

Distribution of Microplastics in Estuarine Sediments along Tamilnadu Coast, Bay of Bengal, India

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Abstract –The occurrence and distribution of large plastic debris in the marine environment are well documented but up to now, only five studies including the Gulf of Cambay, India on the occurrence of plastic fragments have included true microplastics. In the east coast of India, there was no data for the occurrence and distribution of microplastics. Information on the distribution of microplastics in the Tamilnadu coastal environment is necessary to determine the extent of microplastic pollution input into the sea. This study aims to investigate the occurrence and distribution of microplastics in Tamilnadu coastal sediments collected from three estuarine regions.

Keywords– Microplastics, Estuarine sediment, Uppanar River, Vellar River, Coleroon River, Bay of Bengal.

I. INTRODUCTION

Few centuries, humans have been disposing of waste into the sea where it eventually deposits on the coastline or the seabed. Marine pollution occurs when harmful effects, or potentially harmful effects, can result from the entry into the ocean of chemicals, particles, industrial, agricultural and residential waste, noise, or the spread of invasive organisms. Most sources of marine pollution are land-based. The pollution often comes from nonpoint sources such as agricultural runoff and windblown debris.

Many potentially toxic chemicals adhere to tiny particles which are then taken up by plankton and benthos animals, most of which are either deposit or filter feeders. In this way, the toxins are concentrated upward within ocean food chains. Many particles combine chemically in a manner highly depletive of oxygen, causing estuaries to become anoxic. Once in the food webs, these pesticides can cause mutations, as well as diseases, which can be harmful to humans as well as the entire food web.

The global production of plastics was approximately 1.5 million tonnes/year in the 1950s. Currently, it is estimated at almost 250 million tonnes/year and it is still increasing by 10% each year (Plastics Europe, 2008). Plastic product production was estimated at 2.63 billion kg in 2004 in the United States alone. While reliable estimates of the input of produced plastics in the environment cannot be obtained, substantial amounts end up in the marine environment through industrial discharge, littering and terrestrial runoff (Derraik, 2002). This has led to increasing levels of plastic litter in oceans worldwide.

Types of Plastics

Plastics are natural/ synthetic materials. They are produced by chemically modifying natural substances or are synthesized from inorganic and organic raw materials. Based on their physical characteristics, plastics are usually divided into thermosets, elastomers, and thermoplastics. These groups differ primarily about molecular structure, which is what determines their different thermal behavior. The characteristics of the various types of plastics are given in the following Table

1. Thermoset or thermosetting plastics:

Once cooled and hardened, these plastics retain their shapes and cannot return to their original form. They are hard and durable.

2. Thermoplastics:

Less rigid than thermosets, thermoplastics can soften upon heating and return to their original form. They are easily molded and extruded into films, fibers, and packaging. Examples include polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC).




3. Common types of plastics:

Polyethylene terephthalate (PET or PETE): John Rex Whinfield invented a new polymer in 1941 when he condensed ethylene glycol with terephthalic acid. The condensate was polyethylene terephthalate (PET or PETE). PET is a thermoplastic that can be drawn into fibers (like Dacron) and films (like Mylar). It's the main plastic in zip lock food storage bags.

4. Polyvinyl Chloride (PVC): PVC is a thermoplastic that is formed when vinyl chloride ($\text{CH}_2=\text{CH}-\text{Cl}$) polymerizes. When made, it's brittle, so manufacturers

add a plasticizer. When made, it's brittle, so manufacturers add a plasticizer liquid to make it soft and moldable. PVC is commonly used for pipes and plumbing because it's durable, can't be corroded and is cheaper than metal pipes. Over long periods, however, the plasticizer may leach out of it, rendering it brittle and breakable.

Table - I: Types and characteristics of Plastics

TYPE OF PLASTIC	MOLECULAR STRUCTURE	CHARACTERISTICS AND APPLICATIONS
Thermosets		Thermosets are hard and have a very tight-meshed, branched molecular structure. Curing proceeds during shaping, after which it is no longer possible to shape the material by heating. Further shaping may then only be performed by machining. Thermosets are used, for example, to make light switches.
Elastomers		While elastomers also have a cross-linked structure, they have a looser mesh than thermosets, giving rise to a degree of elasticity. Once shaped, elastomers also cannot be reshaped by heating. Elastomers are used, for example, to produce automobile tires.
Thermoplastics		Thermoplastics have a linear or branched molecular structure that determines their strength and thermal behavior; they are flexible at ordinary temperatures. At approx. 120 - 180°C, thermoplastics become a partly liquid mass. The service temperature range for thermoplastics is considerably lower than that for thermosets. The thermoplastics polyethylene (PE), polyvinyl chloride (PVC) and polystyrene (PS) are used, for example, in packaging applications.

5. Polytetrafluoroethylene (Teflon): Teflon was made in 1938 by DuPont. It's created by polymerization of tetrafluoroethylene molecules ($CF_2=CF_2$). The polymer is stable, heat-resistant, strong, and resistant to many chemicals and has a nearly frictionless surface. Teflon is used in plumbing tape, cookware, tubing, waterproof coatings, films, and bearings.

6. Polyvinylidene Chloride (Saran): Dow makes Saran resins, which are synthesized by polymerization of vinylidene chloride molecules ($CH_2=CCl_2$). The polymer can be drawn into films and wraps that are impermeable to food odors. Saran wrap is a popular plastic for packaging foods.

7. Polyethylene, LDPE, and HDPE: The most common polymer in plastics is polyethylene, which is made from ethylene monomers ($CH_2=CH_2$). The first polyethylene was made in 1934. Today, we call it low-density polyethylene (LDPE) because it will float in a mixture of alcohol and water. In LDPE, the polymer strands are entangled and loosely organized, so it's soft and flexible. It was first used to insulate electrical wires, but today it's used in films, wraps, bottles, disposable gloves, and garbage bags. In the 1950s, Karl Ziegler polymerized ethylene in the presence of various metals. The resulting polyethylene polymer was composed of mostly linear

polymers. This linear form produced tighter, denser, more organized structures and is now called high-density polyethylene (HDPE).

7. Polypropylene (PP): In 1953, Karl Ziegler and Giulio Natta, working independently, prepared polypropylene from propylene monomers ($CH_2=CHCH_3$) and received the Nobel Prize in Chemistry in 1963. The various forms of polypropylene have different melting points and hardnesses.

8. Plastics used in the marine environment

The annual global demand for plastics has consistently increased over the recent years and presently stands at about 245 million tonnes. Being a versatile, lightweight, strong, and potentially transparent material, plastics are ideally suited for a variety of applications. Their low cost, excellent oxygen/moisture barrier properties, bio-inertness, and lightweight make them excellent packaging materials. Conventional materials such as glass, metal, and paper are being replaced by cost-effective plastic packaging of equivalent or superior design. Nearly a third of the plastic resin production is therefore converted into consumer packaging material that includes disposable single-use items commonly encountered in beach debris (Andrady, 2003).

Several broad classes of plastics are used in packaging: Polyethylene (PE), Polypropylene (PP), Polystyrene (PS), Poly (ethylene terephthalate) (PET); and Poly (vinyl chloride) (PVC). Their high-volume usage is reflected in their production figures given in Table 2 and consequently, these, in particular, have a high likelihood of ending up in the ocean environment.

Table -II: Classes of plastics that are commonly encountered in the marine environment.

PLASTIC CLASS	SPECIFIC GRAVITY	PERCENTAGE PRODUCTION*	PRODUCTS AND TYPICAL ORIGIN
Low-density polyethylene (LDPE)	0.91 - 0.93	21%	Plastic bags, six-packings, bottles, netting, drinking straws
High-density polyethylene (HDPE)	0.94	17%	Milk and juice jugs
Polypropylene PP	0.83 - 0.85	24%	Rope, bottle caps, netting
Polystyrene PS	1.05	6%	Plastic utensils, food containers
Foamed Polystyrene			Floats, bait boxes, foam cups
Nylon	PA	<3%	Netting and traps
Thermoplastic polyester PET	1.37	7%	Plastic beverage bottles
Poly (vinyl chloride) PVC	1.38	19%	Plastic film bottles, cups
Cellulose Acetate CA			Cigarette filters

* Fraction of the global plastics production in 2007 after (Brien, 2007) Land-based sources including beach litter contribute about 80% of the plastic debris. The entire global fishing fleet now uses plastic gear and some gear is invariably lost or even discarded carelessly at sea during use. Polyolefins (PE and PP) and nylons are primarily used in fishing gear applications (Timmers et al., 2005; Klust, 1982). About 18% of the marine plastic debris found in the ocean environment is attributed to the fishing industry. +Aquaculture can also be a significant contributor to plastics debris in the oceans (Hinojosa and Thiel, 2009).

The rest is derived largely from land-based sources including beach litter. Virgin resin pellets, a common component of debris, enter the oceans routinely via incidental losses during ocean transport or through run-off from processing facilities (Gregory, 1996; Doyle et al., 2011; Ogata et al., 2009). Quantifying floating plastic debris (generally using surface water collection of debris with neuston nets) seriously underestimates the amounts of plastics in the ocean as those in the sediment and mid-water are excluded by the technique.

The visibility of debris as flotsam requires plastics to be positively buoyant in seawater (specific gravity of seawater is ~1.025). However, as seen from Table 1 only a few of the plastics typically used in the marine environment has a specific gravity lower than that of seawater. (The specific gravities given are for the virgin resins; plastics in products are often mixed with fillers and other additives that may alter their specific gravity.) Denser varieties of plastics such as nylons tend to submerge in the water column and even reach the coastal sediment.

8. Microplastics

Marine debris is mainly discarded human rubbish which floats on or is suspended in the ocean. The occurrence and distribution of large plastic debris in the marine environment are well documented and the adverse effects of this type of pollution on marine life have been described extensively by many researchers. However, these larger items eventually undergo fragmentation which leads to the formation of micro- particulates of plastic. These so-called ‘microplastics’ (plastic particulates 1mm) may become widely distributed in the marine environment through hydrodynamic processes and ocean currents.

9. Sources of microplastics

There are two main sources of microplastics:

- Microplastics that are produced either for direct use, such as for industrial abrasives, exfoliants, cosmetics or roto milling or indirect use as precursors (so-called resin pellets or nurdles) for the production of manifold consumer products ("primary microplastics").
- Microplastics formed in the environment as a consequence of the breakdown of larger plastic

material, especially marine debris, into smaller and smaller fragments (so-called "secondary microplastics"). The breakdown is caused by mechanical forces (e.g. waves) and/or photochemical processes triggered by sunlight (especially UVB). The abundance and global distribution of microplastics in the oceans has steadily increased over the last few decades with rising plastic consumption worldwide (Moore, 2008).

9. Microplastics in the oceans

Microplastics, a form of man-made litter, have been accumulating in the oceans for at least over the last four decades (Thompson et al., 2004, 2005). Sampled from surface waters or from beach sand this fraction of litter includes virgin resin pellets, compounded masterbatch pellets and smaller fragments of plastics derived from the larger plastic debris (Moore, 2008).

The term ‘microplastics’ and ‘microlitter’ have been defined differently by various researchers. Gregory and Andrady (2003) defined microlitter as the barely visible particles that pass through a 500 µm sieve but retained by a 67 µm sieve (~0.06–0.5 mm in diameter) while particles larger than this were called mesolite. Others (Fendall and Sewell, 2009; Betts, 2008; Moore, 2008), including a recent workshop on the topic (Arthur et al., 2009) defined the microparticles as being in the size range <5 mm (recognizing 333 µm as a practical lower limit when neuston nets are used for sampling). The global distribution of microplastics is given in Figure _ 1.



Fig .1.Global distribution of microplastics.

Particles of plastics that have dimensions ranging from a few 1m to 500 µm (5 mm) are commonly present in seawater (Ng and Obbard, 2006; Barnes et al., 2009). For clarity, this size range alone is referred to as ‘microplastics’ here; the larger particles such as virgin resin pellets are referred to as ‘mesoplastics’ after Gregory and Andrady (2003). Persistent organic

pollutants (POPs) that occur universally in seawater at very low concentrations are picked up by microplastics via partitioning. It is the hydrophobicity of POPs that facilitate their concentration in the microplastic litter at a level that is several orders of magnitude higher than that in seawater. These contaminated plastics, when ingested by marine species, present a credible route by which the POPs can enter the marine food web.

The extent of bioavailability of POPs dissolved in the microplastics to the biota (Moore, 2008) and their potential bio-magnification in the food web (Teuten et al., 2007) has not been studied in detail. The following is only a suggested procedure derived from published reports as well as the personal experience of the author. Water samples are filtered through a coarse filter to remove mesolite. Sediment or sand samples are slurred in saline water to allow microplastics to float to the surface. A mineral salt may be dissolved in the collected seawater or slurry sample to increase the water density sufficiently to float plastic fragments. Samples of surface water with floating microparticles are carefully removed for study.

Concentrating samples of seawater samples by evaporation can also concentrate the microplastic litter at the surface. Microplastics in surface water samples can be visualized under a microscope using a lipophilic dye (such as Nile Red) to stain them (Andrady, 2010). The water samples will also contain microbiota such as plankton of the same size range but these will not be stained by lipophilic dyes. As a prelude to discussing the mechanisms responsible for the generation of microplastics, understanding the light-induced degradation and biodegradation of plastics in the marine environment is important.

10. Plastics degradation under marine conditions

Degradation is a chemical change that drastically reduces the average molecular weight of the polymer. Since the mechanical integrity of plastics invariably depends on their high average molecular-weight, any significant extent of degradation inevitably weakens the material. Extensively degraded plastics become brittle enough to fall apart into powdery fragments on handling. Even these fragments, often not visible to the naked eye, can undergo further degradation (generally via microbial-mediated biodegradation) with the carbon in the polymer being converted into CO₂ (and incorporated into marine biomass). When this process goes onto completion and all the organic carbon in the polymer is converted, it is referred to as complete mineralization (Andrady, 1994, 1998; Eubeler et al., 2009).

- Degradation is generally classified according to the agency causing it.
- Biodegradation – action of living organisms usually microbes.
- Photo-degradation – action of light (usually sunlight in outdoor exposure).

- Thermo-oxidative degradation – slow oxidative breakdown at moderate temperatures.
- Thermal degradation* – the action of high temperatures.
- Hydrolysis – reaction with water.

*Not an environmental degradation mechanism.

11. Toxicity of ingested microplastics

Seawater already contains numerous natural microparticles most of them <100 nm in size (Rosse and Loizeau, 2003). Filter feeders in the ocean ranging from the nano-zooplanktons to Baleen Whales, routinely interact with these without any apparent ill effect. As no enzymatic pathways available to break down the synthetic polymers in any of these organisms, ingested microplastics are also never digested or absorbed and should, therefore, be bio-inert. Ingestion of microplastics by microbiota, however, presents a very different problem. The concern is their potential for delivery of concentrated POPs, mainly those picked up from seawater, to the organisms (Bowmer and Kershaw, 2010). It is this dissolved POPs that yield the toxic outcomes.

Any toxicity associated with plastics in general, including masoor microplastics, can be attributed to one or more of the following factors:

- Residual monomers from manufacture present in the plastic or toxic additives used in the compounding of plastic may leach out of the ingested plastic. {An example of residual monomer is illustrated by the recent issue on residual bisphenol A (BPA) in polycarbonate products (Vandenberg et al., 2007). The potential toxicity of phthalate plasticizers used in PVC has been widely discussed in the literature (Latini et al., 2004).
- Toxicity of some intermediates from partial degradation of plastics. For instance, burning polystyrene can yield styrene and other aromatics and a partially burnt plastic may contain significant levels of styrene and other aromatics.
- The POPs present in seawater is slowly absorbed and concentrated in the microplastic fragments. Plastics debris does 'clean' the seawater of the dissolved pollutant chemicals. On being ingested, however, these can become bioavailable to the organisms (Endo et al., 2005).

II. DESCRIPTION OF THE STUDY AREA

India has a long coastline of 7,517 km and this 6,000 km is rich in estuaries, creeks, brackish water, lagoons, and lakes. The southeast coast of India is an important stretch of coastline with many significant landmark features, where many major rivers drain into the Bay of Bengal and they are also richer in marine fauna than the western coast of India. Cuddalore town is a significant coastal city in Tamilnadu state that hosts several large-scale industries. The Uppanar River runs parallel to the coast south of Cuddalore town having the number of small streams of domestic, treated and untreated effluent discharges from

industries. Uppanar estuary is constituted by the confluence of the Gadilam and Paravandar rivers that flow into the Bay of Bengal. The main industries that existed along the western bank of Uppanar River include chemicals, beverage manufacturing, tanneries, oil, soap, paint production, paper, and metal processing plants. Cuddalore harbor located in the estuarine region of Uppanar River is occupied by a fleet of mechanized fishing boats, which operates in the coastal zone. This open estuary has an average depth of 2.5 m and a width of 30 m near the mouth and 20 m upstream.

The tidal influence extends to 6 km upstream. The Vellar river originates at the Kalvarayan hills in the Salem district, runs for 90 km and flows through the borders of Villupuram and Perambalur districts for very few kilometers. In its stretch, it enters the Cuddalore district, flowing for another 105 km, and ends its journey into the Bay of Bengal at Parangipettai. The Vellar estuary always remains open and is called a “true estuary.” The maximum amplitude of the tide reaches about 1 m. The tidal influence felt up to 10–15 km upstream. The average depth of the estuary is 2.5 m. The Coleroon River is located in the southern part of the study area.



Fig .2. The study area and sampling locations .

III. MATERIALS AND METHODS

1. Sampling and analysis

To quantify the abundance of microplastics, the estuarine sediments were collected from Uppanar (S1), Vellar (S2) and Coleroon (S3) estuaries using a small trowel during July 2011 (Figure 2). Five kilograms of sediment samples were collected from each location. The microplastics in the sediment samples were extracted using the method of Thompson et al. (2004) with some minor modifications. In short, 3 L of a concentrated saline solution (1.2kg NaCl

l-1) was added to 1 kg of sieved sediment and stirred for 2 min. The sediment was then allowed to settle for 1 hour before the supernatant was poured through a 38 µm mesh sieve. Filters were dried at 20C, sealed in Petri-dishes to prevent contamination. Microplastic concentrations were expressed as a number of particles kg-1dry sediment and on a weight basis (mg microplastics kg-1dry sediment) to allow comparison with other studies. The collected particles were examined using a fluorescence microscope. The particle recovery of the extraction procedure was ascertained by spiking known concentrations of microplastics (of similar dimensions as those encountered in the field) into clean sediment and subjecting it to repeated extractions. The complete analytical procedure has given in Figure 3a. The Photograph Fluorescence Spectroscopy has shown in figure 3b.

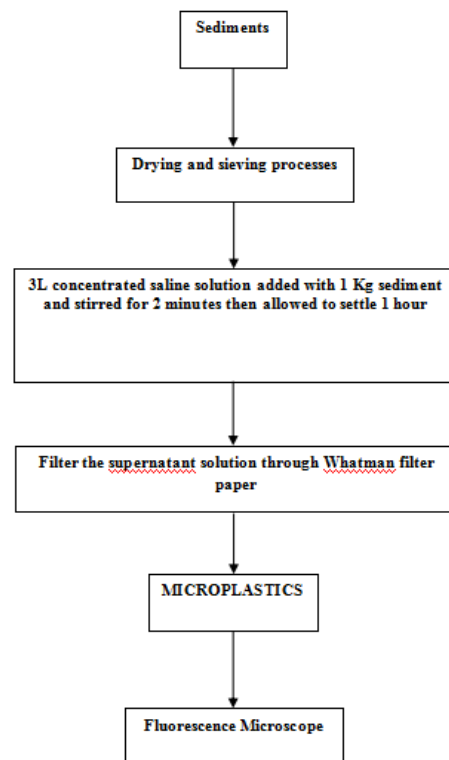


Fig .3a. Procedure for isolation and characterization of microplastics.



Fig .3b. The photograph of Fluorescence Microscopy.

IV. RESULTS AND DISCUSSION

The measured microplastics concentration is illustrated in Figure 4. The mean concentration values of microplastic fragments in Uppanar, Vellar, and Coleroon estuarine sediments are 30.2, 9.4 and 11.7 mg kg⁻¹ respectively. Among the three sampling locations, Uppanar (S1) appeared to have a higher accumulation of total plastics in the sediments than other stations.

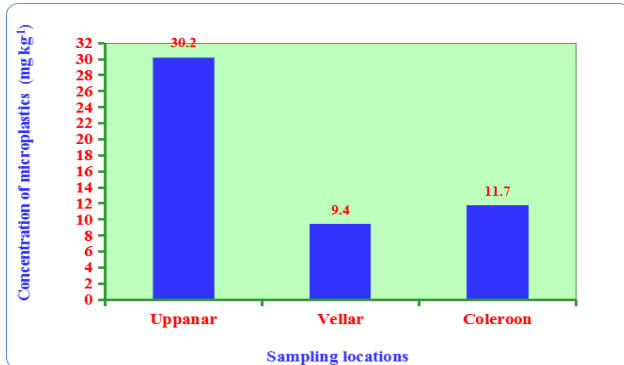


Fig.4.Distribution of microplastics along Tamilnadu coast.

The lowest accumulation of plastic fragments was recorded at Vellar estuarine sediments. The presence of a high concentration of microplastics in Uppanar sediments is likely due to on-going waste disposal particles from industries around the study area, recreational activities and discharge from shipping and boating activities.

Sea-based sources of plastics do not appear to be as significant as land-based sources and are even more difficult for countries in the region to control; the east coast of India has a high density of commercial shipping and fishing vessels. Loss of fishing gear and dumping of rubbish is also enhancing the level of microplastics in the marine environment. Microplastics have the potential to become widely dispersed in the marine environment via hydrodynamic processes and ocean currents, due to their buoyant and persistent properties (Ng and Obbard, 2006). The microplastic fragments were identified using a fluorescence microscope. The morphology of microplastics in Uppanar estuarine sediments is shown in Figures 5 and 6.

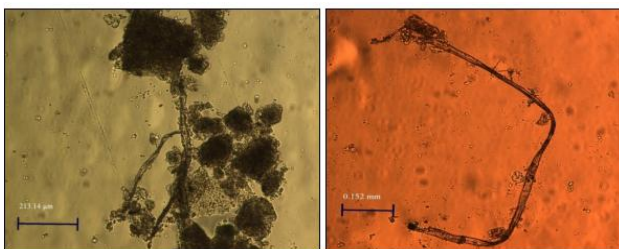


Fig .5.Microscopic images of microplastics in Uppanar estuarine sediments

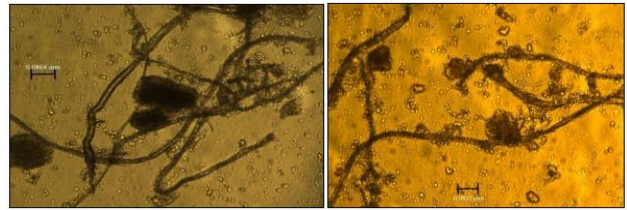


Fig.6.Microscopic images of microplastics in Uppanar estuarine sediments.

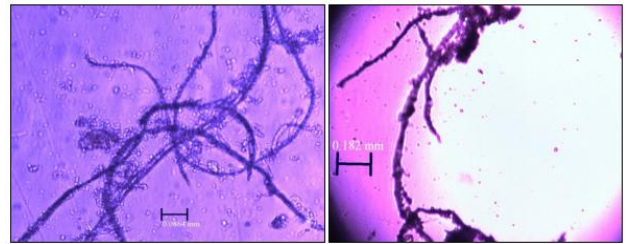


Fig .7. Microscopic images of microplastics in Vellar estuarine sediments.

The morphology of microplastics in Vellar estuarine sediments is shown in Figure 7. The morphology of microplastics in Vellar estuarine sediments is shown in Figures 8 and 9.

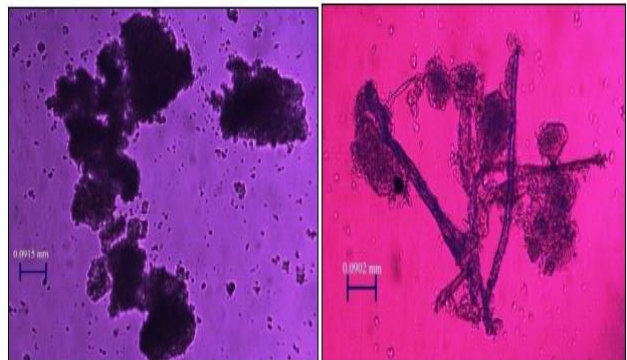


Fig.8. Microscopic images of microplastics in Coleroon estuarine sediments.

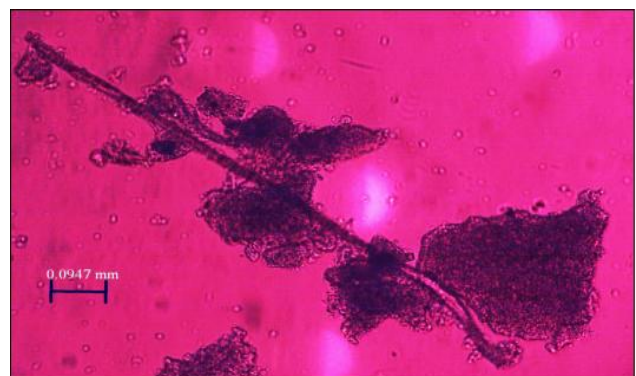


Fig.9. Microscopic images of microplastics in Coleroon estuarine sediments.

The measured microplastic concentrations were compared with the other similar studied and reported areas around

the world and given in Table 3. The mean value of microplastics (17.1 mg kg⁻¹) in the present study is almost three times lower than the average value (81 mg kg⁻¹) reported from the west coast of India (Alang-Sosiya). Like other environmental problems, marine microplastics can be prevented and controlled through an effective collaboration of education, legislation, and innovation. Recognition of microparticles is most likely generated on beaches underlines the importance of beach cleaning as an effective mitigation strategy.

Table -III: The comparison table for the occurrence and distribution of microplastics. Original unit (# fibers 50 mL⁻¹ sediments) converted using an average sediment density of 1600 kg m⁻³ and 1.25 as an average wet sediment/dry sediment ratio.

Country	Location	Concentration	Unit	Reference
Belgian	Harbour	7.21	mg kg ⁻¹	Claessens et al., 2011
Singapore	Beach	3	#Kg ⁻¹	Ng and Obbard, 2006
India (West coast)	Ship-breaking yard	89	mg kg ⁻¹	Srinivasa Reddy et al., 2011
United Kingdom	Beach	8	*Kg ⁻¹	Thompson et al., 2004
United Kingdom	Estuarine	31	*Kg ⁻¹	Thompson et al., 2004
United Kingdom	Sub tidal	86	*Kg ⁻¹	Thompson et al., 2004
India	Estuarine	9.4–30.2	mg kg ⁻¹	Present study

The removal of larger pieces of plastic debris from beaches before these are weathered enough to be surface embrittled can have considerable value in reducing the microplastics that end up in the ocean. Beach cleanup, therefore, can have an ecological benefit far beyond the aesthetic improvements of the beaches, and by reducing microplastics, contributes towards the health of the marine food web.

V. CONCLUSION

Coastal, estuarine and marine ecosystems are highly dynamic and ecologically sensitive. The stress caused by these ecosystems is due to multifarious anthropogenic activities. Among the stress-causing variables, microplastics are posing a serious threat to these ecosystems. Unfortunately, the information on microplastics in the Tamilnadu coastal region is very scarce. Realizing this fact, investigations have been made in the present study to evaluate the microplastics pollution in the estuarine sediments along the Tamilnadu. Three estuarine regions (Uppanar, Vellar, and Coleroon) were selected for the present study along Tamilnadu coast. The sediment samples were collected from the above stations and the level of microplastics were also quantified. The

morphology of microplastics was identified using a Fluorescence microscope. The results of this study have revealed relatively high concentrations of microplastics in sediments of the Tamilnadu coastal zone compared to those in similar areas, confirming the widespread occurrence of microplastics in the marine environment. Moreover, spatial variation in microplastic concentrations was observed on a relatively small scale. The results also suggest that freshwater rivers are a potentially important source of microplastics. Given the ubiquity and prevalence of microplastics in the marine environment further ecotoxicological investigation is warranted.

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