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Characteristics of Microplastics in Sediment at Danang Beaches - Vietnam

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Abstract

Microplastics (MPs) has been recently attracted the attention of many researches due to their existence for a long time in the environment. MPs are considered as an alarming pollution issue such as their occurence in water, sediment, and organism. In this research, the characteristics of MPs such as their concentration, size, shape and composition of the polymers in the shoreline sediments at the two layers (0- 5 cm and 5 -10 cm) collected from seven beaches on Danang city were investigated by the Fourier-transform infrared (μ-FTIR) method. Consequently, the average concentration of MPs was found at 5565 ± 2359 items kg^{-1}dry weight (d.w.) in two layers. The upper layer of beach sediment (0-5 cm) has the majority of MPs with average abundance of 7679 ± 3363 items kg^{-1}d.w. MPs had a wide range of sizes from 22.4 μm to 2797.2 μm with an average of 93.4 ± 18.8 μm in sampling sites. In addition, the range size from 20 to 150 μm was the most dominance in both layers.The morphology of MPs was found in the sediment with the most diversity of fragment. The variation of the polymer types of the MPs in two sampling layers was determined. Polyamide (PA) was the main polymer type found in both sediment layers while polyether sulfone (PES) was observed only at 4/7 sampling beaches.These results showed the diversity of the MPs in the two sediment layers of the seven beaches in Danang city and it is necessary to study on MPs pollution in sediment on beaches of Vietnam in future.

Keywords: Microplastics (MPs), Danang beaches, morphology, sediment, polymer

1. Introduction

Plastics have been used in numerous applications, ranging from household and personal goods, clothing, and packaging [1]. About 3 billion tons of plastic were synthesized in 2016, and 8 million tons of plastics will eventually enter the marine environment annually [2]. Plastics are defragmented into small pieces of plastics in the environment by weathering, time, and natural processes. Plastic debris can be formed in all shapes, sizes, and their dimensions ranging from 1 μm to 5000 μm (5 mm) defined as microplastics (MPs) [1]. Through pathways of environmental activities, ingestion, and inhalation, MPs can enter the water, sediment, and organisms and even recently they were detected in human blood [1]. Thus, MPs can cause adverse impacts on the environment and human beings [3]. The biomagnification and bioaccumulation processes might potentially impact to organism species and people in the ecosystems by the food chains and could pose toxic threats to the environment and human health [1].

Beaches and shorelines accumulated a high quantity of macro - and microplastics, which originated from both inland and seaside anthropogenic activities [4]. Shoreline locations of plastic debris, particularly beaches were habitats that concentrated the breakdown of macroplastics into MPs [5]. Furthermore, the beach sediments were remarkably impacted by both land and sea-based plastic debris, hence a large number of MPs was presented [4, 5]. Gall and Thomson (2015) reported that at least 690 marine species were documented to ingest or entangled in plastic and MPs, at least 17% of those were designated as vulnerable and near threatened. Therefore, study of MPs in the beach habitats was essential to assess both accumulation and occurrence of microplastic waste [6].

A huge amount of research on MPs and plastic debris in the shoreline sediment has been conducted in America and many European, North and South Pacific, Atlantic, Mediterranean, and Hong Kong countries [7]. Some researches in Asia countries showed that MPs concentrations were 5000-8714 items kg^{-1} sediment at Beibu Gulf - China; 1900 items kg^{-1} sediment at Tokyo Bay - Japan; large MPs (1-5 mm): 0-2088 items m^{-2}, and small MPs (0.02-1 mm): 1,400-62,800 items m^{-2}

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at 20 sandy beaches in South Korea and 36.8 ± 23.6 items. kg^{-1} sediment in Singapore [8].

In Vietnam, up to now studies on MPs and plastic debris in the coastal environment and shorelines were still limited. Characterisation of MP debris in three beaches of My Khe, T20 and Son Thuy of Danang was determined from 1,460±758 to 29,232±2,577 items/kg dry sediment. MP sizes less than 150 µm were accounted for the highest proportion of 77.83% at Son Thuy, 87.96% at T20, and 65.91% at My Khe beach. Three dominant polymers were determined as PTFE (Polytetrafluoroethylene (Teflon)), EVOH (Ethylene vinylalcohol), and PA (Polyamide (Nylon)) [9]. In previous research, characteristics of MPs in shoreline sediments from eight beaches in Danang were investigated with MP concentration of 9,238±2,097 items/kg dry weight (d.w). Fiber was the predominant type of microplastics, accounting for 99.2% of the total items [10].

Therefore, microplastic pollution has become a major issue that significantly affects on the coastal ecosystem. In addition, some previous techniques of MPs determination also lacked data of the types of polymers detected in MPs due to limitted techniques for MPs annalysis. MP polymers were basis to look for resources of the MPs pollution and their existence is source of MPs pollution in environment. Thus, it is very necessary to explore the current state of the MPs pollution in all environmental objects such as MPs in water, sediment and organism…Therefore, in such context, this study aims to identify the characteristics of MPs, including their concentration, size, shape and polymer types in sediments collected from seven beaches in Danang city, Vietnam.

2. Material and Methods

2.1. Sampling Sites in Danang Beaches

Field surveys and sampling were conducted in March 2021 in the shoreline sediments of seven sites along the Danang beaches. All sampling sites of the Danang beaches were expressed in Table 1, Fig. 1.

The sampling of MPs in the sediments was adapted from the method described by Eo (2018) [8]. Before sampling, the positions of the sampling sites were reported and the sampling sites were recorded (Fig. 2). At each sandy beach, a crossing - section of three 100 m-width stretches was marked parallelly and compared to the coastline. Among them, the upper shoreline, the middle line and the water edge line were located from the farthest to the closest point to the sea. At each line, four sections of 25m intervals were marked and four subsamples were collected from four dimension separated 0.5 m × 0.5 m randomly along each section (Fig. 2, A). The samples were triplicated with two depths separately, the surface from 0 to 5 cm (layer 1) and the deeper layer (layer 2) from 5 to 10 cm (Fig. 2, B). Each sample was collected in range of the square wooden frame, sediment sample was mixed and collected by stainless steel shovel. When the sampling was done, each sediment sample was primarily sieved through a 5 mm mesh sieve to remove all matters larger than 5 mm to keep compositions as MPs. Then, the sample was poured into a properly labeled glass bottle (Duran® bottle, 1000 mL), transported to the laboratory, and stored in the fridge (4 ºC) for analysis.

Table 1. Sampling sites on Danang beaches

No	Site code	Zone	Longitude	Latitude	Name of beach
1	NNB	Zone 1	108.2741	15.9955	Non Nuoc
2	STB	Zone 1	108.2638	16.0163	Son Thuy
3	T20	Zone 1	108.2517	16.0438	T20
4	HSB	Zone 1	108.2526	16.0948	Hoang Sa
5	PLB	Zone 2	108.1754	16.0773	Phu Loc
6	XTB	Zone 2	108.1448	16.0971	Xuan Thieu
7	NOB	Zone 2	108.1324	16.1116	Nam O

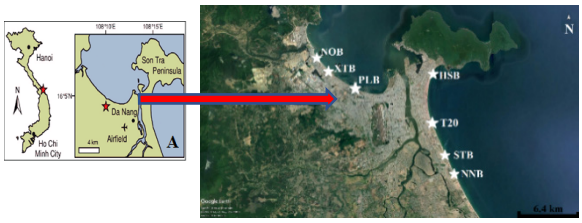

Fig. 1. Danang shorelines and 07 sampling sites

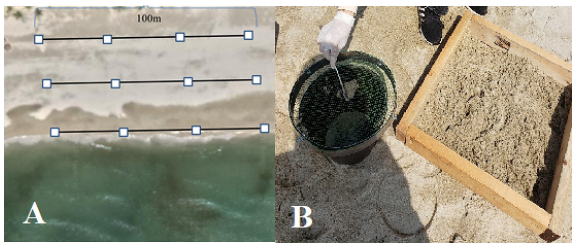

Fig. 2. Sampling area (A) Sampling points along the lines, (B) collecting samples

2.2. Microplastics Analysis

In the laboratory, sediment samples were dried at 55 °C - 60 °C for 72 h in a drying oven to avoid the formation of agglomerates. Then, the extraction was performed with minor modifications as described by [11]. Briefly, 10 g of dried samples were digested by adding 30 mL H_2O_2 30% and 10 ml Fe (II) solution 10g/L and kept at 40 °C for 8 hours. Consequently, the density separation step was conducted with saturated $ZnCl_2$ 5.64 M (d = 1.6 g/ml) by overflow-producing technique. For each sample, the overflow process was performed three times to obtain the highest recovery of MPs. Then, the solution contained MPs was filtered

through glass Whatman GF/F diameter 47 mm, spore size 0.7 µm by the stainless steel 6 - branches vacuum filtration assembly model 168 M6 SS500 Sartorius - German. Afterward, the sample filtered papers were kept in covered glass petri dishes for identifying the MPs. Finally, the Micro Fourier - transform infrared spectroscopy (Micro - FTIR) (Nicolet iN10MX) was used to determine the density, size, shape, and polymer types of the MPs in samples collected from beach sediments. Spectra IFTR for all particles in sample were recorded from 4000 to 650 cm^{-1} in 3s for one measurement. The measured data were signals and images that automatically analyzed by the Particle Wizard and compared with the Ommic polymer spectra library. Component name and match (%) were determined exactly by the spectral library databases. By screening technique of the match greater than 70% and considering to cancel some polymer components as natural polymer, data of MPs were obtained with named polymers. The shape of MPs was determined by their length (L) and width (W) with fragment ($\underline{L:W} \geq 2$), fiber ($L:W < 2$) and bead ($L=W$). The size of MPs was devided with some dimension depended on the measured MPs data.

2.3. Quality Assurance - Quality Control

Ethanol solution 70% was used to clean arouding working area before manupulation of the analysis procedure and to avoid MPs contamination. All experimental instruments and containers were made of glass, stainless steel and metal materials. The laboratory coat and clothes were cotton and nitrile gloves were used. Distilled water and chemical solutions were filtered by filter paper before using. All beakers and glass containers were cleaned at least 3 times by pure water before using. The blank sample was conducted to access MPs in the air of experimental area by using paper of the glass Whatman GF/F diameter 47 mm, spore size 0.7 µm openned in a petri disk. This blank sample was triplicated with 3 glass GF/F papers. This process was carried out in paralled with the analysis of MPs in sediment samples. Then, 3 filtered papers with possible airborne MPs were also measured as other sediment samples by using the Nicolet iN10MX. Finally, there was only 1 particle of MPs in 3 blank samples, thus maybe MPs existed availably in the air but it was very rare. Therefore, it was confirmed that airborne microplastic contamination in the laboratory was neglected and environmental condition was clean for MPs analysis in the sediment samples.

3. Results and Discussion

3.1. Microplastic Concentrations in the Shoreline Sediments

MPs were found at every site sampled, with an average concentration of 5565 ± 2359 items kg^{-1} dry weight (d.w.) at Danang beaches, but varied among sites and depths (Fig. 3). The average abundance ranged from 2094 items kg^{-1} (d.w) at the T20 beach, to 9116 items.kg^{-1} (d.w) at the XTB. Regarding different sandy beaches and depth distribution, the number of MPs measured in layer 1: 0 - 5 cm ranged from 2389 items kg^{-1}(d.w) at T20 to 11027 items kg^{-1}(d.w) at XTB while the concentration in the lower layer 2 (5 - 10 cm) varied from 1460 items kg^{-1}(d.w) at STB to 7206 items kg^{-1} (d.w) also at XTB. In comparison, the concentration of MPs at the 0-5 cm depth (7679±3363 items kg^{-1}(d.w)) was significantly higher than that at the 5-10 cm (3575±2539 items kg^{-1}(d.w)) (t-test, $p = 0.016 < 0.05$). This trend was observed in all sites of Danang beaches, expressing that MPs in the surface sediment were more concentrated than that in the deeper sediment layer. Overall, at the layer 1 (0-5 cm), average MPs concentration in seven beaches in Danang here was less that in the previous research with monitored three beaches and eight beaches depened on sampling sites [9, 10].

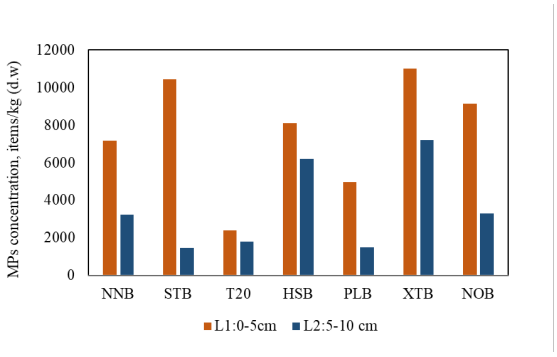

Fig. 3. MPs concentration in the layer 1: 0-5 cm and layer 2: 5-10 cm in Danang beach.

Our results showed that the MPs pollution was ubiquitous in Danang beaches. All 7 sampling sites in this study were found to have levels of MPs contamination. Since the majority of studies were conducted in the upper layer of beach sediment (top 5 cm only), the number of MPs recorded in layer 1 (0-5 cm) was used to compare with the concentration of others. With the MPs average abundance of 7679±3363 items.kg^{-1}(d.w) at layer 1, Danang beaches showed the contamination of MPs in the shoreline sediments more seriously than that in many other beaches in over the world. For example, the MPs concentration in Danang beaches was higher than the concentration found in the Mar Menor lagoon, SE Spain (53.1±7.6 items.kg^{-1}(d.w)), Dubai coast, UAE (59.71 items.kg^{-1}(d.w)); nearly 40 times higher than in Hengchun Peninsula, Taiwan (200 pieces kg^{-1} (d.w)); nearly 30 times denser than that in Chabahar Bay, Iran (262±17 items.kg^{-1}); more than doubled the abundance recorded in Qinzhou Bay, China (3266.0±6390.8 item.kg^{-1}). In comparison with MPs abundance found on sandy beaches in Vietnam, our results were much higher than that on beaches in Vung Tau city (44.6 items.kg^{-1}) [12] but smaller than the

study of Tran (2020) [13] also in Da Nang beaches (9238±2097 items.kg^{-1}(d.w.)). The variation in MPs abundance could be due to various reasons, including both natural-drive and anthropogenic factors. Lo (2018) highlighted the effect of particle size and wave energy, as they allowed MPs to drift further distances and thus, depending on the wave energy, it affected the rate of depositions [14]. Natural events such as storms or heavy rain could make an influence on MPs variation as water effluent and road runoff is another major pathway for MPs into beach sediment or the sea. Since a majority of our sampling sites were beaches for tourism purposes or easy to be accessed by people living nearby, MPs were likely to be transported from nearby anthropogenic sources, ranged from maritime recreational activities along the coastline to distant sources. The sewage effluents from hotels, restaurants, and laundry facilities could increase the amount of MPs in tourist areas [4]. Chen (2020) reported in their study that tourism activity could become the primary factor affecting MPs abundance in major tourist attractions. Hence, sediment MPs in Danang was possiblly sourced from tourism development over longtime.

Regarding depth distribution, our results showed that MPs were more concentrated in the upper layer than in the lower depth, which was agreeable with the study of Yu (2016) in the sediment of Bohai beach, China [15]. The abundance of MPs decreased significantly with the depth of the sediments, as reported by Turra (2014) in the Amazon at 61.5%, 25.0%, and 13.5% from the surface to 20, 20 - 40, and 40- 60 cm, and in Brazilian sandy beaches [16]. The reason for that phenomenon could be due to various factors, including the surface layer exchanges directly with the seawater or wastewater which might retain more MPs, or the larger plastics in the top layer that could be divided into smaller pieces by people's activities at the surface [15].

3.2. Distribution of the Sizes and Shapes of MPs in the Shoreline Sediments

MPs in Danang sandy beaches had a wide range of sizes with an average size of 93.4 ±18.8 µm (min - max: 22.4-2797.2 µm). In both layers, there was a dominance of MPs in the size range of 50 to 150 µm, accounting for 91% of the total MPs recorded. Among the range of 20 to 150 µm, MPs of size from 50 to 150 µm were more frequently observed than other size fractions, with 46% and 48% of the total MPs recorded in layers 0-5 cm and 5-10 cm, respectively. There was a similar trend in the size composition of MPs in all beaches sampled as most of the MPs measured within the range of 20-150 µm. In the upper layer (0-5 cm), MPs within the size class of 50 to 150 µm varied from 24% at PLB to 92% at T20 and it had a quite similar trend with MPs of size 20 to 50 µm (min: 25% at HSB and max: 62%, at PLB) except for site NNB and T20 which had MPs bigger than 50 µm only (Fig. 4, A). For the deeper layer (5-10 cm), it reached up to 71% at NOB (size class 50-150 µm) and 58% at XTB (size class 20-50 µm) (Fig. 4, B). The average size of MPs in the layer 0-5 cm was 92.1±21.7 µm and less than 95.9±24.2 µm in the layer 5-10 cm. However, there was no significant difference between them.

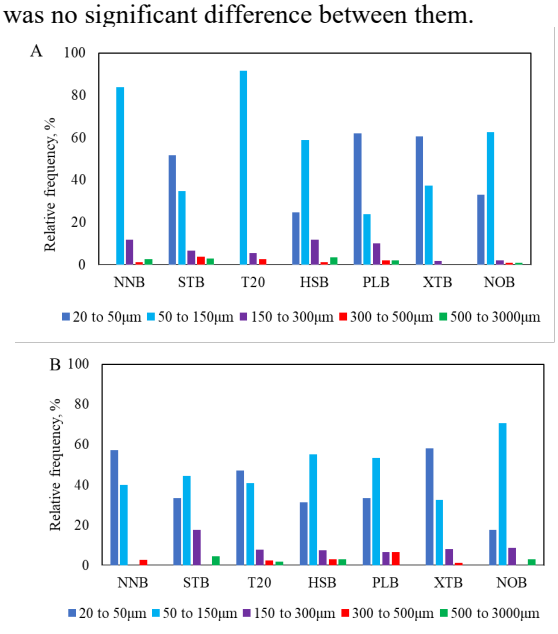

Fig. 4. Size distribution of MPs in A) layer 1: 0-5 cm, B) layer 2: 5-10 cm

With regards to the size of the MPs analyzed, the dominant size in our study ranged from 20 to 150 µm (91% and 88% in upper and lower layers, respectively) with the most abundance group belonging to 50 to 150 µm (46% in (0-5 cm) and 48% in (5-10 cm) layers). Godoy (2020) also reported similar observations for the majority of MPs found in beach sediments (more than 50% of the particles found were smaller than 100 µm) [17]. Our results also agreed with the MPs size distribution reported on the North coast of the Persian Gulf (70 -97% of MPs were between 10 - 300 µm) [18].

Considering the variation in the average size of MPs found in the layer 1 (0-5 cm) and layer 2 (5-10 cm), the reason could due to the fragmentation of larger plastics in the top layer to smaller particles by anthropogenic factors [15]. Additionally, the variation of MPs size distribution in different studied areas could be explained by numerous reasons including different sources, rates of degradation, environmental factors such as temperature and solar radiation, variation of sampling methods, and resultant detection limits. Combing all, this might also explain the reason why the length of microfibers in our study in the top 5 cm depth was shorter than that in the deeper layer (5 -10 cm).

The morphology of MPs found in Danang beach, fragment was the most dominant shape (76% of the total MPs found), fiber was second in number with

18% and bead was the scarcest shape with only 6%, other morphology was not detected. Regarding the layer 0-5 cm, MPs fragment ranged from about 69% to nearly 90% (Fig. 5, A) and fiber ranged from approximately 10 to 30%. While fragments and fiber were observed in the upper layer of all sampling sites, the bead was recorded in 5/7 sampling sites with a maximum percentage of 10%. The same trend has also happened in the lower layer (5-10 cm) (Fig. 5, B). Except for NOB where fragments accounted for 62%, other sites had fragment occurrence of more than 70% with a maximum of 91% in NNB. The occurrence of fiber varied from 9 to 38% and bead appeared in 4/7 sampling sites with an average percentage ranging from about 5 to 10 %.

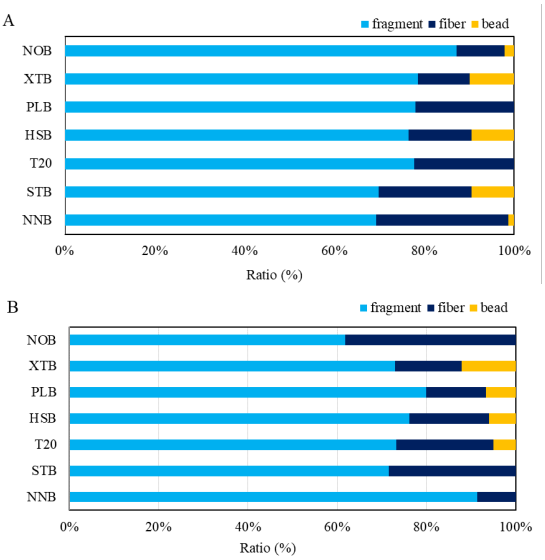

Fig. 5. Shape distribution in (A) layer 1: 0-5 cm, (B) layer 2: 5-10 cm

In this study, at all sampling sites, the fragment was the predominant shape of MPs found in beach sediment, with higher occurrence than fiber or bead. The dominance of fragment MPs was in line with other studies from both European and worldwide [15]. The proportions of different shapes of microplastics were affected by the land-based activities near the beaches. Alvarez-Zeferino (2020) reported that wastewater discharge and estuaries could increase the fibrous MP's proportion [19], while beaches with high touristic activities at the Danang beach could have higher concentrations of rigid and foamed fragments, which originate from the degradation of disposable cutlery and plastic bags.

Due to the variation of pollutant sources and environmental processes (e.g. erosion, solar radiation, biodegradation), microplastics detected in coastal environments may be in regular and irregular shapes. While regular and common shapes normally are derived from a direct release (primary MPs), the irregular shapes mainly come from the fragmentation of large plastic debris (secondary MPs), which include fragments and fiber [4]. Those uncommon shapes of plastic particles are primarily formed through the weathering process of household plastic materials to urban runoff by the sea. Recently studies suggested that the distribution of fragments and fibers was higher than compared to other forms in the beach sediments [13] and their concentration was higher in urban beaches than in natural or semi-natural beaches.

3.3. Composition of the Polymers in Microplastics

There was variation in the polymer type's composition between two layers. The occurrence of 52% of the total MPs found in the layer 1: 0-5 cm, Polyamide (PA), or Nylon, was the dominant polymer type with a minimum level of 27% at HSB and NOB, and maximum level of 68% at PLB (Fig. 6, A). PTFE (Polytetrafluoroethylene) was the second most common type and it could reach up to 67% at NOB. Aside from PA and PTFE, PES (Polyester), EVOH (Ethylene-vinyl alcohol), and MUF (Melamine-Urea-Formaldehyde Resin) occurred less frequently. Among them, PES was the scarcest type as it only appeared in 4 sampling sites, and occupied up to 1% in all sites, except for site PLB (6%). EVOH and MUF each could come up with a maximum occurrence of 13% in all sampling sites. OTHERS including 16 polymer types (Polydimethylsiloxane (PDMS), Cellophane, Polyvinylidene fluoride (PVDF), chlorinated polypropylene (CPP), Urea-formaldehyde (UFs), Polyurethane (PU), Silicone rubber, Acrylic, Melamine formaldehyde resin (MF), Epoxy resin, Phenol formaldehyde resin (PF), Poly (vinyl alcohol) (PVA), Polyetherimide (PEI), Polyethylene Low Density (LDPE), Poly (oxymethylene) (POM), and Ionomer resin, accounted up to 18% in all sampling sites in the layer 1.

There were differences in the composition of polymer types found in the layer 2: 5-10 cm (Fig. 6 B). In particular, PES appeared less in the deeper layer (3/7 sites) but with higher concentration (ranging from 4% at T20 to 13% at STB). PA, PTFE, EVOH, and OTHERS appeared in all sampling sites at a depth of 5-10 cm and with varied numbers. Similar to layer 1, PA was the dominant polymer type found in layer 5-10 cm with 32% of the total MPs recorded (min-max: 3% (HSB)-74% (NNB). However, PTFE was the second most common with a similar occurrence 28% of the total MPs, min-max: 11% (NNB)-54% (T20).

Besides the beach difference, the sampling sites into 2 areas were divided to the southern area (zone 1) and the northern area (zone 2) to compare the difference in polymer distribution in those two separate areas. The most noticeable statement was showed that there were more OTHERS polymers in zone 1 than in zone 2 in both layers. In detail, for the layer 1 (0-5 cm), OTHERS occupied only 3% in zone 2, while it rose to 13% in zone 1. As for the layer 2

(5-10 cm), the Fig. 6 were 22% (zone 1) greater than 16% (zone 2). Aside from OTHERS, PA and PTFE seemed to have a reverse characteristic in zone 2 of the layer 2, as PA became more and PTFE became less frequent from PLB to NOB in zone 2 (from South to North).

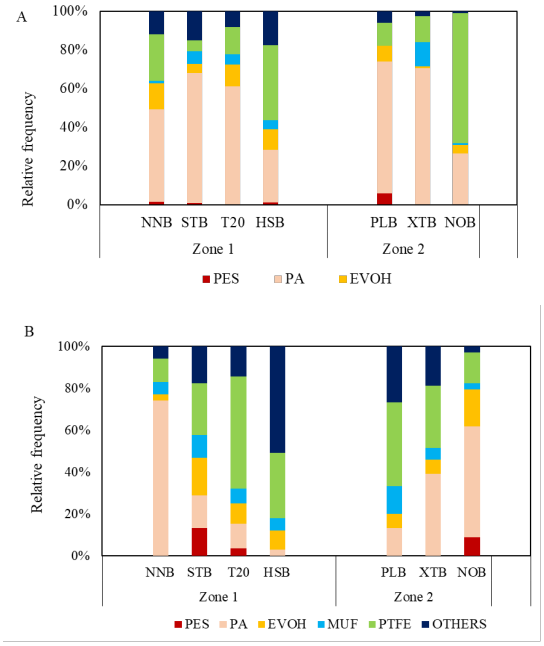

Fig. 6. Polymer distribution in Da Nang beach sediment in, A) layer 1: 0-5 cm, B) layer 2: 5-10 cm

In our study, PA or Nylon and PTFE were considered the most dominant polymer types in both layers. PA was also observed to be in the common groups of polymer reported in Niterói, RJ, Brazil (for fragments, pellets, and fibers) [20], one of the four polymers groups found in Kiel Fjord, Western Baltic Sea. Since nylon was used to make fishing nets and threats, it was considered the most important source of marine plastic contamination. Nylon was not only used in the majority of fibrous materials such as clothes, carpets, ropes, and airbags but also fishing nets. In addition, nylon was also utilized in balloons, frozen foods, liquids, and medical supplies packaging, as well as vacuum packaging material.

While PA belongs to the groups of quite common polymer types found in beach sediment on several beaches worldwide, PTFE was the opposite. It was stated that although PTFE was resistant to the environment and not an important component of MPs pollution because of its highly stable chemical properties, it has become the most widely consumed fluororesin. Consequently, applications of PTFE in food processing can lead to reduce fouling, food contamination, and biofilm formation [5]. Thus, the origin of PTFE found in Danang beach could come from the fragmentation of packaging with the economic massive development related to fishery and tourism.

4. Conclusion

In this study, the occurrence of MPs in the sediment of different beaches in Danang city was evaluated. The results showed that the number of MPs items in the surface layer (7679±3363 items. kg^{-1}(d.w.)) was higher than double in the deeper layer (3375±2539 items.kg^{-1}(d.w.)). The MPs size was found the most dominant in the range from 20 to 150 µm in both layers with a contribution of more than 91%. Among the morphology of MPs, the fragments were the dominant shape observed with an average of 76% while bead was found with an average of 6% in all samples. Finally, the MPs identification was performed with the µ-FTIR technique, the polyamide (PA) and Polytetrafluoroethylene (PTFE) were the most types of polymers detected. The PES was found in only 4/7 sampling sites. These charactersitics of MPs in this reseach will enhance further studies to perform the investigation of the behavior as well as the impact and risks of MPs to the environment and human health in future.

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