased with decreasing size at the HWL (from about 72% in coarse to about 26% in fine)
- whereas pellets (about 50%) and fibres (about 65%) were most important in the medium and
- fine fractions, respectively. By contrast, at the disposal sites, fragments dominated the
- medium size fraction (nearly 90%) whereas pellets and fibres were most important in the
- fines.
- 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
- microplastics, blue fragments and fibres, yellow and green pellets and fragments, and brown
- fibres. More abundant at the disposal sites, and above the line of unit slope, are white
- microplastics, blue pellets, yellow fibres and brown fragments and pellets.
- 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 colour distribution.
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259 Figure 4: Percentage distribution of microplastics by size and shape across the HWL and at

260 the disposal sites.

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

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

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

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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.

3.3. Polymeric composition of microplastics

 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 most abundant and polyester-, epoxy-, polystyrene- and piperylene-based resins also 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 281 peaks in the region 1000-1200 cm⁻¹ through asymmetric stretches associated with silicate glass (Hopkinson et al., 2021). At the disposal sites, there was a combination of resin-based 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 were not identified and polyethylene was the most abundant polymer among the various fragments and fibres tested.

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

category). Note that polyester includes polyethylene terephthalate.

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 294 concentrations of Cu range from 1.1 mg kg⁻¹ to 11.0 mg kg⁻¹, with relative standard deviations for replicate analyses ranging from about 5% to 35%. Median concentrations are 296 1.5 mg kg⁻¹ and 3.6 mg kg⁻¹ at the HWL and disposal sites, respectively, with a Mann- Whitney U-test (Minitab, *v*19) revealing a significantly higher median (*p* < 0.05) at the latter. Mean concentrations of Pb are greater than Cu in each sample and are more variable 299 (ranging from about 3 mg kg⁻¹ to 400 mg kg⁻¹). As with Cu, median concentrations are significantly higher at the disposal site than the HWL.

- Concentrations of Cu and Pb in the mesoplastic FRP fragments analysed by XRF are summarised in Table 6. Where detected, Cu concentrations are greater than those found in
- 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.
- Lead was detected in fewer mesoplastics but median concentrations are significantly higher
- than Cu in both the HWL and disposal sites.

310 Table 5: Concentrations of Cu and Pb (in mg kg⁻¹) 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

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

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317

315 Table 6: Summary statistics for the concentrations of Cu and Pb (in mg kg⁻¹) in the

316 mesoplastics sampled from the HWL and disposal sites.

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 disposal sites and along the HWL, but differences were evident between the colour and size distributions at the different location types. Analysis of a selection of microplastics by FTIR 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 densities of polyethylene and polypropylene than FRP. Thus, while the density of polyolefins 332 is below 1 g cm⁻³, 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⁻³ (Abbood et al., 2021), with slight modifications possible when the surface is painted. The lower density plastics may be more readily transported from the disposal sites to the HWL and, with buoyant plastics derived from offshore, are subject to redistribution, recirculation and accumulation in the intertidal zone (Graca et al., 2017). Conversely, higher density FRP is less readily transported to the HWL (Ciocan et al., 2020), and any material reaching this area is more likely to be subject to transportation with subsurface offshore currents and sinking and burial.

 Analysis of the painted surfaces of larger fragments of meso-sized FRP from the disposal 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 environmental perspective, however, are variable but higher (median) concentrations of Pb. Quantitatively, our results are similar to those reported by Hopkinson et al. (2021) for 14 samples of FRP sourced from various breakers yards in southern England (Cu ~ 150 to 1600 347 mg kg⁻¹; Pb ~ 95 to 10,400 mg kg⁻¹). This suggests that, despite extensive restrictions on the Pb content of consumer paints, leaded paint is still used or has been applied relatively recently on FRP more generally. With regard to India, a maximum Pb concentration in 350 consumer paints of 90 mg $kg⁻¹$ has been required since 2017. However, we note that in a chemical assessment of Indian paints undertaken since the restrictions, Arora et al. (2018) found that paints with very high levels of Pb are still widely sold across the country and that

 consumer awareness of the problem is extremely low. In the present setting at least, a 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 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, the Pb concentration in one sample at disposal site D3 exceeded the CCME marine 359 sediment quality guideline for the protection of aquatic life (112 mg kg⁻¹; Canadian Council of Ministers of the Environment, 2012).

 There is very little information on the occurrence, fate and effects of FRP in the marine science literature, and especially regarding microplastics in sediment, from which to draw comparisons with the present study. The main reason for this is, likely, that conventional means of isolating microplastics from sediments involves flotation in a saturated salt solution, 365 and common solutions employed, like NaCl, NaI and ZnCl₂ (Cutroneo et al., 2021), have a 366 density range $(-1.2 \text{ to of } 1.8 \text{ cm}^{-3})$ that would preclude many glass-reinforced resins. Nevertheless, given the importance of FRP in the leisure and commercial boating sectors (Ciocan et al., 2020) and with regard to other maritime structures (Summerscales et al., 2016), and the documentation of other boat-derived debris (mainly paint particles) in the 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 contamination is predicted to be a more general problem. 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, they are potentially more hazardous than "conventional" MPs that are commonly described 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 leaded pigments in associated paints. In the only study of the ecotoxicological impacts of FRP that we are aware of, Ciocan et al. (2020) exposed fine particles ground from a 386 laminated sheet sourced from a boatyard in southern England ≤ 4 mm and up to 120 mg L⁻¹) to the mussel, *Mytilus edulis*, and water flea, *Daphnia magna*. In *M. edulis*, FRP was

 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.

5. Conclusions

 High concentrations of plastic debris and microplastics have been found on and within the sand in the vicinity of boat disposal sites on the landing stage at Chellanam. Plastic was 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 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 399 concentrations of the latter being sufficient to contaminate sediments from a few mg $kg⁻¹$ up 400 to about 400 mg kg⁻¹. Because of its relatively high density, FRP is generally overlooked in studies of plastics, and in particular microplastics. However, given its association with asbestos-like glass fibres and metal-bearing paints, further environmental studies are recommended.

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