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# Spatial and temporal distribution of microplastic in surface water of tropical estuary: Case study in Benoa Bay, Bali, Indonesia

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#### ABSTRACT

In 2010, Indonesia estimated as the second-largest country in the world that donates plastic to the sea. This study aims to investigate the spatial and temporal distribution of microplastics in tropical estuaries. The sampling was carried out in Benoa Bay with four repetitions representing the wet and dry seasons. Spatially it was found that the highest microplastic abundance around the Suwung landfill, while the lowest at Badung River Estuary, middle, and an inlet of the Benoa Bay. The highest percentage to the lowest microplastic based on the size was  $500-1000~\mu m$  (37.9%),  $>1000~\mu m$  (35.7%),  $300-500~\mu m$  (22.1%), and  $<300~\mu m$  (4.3%), while based on the shape were fragments (73.19%), foam (17.02%), fiber (6.38%), and granule (3.40%). No significant differences were found between the wet and dry seasons based on the abundance, but significantly varied based on size and shape. Polymers of microplastics were dominated by polystyrene, polypropylene, and polyethylene.

#### 1. Introduction

Marine waste has become a concern of the central and regional governments in Indonesia (Purba et al., 2019) since the research revealed that 4.8-12.7 million metric tons (MMT) of plastic waste annually poured out into the oceans and Indonesia (0.48–1.29 MMT) is the second-largest country contributing plastic to the sea after China (1.32-3.53 MMT) (Jambeck et al., 2015). Plastic is tough to degrade (Andrady, 2015; Shah et al., 2008; Worm et al., 2017), thus making it have a long resident time in the ocean (Brandon et al., 2016) and can be accumulated for decades in aquatic ecosystems (Thompson, 2005; Thompson et al., 2004). Plastic waste can be categorized into several types based on its size, namely megaplastic (>50 cm), macroplastic (5-50 cm), mesoplastic (0.5-5 cm), microplastic (0.05-0.5 cm) (Cordova, 2019; Lebreton et al., 2018; van Emmerik et al., 2018), and nanoplastic (1–1000 nm) (da Costa, 2018; Gigault et al., 2018; Ter Halle et al., 2017). One focus of plastic waste research in the ocean is microplastics (Andrady, 2011; Purba et al., 2019).

Microplastics can be divided into primary and secondary

microplastics (Boucher and Friot, 2017). Primary microplastic is a plastic that is deliberately produced with diminutive size (Auta et al., 2017) and generally used as a cleaning agent in personal care products (Murphy et al., 2016) such as in toothpaste, facial cleansers, scrubs, bath soap, and many others (Cole et al., 2011). Secondary microplastics are derived from larger plastic fragmentation (Auta et al., 2017; Estahbanati and Fahrenfeld, 2016) caused by physical, chemical, and biological processes (Auta et al., 2017) such as exposure to the sun, wind, water, and various environmental pressures (Murphy et al., 2016). Microplastic is challenging to observe directly with the naked eye (Andrady, 2011). Microplastics are more dangerous than larger size plastics because of their microscopic size so a low trophic level can eat them, the color is similar to natural food, high abundance, and low density (Guzzetti et al., 2018; Wright et al., 2013). In addition to physical microplastics that are easily swallowed, the dangers microplastics additives such as polybrominated diphenyl ethers (PBDE), phthalates, nonylphenols (NP), bisphenol A (BPA), and antioxidants that can increase toxicity to organisms (Hermabessiere et al., 2017). These additives can be absorbed and accumulated by microplastics from polluted aquatic environments

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(Guzzetti et al., 2018). Some materials, such as heavy metal, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and dichlorodiphenyltrichloroethane (DDT) were found binding to microplastic (Purwiyanto et al., 2020; Van Cauwenberghe et al., 2015).

Microplastic properties that are easily swallowed at low tropic levels cause it can accumulate (Batel et al., 2018; Karlsson et al., 2017) in cells and tissues (Cordova et al., 2020; Sharma and Chatterjee, 2017), and can move to higher trophic levels (including to humans) through the food chain (Carbery et al., 2018). Microplastics can enter the human body through food, drink, and breathing (Vethaak and Leslie, 2016). Bio-accumulation and biomagnification will cause an increased risk and toxicity of microplastic to the top predators in the food chain (Carbery et al., 2018; Ferreira et al., 2018; Fonte et al., 2016). Furthermore, microplastics in the human body stimulate infertility, obesity, and cancer (Sharma and Chatterjee, 2017) because they can be absorbed in the spleen, veins, and even to the brain (Barboza et al., 2018). The level of human exposure to microplastics is a critical parameter that affects health (Wright and Kelly, 2017), and can cause death even in chronic conditions (Prata, 2018).

The primary sources of microplastic pollution in the sea are rivers, runoff, tides, wind, fishing gear, fish farming, and the atmosphere (Akdogan and Guven, 2019; Law and Thompson, 2014). Of all these sources, rivers are the most significant contributor to marine plastic waste, with a load between 1.15 and 2.41 million tons annually (Lebreton et al., 2017). One of the highest microplastic accumulation sites is in the estuary region (Antunes et al., 2018; Zhang, 2017). The Surface waters contain microplastics three times higher than the water column (Eo et al., 2019). The presence of microplastics in estuaries is strongly influenced by human activities, where human existence will increase microplastic pollution (Hitchcock and Mitrovic, 2019). Besides that, the presence of microplastics in an aquatic ecosystem is strongly associated with seasons (Cheung et al., 2016; Eo et al., 2019; Germanov et al., 2019; Han et al., 2020). Several studies have shown that the level of microplastic pollution in estuarine waters is very high in various parts of the world such as Shanghai estuary, China (Zhang et al., 2019), Bohai Bay, China (Wu et al., 2019), Estuary in South America (Barletta et al., 2019), an estuary in South Carolina, United States of America (USA) (Gray et al., 2018), and Solent Estuary, United Kingdom (Gallagher et al., 2016).

Research on microplastic pollution on Indonesia's surface waters has been reported quite a lot, such as in the Surabaya north coast (Cordova et al., 2019), Cilacap beach (Bouhroum et al., 2019; Syakti et al., 2017), East Nusa Tenggara Beach (NTT) (Germanov et al., 2019; Hiwari et al., 2019), Banyuurip coastal water, Gresik (Ayuningtyas, 2019), Nusa Penida, Bali, and Bentar, East Java, coastal water (Germanov et al., 2019), however information about the presence of microplastics in Indonesian estuary waters is still limited. Besides that, research on microplastics in Indonesia is incidental, and no one has reported spatially and temporally fluctuations in the estuary (Falahudin et al., 2020; Purba et al., 2019). Temporal variations of macro debris have been reported only in large rivers in Jakarta, the capital of Indonesia (Cordova and Nurhati, 2019). In contrast, microplastic spatial and temporal variations on the estuary surface water are significant to identify for obtaining a complete frame of pollution.

One estuary in Indonesia territory is Benoa Bay, Bali, as a case study of tropical estuaries in this research. Benoa Bay is an urban estuary and is significantly influenced by the activities of the surrounding population. Benoa Bay is the estuary of six rivers in Bali and already polluted by suspended material, nutrients, and heavy metals (Rahayu et al., 2017; Risuana et al., 2017; Suteja et al., 2020b; Suteja and Dirgayusa, 2018a) which originates from activities on land. Although in a polluted condition, Benoa bay is used by local people as a fishing zone for fish, bivalve, and crab. Moreover, Benoa Bay is also used as a crab and fish cultivation area by using the floating net cages (FNC) method. Benoa Bay is potentially contaminated by microplastics because there are reports that

rivers are the primary source of plastic pollution in various regions of the world, and their abundance is greatly influenced by seasons. Therefore, this study aims to investigate the spatial and temporal distribution of microplastics in surface water of tropical estuary, especially in Benoa Bay.

#### 2. Materials and methods

## 2.1. Study site description

Benoa Bay is one of the biggest estuaries in Bali Island, with an area around 1243.41 ha. In 2011, Benoa Bay was designated as a water conservation area by Presidential Decree no. 45 of 2011. Benoa Bay status changed in 2014 to utilization area, especially for reclamation activities by Presidential Decree no 51 of 2014. This status was rejected by the local community and change by the government in 2019 as a maritime conservation area through the Minister of Maritime Affairs and Fisheries Decree number 46/KEPMEN-KP/2019. Benoa Bay is surrounded by a 1002.22 ha mangrove ecosystem (Pratama et al., 2019), which belongs to the Ngurah Rai Forest Park area, and on the western side is bordered by I Gusti Ngurah Rai International Airport. Some of the land sides of Benoa Bay are used as a regional final disposal site, called Suwung landfills, from Denpasar, Badung, Gianyar, and Tabanan (SARBAGITA) district. This landfill is planned as a location for waste processing into electrical energy. In addition, Benoa Bay is also used as a water sports zone and a port (Benoa Port). Benoa Bay is an estuary of Bualu, Sama, Mati, Badung, Loloan, and Buaji rivers. This river carries pollutants from the mainland to Benoa Bay (Perwira et al., 2019; Sari and Yudha, 2019; Suteja and Purwiyanto, 2018). Previous studies have shown that Benoa Bay is in the eutrophic category base on nutrient (Suteja and Dirgayusa, 2018a), polluted category based on a saprobic index (Ananingtyas et al., 2017), and a high sedimentation process base on numerical simulation (Maharta et al., 2018). The Suspended material (Risuana et al., 2017), nutrient (Dewi et al., 2017; Raharja et al., 2018; Rahayu et al., 2017), and organic matter (Yuspita et al., 2017) concentration in Benoa Bay was reported in general it had passed the quality standard set by the local and central government. Benoa Bay is also reported to have been contaminated by chromium (Cr) in crabs, mangroves, water, sediments, and plankton (Dirgayusa et al., 2017; Suteja et al., 2020b; Suteja and Dirgayusa, 2018b). Most of the Benoa Bay area is tidal flat, and the substrate is dominated by sand (Prinasti et al., 2020; Suteja et al., 2020b).

## 2.2. Sampling collection

Field sampling is carried out based on seasonal differences (wet and dry seasons) in Indonesia. The wet (dry) season is characterized by the northwest (southeast) wind that carries a lot of (a little) water vapor (As-Syakur et al., 2011; Prasetia et al., 2013). The wet season ranges from November to April, while the dry season is from May to October (Aldrian and Djamil, 2008; As-Syakur et al., 2011; Chang et al., 2005; Hendon, 2003). Rainfall will cause an increase in river flow into Benoa Bay (Suteja and Purwiyanto, 2018). The sampling at Benoa Bay was conducted in March 2018 and April 2019 for representing the wet season, and October 2018 and 2019 for defining the dry season. Water samples were taken at eight stations (Fig. 1). Station 1 and Station 2 are located in the estuary of Mati and Badung Rivers, which represent a permanent river that empties into Benoa Bay. The length (maximum discharge) of Mati and Badung rivers is 12 km  $(46.2 \text{ m}^3/\text{s})$  and 17 km  $(25.6 \text{ m}^3/\text{s})$ , respectively (Suteja and Purwiyanto, 2018). At the downstream of the Badung River, there is a dam that is used as a source of water for local drinking water companies. Station 3 and Station 6 are located in the estuary of the Sama and Bualu Rivers which represent the seasonal rivers in Benoa Bay. The discharge from these rivers is very low and generally flows only during the wet season. Station 4 and Station 5 are located at the inlet and middle of the bay which represents the areas with the

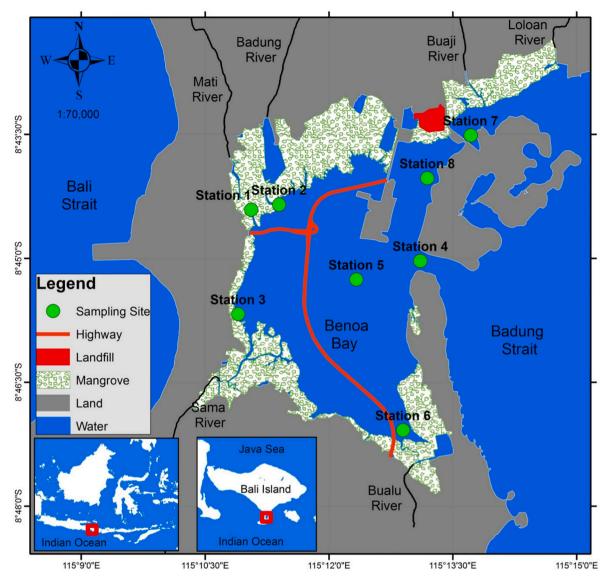


Fig. 1. Sampling location of microplastic in Benoa Bay surface water.

deepest water and fastest currents velocity in Benoa Bay. Station 4 is the main shipping channel of Benoa Harbor, while Station 5 is a ship parking area. Station 7 and Station 8 are located near a Suwung landfill which is characterized by shallow waters and calm currents. Station 7 is also adjacent to the FNC. The sampling process was done during the ebb tides to get maximum input from the mainland through the river flow. The sampling time was determined based on tidal prediction at Benoa Port, issued by Balai Riset dan Observasi Laut (BROL) and available on the website <a href="http://bpol.litbang.kkp.go.id/imro-ofs/#show\_select\_tides\_predictor">http://bpol.litbang.kkp.go.id/imro-ofs/#show\_select\_tides\_predictor</a>.

The Microplastic sampling procedure in Benoa Bay followed previous research (Herrera et al., 2020; Kang et al., 2015; Mani et al., 2015; Tamminga et al., 2018). Microplastic sampling in water using a mini manta trawl net (mesh size 300  $\mu m$ , net length 1.5 m, rectangular opening area 0.45  $m^2$ ) equipped with a flowmeter (Hydro-Bios, model 438–115) which mounted in the center of the opening. The number of flowmeter rotation at each station was recorded to calculate the volume of filtered water. To calculate the amount of filtered water was done by multiplying the opening area of the mini manta trawl net by the number of rotation at each station and the correction factor (0.3). The Mini manta trawl net was pulled around the sampling point for 20 min at 2–3 knots of ship speed. The towing duration followed previous research,

which recommended the towing period <60 min to prevent net clogging by organic and suspended material, which might be resulting in low microplastic obtained (Fischer et al., 2016; Tamminga et al., 2018). Especially of sampling point around river estuary, the towing was done in a parallel direction to the river with shorter towing time (around 5–10 min) due to the high water turbidity (base on visual observation). After the towed was complete, the mini manta trawl net is carefully rinsed by seawater from the outside and Double Distillate Deionized Water (DDDW, please see Section 2.5. Quality control and quality assurance) from inside to make sure all the marine debris goes down to the cod-end bucket. Water samples obtained in the cod-end bucket were then put into a sterile glass bottle. To ensure all the samples were recovered, the cod-end bucket is washed three times using DDDW. The water samples obtained were stored at 4  $\pm$  2 °C until further analysis.

## 2.3. Microplastic extraction

The initial stage of the laboratory process follows previous research (Kang et al., 2015; Maes et al., 2017). The water sample is separated between plastic and non-plastic material from the glass bottle. Non-plastic samples that were mostly caught in Benoa Bay were dominated by living tissue from the aquatic organism (mangrove, seagrass,

macroalgae, macro, and mesofauna). The next step was to separate large plastic samples (Mesoplastic and macroplastic). All separated samples are washed using DDDW to remove the attached microplastics. The separation process was carried out carefully using stainless-steel tweezers, and intermixed plastic was observed by the visual method (naked eye).

The next step has followed the procedure conducted by Cordova et al. (2019), namely filtering water samples using multilevel stainless steel filters (3 in. in diameter). Mesh size 5 mm was in the first level and mesh size 200  $\mu m$  was in the second level. The filtering aims to separate water from microplastic samples. The filtered sample then transferred into a sterile test tube using DDDW and dried using an oven for 24 h at 50  $^{\circ}$ C. It sample was not ready for further analysis because microplastics were still mixed with organic material, so the next step is to destruct the organic material. This destruction is done by adding 3-5 mL of hydrogen peroxide ( $H_2O_2$ ) 30% to the test tube and heated at 60 °C for 24–48 h in the water bath (Shibata water bath, model WB-6C). We choose H<sub>2</sub>O<sub>2</sub> 30% as an agent for organic material destruction because of the high recovery of the microplastic sample (85% to 91%) (Majewsky et al., 2016), compared to H<sub>2</sub>O<sub>2</sub> 6% which only had 78% of sample recovery (Sujathan et al., 2017). The H<sub>2</sub>O<sub>2</sub> 30% is the most commonly used previous studies for organic material destruction (Stolte et al., 2015; Zhao et al., 2014), although some other experiments add H<sub>2</sub>O<sub>2</sub> 50% (Tamminga et al., 2018), and H<sub>2</sub>O<sub>2</sub> 35% (Nuelle et al., 2014), and H<sub>2</sub>O<sub>2</sub> 20% (Kang et al., 2015). Furthermore, Nuelle et al. (2014) explained that a higher percentage of H<sub>2</sub>O<sub>2</sub> used would cause microplastic color fading. In this study, microplastic colors were not analyzed. After the destruction of organic matter was completed, microplastic samples were transferred to sterile filter paper (Whatman cellulose nitrate, pore size  $0.45~\mu m$ , diameters 47 mm) by vacuum method. The microplastic sample were dried using an oven for 24 h at 60 °C (de Carvalho and Baptista Neto, 2016; Kang et al., 2015).

## 2.4. Microplastic identification

Microplastic identification was divided into three categories; namely based on the shape, size, and polymer type. The shape and size of the microplastic were identified using a stereo microscope (Nikon Eclipse Ni-U) equipped with a camera (Nikon DS-L4). Microplastic identification is carried out based on the standard criteria recommended by the previous research (Cordova et al., 2019; Mohamed Nor and Obbard, 2014). The rules were: no organic structure is seen; particles have homogeneous and non-shiny colors; fiber is not segmented or branched, has a homogeneous thickness, and is not tapered at the edges. Specifically, the criteria for homogeneous and shiny plastic particles have been criticized by Mohamed Nor and Obbard (2014) because these rules insufficient and lead to miss identification of microplastic. To avoid misidentification, we applied an additional requirement of microplastic suggested by a previous study (Horton et al., 2017). The additional categories were: 1) the particles have unnaturally color, and the texture is homogenous, 2) unnaturally contrast colored particles, 3) unnatural shapes (such as perfectly rounded), 4) flexible or compressible and not fragile, 5) fibers that remain intact when handled with tweezers, and 6) particle are glassy or shiny. Horton et al. (2017) explained the particles are categorized as microplastic if they meet at least two additional criteria. The Particles that eligible as plastic were then divided into fibers, fragments, granules, and foam based on previous research (Bosker et al., 2018; Cordova et al., 2019; Syakti et al., 2018). Microplastic sizes were divided into four following main categories: <300, 300-500, 500–1000, and >1000  $\mu m$  (Cordova et al., 2019). The Microplastic size was measured using Nikon NIS-Elements software. Random particles (34 out of 235, microplastic particles found) were chosen to confirm the chemical composition of microplastics. Microplastic polymer type was identified by using µFTIR spectroscopy (JASCO FTIR Microscopes IRT-7200 VC with FT/IR-6800FV Full-vacuum System). The FT-IR is used for polymer analysis because it can analyze samples directly (Käppler

et al., 2015). FT-IR is operated based on the experimental setup of Löder and Gerdts (2015), in a single reflection mode with resolution 8 cm, range of 600 and  $3800 \text{ cm}^{-1}$ , and 32 scans per analysis.

## 2.5. Quality control and quality assurance

This procedure is done to ensure the accuracy of the data obtained (Mai et al., 2018). Before using µFTIR spectroscopy, a comparison was made between library data on the  $\mu FTIR$  spectrum, as well as between particles found with polymer standards from Thermo-scientific, Shimadzu, and plastic standards from the Research Center for Geosciences, University of Bayreuth. In this study, the field blank was not carried out, but we applied contamination prevention during the field sampling process. To minimize cross-contamination during field sampling, we wore latex gloves and clothes made from 100% cotton. We also placed the manta net beside the boat (port or starboard side, close to the bow) during the towing process to prevent vessel-based contamination. Another goal was to reduce contamination from the bottom waters due to the sediments resuspension by boat propellers. For reducing crosscontamination between stations, the net and cod-end buckets were rinsed separately before the next towing by using DDDW. To minimize contamination, during the extraction and microplastic observations in this study, following the procedures carried out by previous studies such as using latex gloves, laboratory clothing made of 100% cotton, using a clean and closed room (Falahudin et al., 2020; Lusher et al., 2015; Mai et al., 2018; Mani et al., 2015; Wesch et al., 2017). Besides that, all laboratory equipment (sample bottles, test tubes, tweezers, and filters) used are made of non-plastic material (glass or stainless steel). All laboratory equipment is sterilized before use, samples are kept in a closed condition when not analyzed, and used the DDDW during all process. All of these procedures are in line with previous research (Cordova et al., 2019; Mai et al., 2018; Nuelle et al., 2014). The DDDW is reverse osmosis water produced from a multilevel distillation process, and the electricity ion content is removed. The DDDW then filtered using a sterile filter paper before use to ensure there is no contamination. To control contamination from the air, particularly from synthetic fiber materials (Chen et al., 2020; Dris et al., 2016), we applied a blank procedure during laboratory analysis (Falahudin et al., 2020; Torre et al., 2016). The filter paper was then observed under a microscope after all laboratory processes are complete. From blank procedure observations, there were no microplastics found in filter paper, which meant there was no contamination during the laboratory processes.

## 2.6. Statistical analysis

Microplastic statistical analysis was performed using Excel and PAST3.5 software (Hammer et al., 2001). Microplastic abundance in this presentation is presented in the form of average values and standard deviations. To observed variations between stations, seasons, and sampling period, the Kruskal-Wallis and Mann-Whitney pairwise post hoc non-parametric tests with a significant level at 0.05 were performed. The data was spatially mapped using QGIS 3.10.7.

## 3. Result and discussion

# 3.1. Microplastic abundance

Observations at Benoa Bay showed microplastics were found at all stations (Fig. 2) with different abundances at each sampling period (Table 1, Table S1). Based on non-parametric tests, it was found that the microplastics abundance in Benoa Bay was significantly different between all stations. The significant difference (Mann-Whitney pairwise post hoc with a significant level of p < 0.05) mainly originated from Stations 7 and 8, near the Suwung landfill. The location of the minimum abundance of microplastic in Benoa Bay was found in a different location at each sampling period. Minimum abundances in March 2018

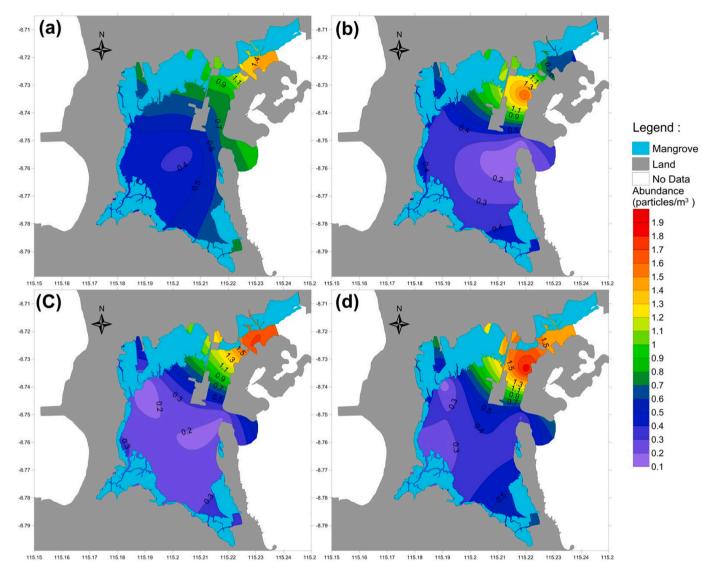


Fig. 2. Microplastic abundance in March 2018 (a), October 2018 (b), April 2019 (c) and October 2019 (d) at Benoa Bay surface water.

**Table 1** Microplastic abundance in Benoa Bay.

No	Sampling period	Microplastic abundance (particles/m³)			
		Minimum	Maximum	Average	Standard deviation
1	March 2018	0.36	1.41	0.69	0.32
2	October 2018	0.15	1.65	0.54	0.47
3	April 2019	0.13	1.71	0.54	0.57
4	October 2019	0.11	1.88	0.70	0.63
5	Wet season	0.13	1.71	0.61	0.46
6	Dry season	0.11	1.88	0.62	0.55
7	Overall	0.11	1.88	0.62	0.50

(0.36 particles/m³) and October 2018 (0.15 particles/m³) were found in the central and inlet of Benoa Bay, respectively. It is because the two locations are far from the river estuary, which is the primary source of plastic pollution in the ocean (Lebreton et al., 2017). These results were in line with previous studies in the Yangtze estuary, China (Zhao et al., 2014), and the Yellow River Estuary, China (Han et al., 2020) which found that areas far (close) to river estuary have a low (high) microplastic abundance. The low microplastic abundance in areas far from river estuary is also caused by microplastic sinking into sediments due to

heteroagregation with fine suspended material in the water column (Bakir et al., 2014; Besseling et al., 2017; Leslie et al., 2017). Heteroaggregation will reduce microplastic abundance in the water column (Besseling et al., 2017). Previous studies conducted by Risuana et al. (2017) conclude that the highest suspended material in Benoa Bay was observed in the estuary area. Furthermore, numerical simulations showed that the highest sedimentation process in Benoa Bay occurs in Mati and Badung rivers estuary (Maharta et al., 2018). The low abundance of microplastics in the central and inlet of Benoa Bay was also affected by the sampling time. The sampling time of this research was carried out at ebb, which means that the water density was lower (Suteja et al., 2020a) so that the microplastics were easier to sink around the river estuary. The opposite condition occurs at the flood.

The location of minimum abundance obtained in 2019 was different from 2018. Minimum abundance in April 2019 (0.13 particles/m³) and October 2019 (0.11 particles/m³) was obtained at the Badung River estuary (Table S1). It is due to the low discharge of the Badung River, which flows into Benoa Bay. There is a dam at the Badung river mouth utilized as the main source of clean water for southern Bali's regional drinking water company (Suteja and Purwiyanto, 2018). Microplastic abundance, which is influenced by freshwater input from rivers, has been explained by previous studies. For example, Lima et al. (2015) found that microplastic abundance in the Goiana estuary, Northeast

Brazil, decreases with decreasing water discharge. When the water discharge decreases, the water flow also decreases so that deposited microplastics in sediment are not resuspended (Hurley et al., 2018) and cause the low abundance of microplastic in the waters (Eo et al., 2019; Lima et al., 2014). A study conducted by Gündoğdu et al. (2018) in the Northwest Mediterranean Sea concludes that the microplastic abundance increased 14 times during floods, which triggered by heavy rains. Geographically, the Badung river estuary is close to a Mati river estuary, but at ebb, there is no garbage transfer between estuaries. It is due to the presence of dense mangrove vegetation between the two stations (Pratama et al., 2019), thus preventing plastic waste movement (Balas et al., 2001; van Emmerik et al., 2019). Moreover, at low tide, the two estuaries are separated by a tidal flat.

The highest microplastic abundance  $(1.41-1.88 \text{ particles/m}^3)$  at the surface of Benoa Bay waters during the sampling period was found around the Suwung landfill (Stations 7 and 8). We suspect that the Suwung landfill is a primary source of microplastic pollution in the surrounding area. It is supported by previous research, which states that landfill is not the final sink for plastic but instead becomes a source of microplastic pollution (He et al., 2019) especially those flows into the marine environment (Kazour et al., 2019). It is estimated that about 227 million microplastics per day were released from landfills into the marine environment, and its abundance increases with decreasing distance from the landfill (Kazour et al., 2019). The main routes of microplastics transfer from landfill to aquatic environments are from the airborne and leachate (Chen et al., 2020; Huang et al., 2020; Lassen et al., 2015). Research conducted in China shows that leachates contain microplastics between 420 and 24,580 particles/m<sup>3</sup> in the Shanghai, Wuxi, Suzhou, and Changzhou landfills (He et al., 2019) and between 4000 and 13,000 particles/m<sup>3</sup> in the Laogang Landfill (Su et al., 2019). Further explained by Su et al. (2019) that the mechanism of polymer degradation, the amount of plastic present in the landfill, and the lifetime of wasted plastic products were strong influences microplastics abundance in the landfill. Specifically, for the Suwung landfill, we suspect that microplastic influx into seawater through leachate water discharges, which located in the mangrove ecosystem. The pollution from landfills is also indicated by the number of dead mangroves (the color turns brown) around the leachate channel from the Suwung landfill. Airborne microplastics in landfill originated from macroplastic degradation that flew into the air (Chen et al., 2020; Dris et al., 2016) or from imperfect plastic waste incineration (Liu et al., 2019b). The wind is an essential factor that transfers microplastics from soil to air (Liu et al., 2019a), and transports them to other areas (Prata, 2018), such as mountainous areas (Allen et al., 2019) or waters environments (Chen et al., 2020). However, the information of airborne microplastics influence from landfill to the surrounding environment (especially in the estuary area) is very limited and requires more in-depth study. The input of marine waste from landfills is a serious problem and must be addressed immediately because recently, there has not been a single country that makes regulations that microplastic is one of the pollutants from the landfill (Kazour et al., 2019).

Microplastic abundance at Benoa Bay surface water between the sampling periods was not significant variation based on non-parametric statistical tests. The average abundance of microplastics from the lowest to the highest sequentially is October 2018 and April 2019 (0.54 particles/m³) < March 2018 (0.69 particles/m³) < October 2018 (0.70 particles/m³). For calculating microplastic abundance in each season, averaging was done in March 2018 and April 2019 for the wet season, while the average October 2018 and 2019 for the dry season. The average microplastics abundance in the wet season (0.61 particles/m³) and dry season (0.62 particles/m³) are similar. There is no significant difference based on non-parametric statistical tests between seasons. It is due to the abundance of microplastics on the surface water of Benoa Bay not affected by rainfall and river flushing but influenced by the input from the landfill. Its results were supported by previous research in the Clyde and Hunter estuary, Australia, which did not get a significant

correlation between rainfall and river flushing with microplastic abundance in surface waters (Hitchcock and Mitrovic, 2019). Rodrigues et al. (2018) also explained that changes in rainfall intensity during the sampling process cause its influence on microplastic abundance can be ignored. The influence of seasons (rainfall) on microplastic abundance varied between regions. Research conducted in Nusa Penida Island, Bali, Indonesia (Germanov et al., 2019), Pearl River Estuary, Hong Kong (Cheung et al., 2018), Goiana Estuary, Brazil (Lima et al., 2014), Tamsui River, Taiwan (Wong et al., 2020), and the Nakdong River, South Korea (Eo et al., 2019) found that the highest microplastic abundance was obtained during the wet season and vice versa during the dry season. Lima et al. (2014) explained that the high microplastics abundance in the wet season due to the increase of river flow, which carries many plastic fragments from upstream to the estuary. The opposite results were obtained in the Maozhou River, Macao (Wu et al., 2020b), Antua River, Portugal (Rodrigues et al., 2018), the Yellow River estuary, China (Han et al., 2020), and the southern part of Yellow Sea (Jiang et al., 2019) which got the highest abundance of microplastic during the dry season and vice versa during the wet season. It is because the increase of rainfall will increase the river flux (Zhao et al., 2019), and dilute the microplastic abundance in the waters (Yan et al., 2019).

#### 3.2. Microplastic abundance compared with the previous study

The microplastic abundance in Benoa Bay during the wet season (0.61 particles/m<sup>3</sup>), the dry season (0.62 particles/m<sup>3</sup>), and overall (0.62 particles/m<sup>3</sup>) had different values compared to previous studies. Microplastic abundance in Benoa Bay is much lower than that reported in the Yellow River Estuary, China (Han et al., 2020), small-scale estuaries in Shanghai, China (Zhang et al., 2019), Changjiang Estuary, China (Xu et al., 2018; Zhao et al., 2019), Yondingxinhe Estuary, China (Wu et al., 2019), Yangtze Estuary, China (Zhao et al., 2014), the southern part of Yellow Sea, China (Jiang et al., 2019), Tampa Bay, Florida, USA (McEachern et al., 2019), dan Río de la Plata estuary, South America (Pazos et al., 2018), and the northern coast of Surabaya, Indonesia (Cordova et al., 2019) (Table 2). We suspect that two factors cause a high abundance of microplastics in these locations. The first factor was the high level of pollution. High microplastic abundance is mostly reported from China, which is based on previous studies declare as the primary contributor of plastic waste from land to sea (Jambeck et al., 2015). The second factor is that the sampling device was used. The area that has a high microplastic abundance (Table 2) did not use the nondiscrete device (pump, plankton net, or manta net) but instead used a discrete device (Niskin bottle, Van Dorn water sampler, rosette, or bucket). The net mesh size also determines the effectiveness of a nondiscrete device for filtering. The finer mesh size is generally used in the pumping method (Cutroneo et al., 2020). The non-discrete tool still allows smaller particles than the mesh size to escape during field sampling, while the discrete method would collect all microplastic sizes and shapes (Cutroneo et al., 2020; McEachern et al., 2019; Wu et al., 2020a). A study in Tampa Bay, USA, which used two different sampling devices, showed that the discrete method collects microplastic abundance 230 times higher than the non-discrete tool (McEachern et al., 2019). Similar results were also obtained by other studies that conclude microplastic abundance from the bulk sample was four times higher (Song et al., 2014), and the grab samples were three times higher (Barrows et al., 2017) compared to the non-discrete method. However, it should be noted when using the discrete device that the sample volume is crucial for microplastic abundance. It can be seen in Table 2 that the higher of sample volume, the lower microplastic abundance. It was also confirmed by Kanhai et al. (2017), which only got 1.1 particles/m<sup>3</sup> of microplastic by filtering 2000 L water samples.

The average abundance of microplastics in Benoa Bay, using the same method (net-based), was higher than that obtained by surface water in Goiana Estuary, Brazil (Lima et al., 2014), United Kingdom offshore water (Maes et al., 2017), English Channel (Cole et al., 2014),

**Table 2** Microplastic in surface water from various locations in the world. The abundance with particles/L unit in some reference converted to particles/ $m^3$  units. DS (dry season), WS (wet season), and O (overall).

DS (dry season), WS (we			
Location	Sampling device	Abundance (particles/m³)	References
South Yellow Sea, China	Pump, 100 L, 50 μm	Mean: $6500 \pm 2100 \; (DS)$ Mean: $4500 \pm 1800 \; (WS)$	(Jiang et al., 2019)
Pearl River Estuary, Hong Kong	Manta net, 333 μm	Mean: 0.256 (DS) Mean: 6.124 (WS)	(Cheung et al., 2018)
Nusa Penida waters, Bali, Indonesia	Small round net, 200 μm	Mean: 3.973 (O) Range: 0.04–0.06 (DS) Range: 0.56–0.90 (WS)	(Germanov et al., 2019)
Maozhou River, Hong Kong	Stainless-steel bucket, 5 L	Range: 4000–25,500 (DS) Range: 3500–10,500	(P. Wu et al., 2020)
Near estuary Yellow River, China	Stainless-steel bucket, 5 L	(WS) Mean: 930,000 (DS) Mean: 497,000 (WS)	(Han et al., 2020)
Small-scale estuaries in Shanghai, China	Stainless-steel bucket, 5 L	Mean: 27,840 ± 11,810	(Zhang et al., 2019)
Changjiang Estuary, China Changjiang Estuary,	Pump, 100 L, 60 μm Pump, 100 L,	Mean: 45.4–122.8 Mean: 231 ±	(Zhao et al., 2019) (Xu et al., 2018)
China Haihe Estuary, China	70 μm Pump, 20 L, 48 μm	182 Mean: 1485.7 $\pm$ 819.9	(Wu et al., 2019)
Yondingxinhe Estuary, China	Pump, 20 L, 48 μm	Mean: 788.0 ± 464.2	(Wu et al., 2019)
Yangtze Estuary, China	Pump, 20 L, 32 μm	Mean: 4137.3 $\pm$ 2461.5	(Zhao et al., 2014)
Pearl River estuary, China	Water sampler, 20 L	Mean: 8902	(Yan et al., 2019)
Tampa Bay, Florida, USA Costal Water of	Van Dorn sampler HDPE Bottle,	Mean: 940 $\pm$ 520 Mean: 490	(McEachern et al., 2019) (Cordova et al.,
Surabaya, Indonesia Río de la Plata estuary,	20 L Bucket, 100 L	Mean: 139	2019) (Pazos et al.,
South America Coastal water of southern California,	Manta trawl net, 300 μm	Mean: 8	2018) (Moore et al., 2002)
USA Tampa Bay, Florida, USA	Plankton net, 330 μm	Mean: 4.5 $\pm$ 2.3	(McEachern et al., 2019)
Eastern Mediterranean Basin	Manta trawl net, 52 μm	Mean: 6.7	(Kazour et al., 2019)
Goiana Estuary, Brazil	Plankton net, 300 μm	Mean: 0.26	(Lima et al., 2014)
Pacific Ocean  Urban Estuary in South	Manta net, 330 μm Plankton net,	Mean: $0.13 \pm 0.11$ Ranges: $1.0-7.0$	(Mu et al., 2019) (Naidoo et al.,
Africa United Kingdom offshore water. North-	300 µm Manta net, 333 µm	Mean: 0.14	2015) (Maes et al., 2017)
East Atlantic English Channel	Plankton nets, 200 µm and 500 µm	Mean: 0.27	(Cole et al., 2014)
Tamar Estuary, Southwest England	Manta net, 300 μm	Mean: 0.028	(Sadri and Thompson, 2014)
Mediterranean Coast of Israel Coastal water of Portuguese Benoa Bay, Bali, Indonesia	Manta net, 333 μm Neuston net, 280 μm Mini Manta net, 300 μm	Mean: 7.68 ± 2.38 Ranges: 0.01-0.32 Mean: 0.62 (O) Mean: 0.62 (DS) Mean: 0.61 (WS)	(van der Hal et al., 2017) (Frias et al., 2014) This study

Tamar Estuary, England (Sadri and Thompson, 2014) and the Portuguese Coast (Frias et al., 2014) (Table 2). However, the average abundance of microplastics in Benoa Bay was lower than that obtained in coastal waters of Israel (van der Hal et al., 2017), an urban estuary in South Africa (Naidoo et al., 2015), northeast Basin of Mediterranean Sea (Kazour et al., 2019), Tampa Bay, USA (McEachern et al., 2019), Pearl River estuary, Hong Kong (Cheung et al., 2018), and Southern California Coastal waters, USA (Moore et al., 2002) (Table 2). The difference in microplastic abundance was greatly influenced by the pollution level, sampling location, and water hydrodynamics in each study area.

## 3.3. Microplastic size

Microplastic is a synthetic solid particle that is difficult to dissolve in water and has a regular or irregular shape with 1–5000  $\mu m$  in size (Frias and Nash, 2019). In this study, microplastics were divided into four categories based on size, namely <300, 300-500, 500-1000, and >1000  $\mu m$ . The microplastic size was significantly varied between the sample median based on the non-parametric statistic tests. Overall, microplastics at Benoa Bay surface water was dominated by sizes of 500-1000 um (37.9%), while the sizes <300 um was the lowest percentage (4.3%). Furthermore, the percentage of microplastic categories 300-500 and >1000 μm in size were 22.1% and 35.7% respectively in the whole sampling period. The dominant microplastic sizes between stations in each sampling period were different (Fig. 3, Table S1). It is the difference was also found between the wet and dry seasons. In the wet season (dry season), plastic is dominated by category 500–1000 μm  $(>1000 \mu m)$  in size, with a percentage of 47.71% (43.65%). The dominant percentage of large microplastic size is Benoa Bay also strengthens the possibility of microplastic airborne contributions from landfills since the bigger sizes are more challenging to blow to more distant areas. However, this possibility needs to be proven by further studies. Microplastic size <300 µm has the smallest percentage either during the wet (1.83%) or dry (6.35%) seasons. It is related to the use of manta net mesh size of 300 µm, which allows smaller size microplastics to escape from the net during field sampling. Cole et al. (2011) explained that the smaller microplastic size, the higher possibility of being swallowed by organisms and accumulate in the food chain. The difference in microplastic size of marine debris was strongly related to the wind direction and speed (González-Hernández et al., 2020) the presence of biofouling (Leiser et al., 2020), and the hydrodynamic conditions of the waters (Zhang, 2017) such as currents, wave, tides (Jasmin et al., 2019; Kubota, 1994; Zhang et al., 2020).

Based on the particle size category, the percentage of microplastic size in Benoa Bay was larger, smaller, or even the same as other previous studies. For example, the microplastics in this study were larger than those obtained at the surface of the yellow river estuary water, China, which was dominated by size  $<200 \mu m$  (87.94%) (Han et al., 2020). In addition, the microplastic sizes between 45 and 200 µm also dominate (68-73%) in the Hunter, Clyde, and Bega estuaries, Australia (Hitchcock and Mitrovic, 2019). This study was in line with previous studies that obtained microplastics 500–1000  $\mu m$  in size dominating the small-scale estuaries in Shanghai, China (Zhang et al., 2019), Changjiang Estuary, China (Zhao et al., 2019), Goulburn river, Australia (Nan et al., 2020), and the northern coast of Surabaya, Indonesia (Cordova et al., 2019). The percentage of the dominant microplastic size in this study was smaller than that found in the sediment of Ciwalengke River, Indonesia, in which 72% has a size of  $>1000 \, \mu m$  (Alam et al., 2019). It is because an increase of microplastic size will be in line with the density so that it will sink into sediments easier (Nizzetto et al., 2016).

# 3.4. Microplastic shape

Microplastic shapes in this study were divided into fibers, fragments, granules, and foam. In general, this study was found that the percentage of the highest to lowest microplastic shape in Benoa Bay was fragment

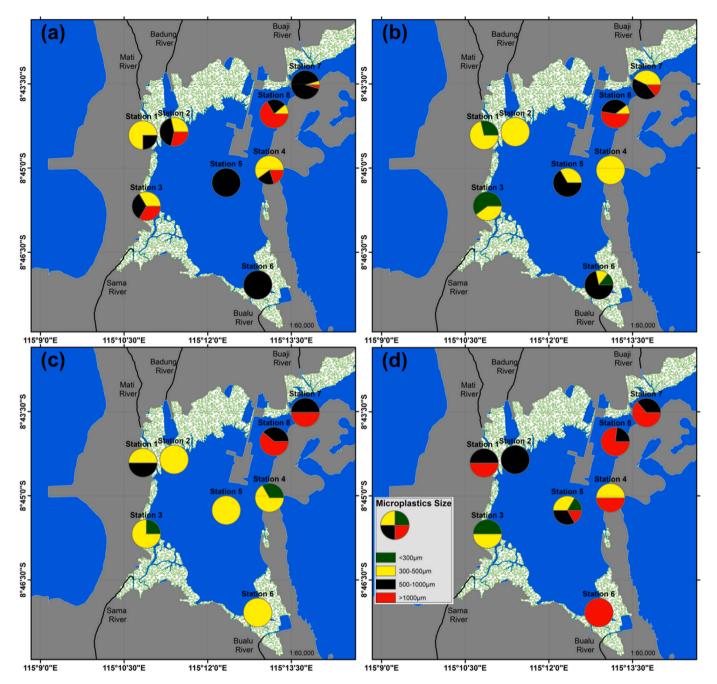


Fig. 3. Microplastics sizes  $<300 \, \mu m$  (green),  $300-500 \, \mu m$  (yellow),  $500-1000 \, \mu m$  (black), and  $>1000 \, \mu m$  (red) in March 2018 (a), October 2018 (b), April 2019 (a) c) and October 2019 (d) at Benoa Bay surface water. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(73.19%) > foam (17.02%) > fiber (6.38%) > granule (3.40%). Specifically, the percentage of fragments, foam, fiber, and granules was 69.72%, 18.35%, 8.26%, and 3.67% in the wet season and 76.19%, 15.86%, 4.76%, and 3.17% in the dry season, respectively. Fragments were the dominant shape of microplastic in both the wet and dry season, and each sampling period (Fig. 4, Table S1). Statistically, the microplastic shape was significantly varied between the sample median. The high fragment shape in an aquatic environment indicates that microplastics are produced from waste originating from population activities (Cordova et al., 2019) either through rivers or landfills. It is supported by previous studies that found that TPA leachate is dominated by fragment-shaped particles (He et al., 2019). The fragment is a microplastic shape formed from macroplastic fragmentation due to weather, mechanical processes (Barnes et al., 2009), and the incineration process

in landfills (He et al., 2019; Morét-Ferguson et al., 2010). The foam was the second dominant microplastic shape in the Benoa Bay surface waters. Foam is thought to originate from Benoa Harbor's activity as a fish landing, and foam is widely used as a fish storage container. Another foam source was styrofoam, which is intensively used in FNC in Benoa Bay. The granule is the smallest percentage shape of microplastic in Benoa Bay. It is because microplastics with a granule (round) shape were sinking faster than irregular shapes (Kaiser et al., 2019). We suspect that these granules will be found abundantly in Benoa Bay sediments, but this hypothesis needs further research.

The dominance of the microplastic shape in surface waters is related to the sampling methods and its surrounding activities. Sampling using a net allows fiber particles to escape the filtering process, so they are not filtered optimally, and the abundance is underestimated (Liu et al.,

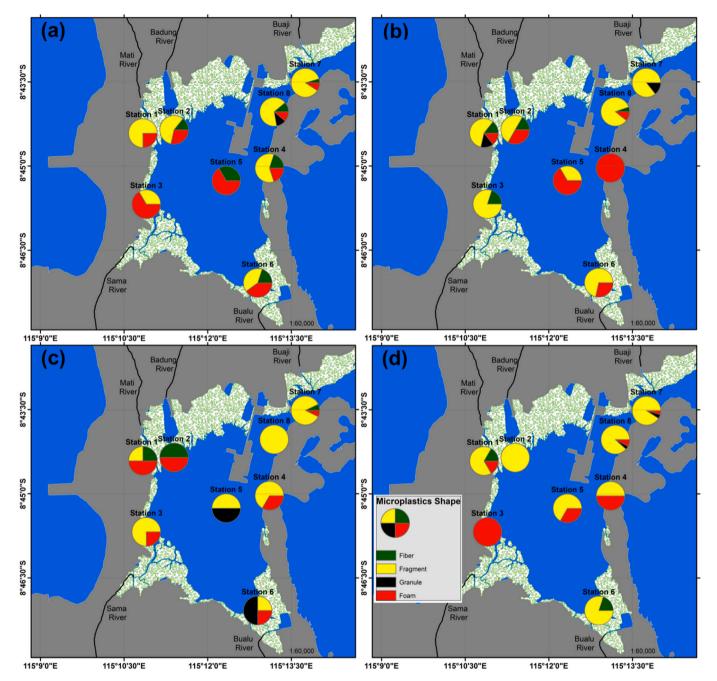


Fig. 4. Microplastic shapes of fiber (green), fragment (yellow), granule (black), and foam (red) in March 2018 (a), October 2018 (b), April 2019 (c) and October 2019 (d) at Benoa Bay the surface water. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2020). Foam shape was most commonly found in Chinese waters because they were widely used as food wrappers (Cheung et al., 2018), storage media, and fish transport (Liu et al., 2020). Granule (pellet) was the dominant shape found in areas where many residents use beauty or personal care products (Tsang et al., 2017). The spherical shape in microplastics was rarely created from large plastic degradation (Isobe, 2016), but mostly produced from primary microplastics (Tsang et al., 2017). Meanwhile, the fiber generally comes from the washing clothes process (Abidli et al., 2018; Hernandez et al., 2017).

The shapes of surface microplastic in Benoa Bay were in line with and contradicted with previous studies. Hitchcock and Mitrovic (2019) found a fragment was the dominant microplastic observed in the Hunter, Bega, and Clyde estuaries with an average percentage of 66%, 77%, and 72%, respectively. Liu et al. (2020) noted that fragment forms had the highest percentage (52%) compared to fiber (29%) and foam (19%) in

Jiaozhou Bay, China. Fragments were also reported as the most dominant shape of microplastic (63%) compared to pellets, films, foam, and lines on the surface water of the northeastern Atlantic ocean (Maes et al., 2017). However, the microplastic shape in Benoa Bay is different from previous studies in the Pearl River estuary (Cheung et al., 2018), and the coast of Guangdong, southern China (Fok et al., 2017) which got foam as the dominant shape. In addition, Pellet is the most common shape (96.8%) on the surface seawater of Hong Kong (Tsang et al., 2017).

## 3.5. Microplastic Polymer

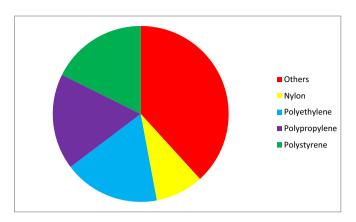
The identification of polymer was not in all samples but done by taking random microplastic particles. The total microplastic samples found in Benoa Bay were 235 particles, but the polymer type was identified by  $\mu FTIR$  spectroscopy only in 34 (14.47%) particles.

Identification of microplastic polymers in Benoa Bay (Fig. S1) showed that polypropylene (17.6%), polyethylene (17.6%), and polystyrene (17.6%) were the most dominant types of polymer found (52.9%) (Fig. 5). The other components (38.3%) were a combination of nylon, polybutadiene, poly-2-decyne, poly-vinyl chloride, polybutylene terephthalate, poly-2-butylene, poly hydroxyethyl ethylene oxide, chlorinated polyolefin resin, polyoxyethylene, and polyurethanes with percentages of 8.8%, 5.9%, 5.9%, 5.9%, 5.9%, 2.9%, 2.9%, 2.9%, 2.9%, and 2.9%, respectively (Fig. 5). Polypropylene, polyethylene, and polystyrene were the most commonly obtained because these polymers were widely used in everyday life. For example, polyethylene was widely used as a bottle or bottle cap, while polypropylene and polystyrene were widely used in the textile industry and product packaging (Avio et al., 2017). The presence of polypropylene, polyethylene, and polystyrene in Benoa Bay indicates that the waste originates from population activities (Liu et al., 2020), and can be classified as secondary microplastic (Cordova et al., 2019). The indication of waste from population activities is strengthened by the dominance of fragment and foam (secondary microplastic) compared to granule (primary microplastic).

The results of this study were in line with previous studies that found that >50 microplastics found in water and sediments of the Antuã river, Portugal was polyethylene and polypropylene (Rodrigues et al., 2018). The predominance of polystyrene, polyethylene, and polypropylene, were also found on surface water microplastics in Surabaya, Indonesia (Cordova et al., 2019), and the Tunisian Coast (Abidli et al., 2018). Polyethylene in microplastics was found in large quantities (82%) in the Changjiang River estuary, China (Xu et al., 2018). The opposite result was found in the Pearl River estuary, China, which observed that polyamide dominated microplastic particles (Yan et al., 2019). Polyamide is widely used as a fishing string or food wrapper (Naji et al., 2017). The variations of microplastic polymers found in waters are strongly related to plastics uses on land or ocean activities.

# 4. Conclusion

This research has succeeded in describing the spatial and temporal variations of microplastic in tropical estuaries. Spatially, it was found that the lowest microplastic abundance in 2018 was found in the inlet and middle areas of Benoa Bay. It is because the locations were far from the river estuary, which is the primary source of plastic pollution in the ocean. The lowest microplastic abundance in 2019 was found in the Badung River estuary due to the water discharge from the dam being closed. The highest microplastic abundance (1.41–1.88 particles/m<sup>3</sup>) at the Benoa Bay surface water during the sampling period was found around the Suwung landfill. This landfill is strongly suspected of providing microplastic into the surrounding area through airborne and leachate. In Benoa Bay, it was found that there was no significant difference between the average abundance of microplastic in the wet (0.61 particles/m<sup>3</sup>) and dry season (0.62 particles/m<sup>3</sup>). The influence of seasons (rainfall) on microplastic abundance is still widely debated by experts. Some studies find high microplastic abundance during the wet season and low during the dry season, but the opposite was found in other areas. An interesting note was found when comparing the abundance of microplastics in the Benoa Bay area with other regions. Research that uses discrete device got a greater abundance of microplastics than the non-discrete. The non-discrete device still allows smaller particles than the mesh size to escape during field sampling. The highest to the lowest percentage of microplastic based on the size in Benoa Bay was 500–1000  $\mu m$  (37.9%), >1000  $\mu m$  (35.7), 300–500  $\mu m$ (22.1%), and  ${<}300~\mu m$  (4.3%), while based on the shape was fragments (73.19%), foam (17.02%), fiber (6.38%), and granule (3.40%). The low percentage of microplastic <300 µm was presumably due to the use of net a 300  $\mu m$  in mesh size. The high percentage of fragment shape in an aquatic environment indicates that microplastics are produced from population activities, either through rivers or landfills. Investigation of



**Fig. 5.** Microplastic polymer of polystyrene (blue), polypropylene (purple), polyethylene (green), nylon (yellow), and other (red) at Benoa Bay water surface water. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

polymer types using  $\mu$ FTIR spectroscopy shows that 52.9% of microplastics in Benoa Bay are dominated by three polymers type (polystyrene, polypropylene, and polyethylene). Its polymer is widely used in various food and beverage wrapping products and the textile industry.

## CRediT authorship contribution statement

Yulianto Suteja: Conceptualization, Investigation, Visualization, Data curation, Formal analysis, Validation, Writing – original draft. Agus Soleh Atmadipoera: Validation, Writing – review & editing. Etty Riani: Validation, Funding acquisition, Writing – review & editing. I. Wayan Nurjaya: Validation, Writing – review & editing. Dwiyoga Nugroho: Validation, Writing – review & editing. Muhammad Reza Cordova: Conceptualization, Investigation, Funding acquisition, Resources, Methodology, Formal analysis, Validation, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix B. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:https://doi.org/10.1016/j.marpolbul.20 21.111979. These data include the Google map of the most important areas described in this article.

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