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# Microplastic abundance and characteristics in seawater and inter-tidal sediments of the Tokin Bay Coast (North Vietnam)

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# Abstract

Microplastics (MPs, plastic items from 1  $\mu\text{m}$  to 5 mm in size) are present in all the environmental compartments. The evaluation of their concentration, fate, and spatial distribution is still a challenge for the scientific community. This concern is just debuting in developing countries, *i.e.* Asia, South America and Africa. This study deals with the MP contamination in the abiotic marine compartments of the Northern of Vietnam: seawater and intertidal sediments. Four sites located in the intertidal zone or near the coastline in the Tonkin bay, Vietnam were studied. A total of 16 samples (8 for each compartment) were collected in July 2020 (rainy season) and January 2021 (dry season). Plastic-like items were found at levels ranging from 3 to 303 items/ $\text{m}^3$  in seawater and from 63 to 955 items/kg dry weight in sediments. Most of these were fibers less than 300  $\mu\text{m}$  long. Higher levels in seawater were found in the rainy season compared to the drier one. As the river flow was estimated six times higher during the rainy season than during the dry season, these results suggest the river discharge as a potential source of contamination for coastal zone. The temporal variability was lower for the sediments than for the seawater, demonstrating the long-term integration of the plastic-like items in this compartment. While the nature of fibers wasn't investigated, the plastic-like fragments were analyzed on  $\mu\text{FTIR}$  imaging spectrometer. The determination of polymer composition indicated that most of the fragments were made of polypropylene (PP, 82%), followed by polyethylene (PE, 9%) and polystyrene (PS, 9%). The fragment size was similar in two studied compartments but it was depending on polymer types, since PS fragments ( $140 \pm 17 \mu\text{m}$ ) were smaller than those made of PE ( $622 \pm 123 \mu\text{m}$ ) and PP ( $869 \pm 905 \mu\text{m}$ ).

## 1. Introduction

The plastic pollution has been recognized as a high challenge for our society (GESAMP, 2019). It was found in all environmental compartments and even in human consumption daily products. The microplastics (MPs), with sizes from 1  $\mu\text{m}$  to 5 mm, were reported in remote areas, such as the Antarctica (Reed et al., 2018), and in the six continents (Phuong et al., 2021). It was also found in table salt (Iniguez et al., 2017; Kosuth et al., 2018), beer, honey (Diaz-Basantos et al., 2020), milk (Kutralam-Muniasamy et al., 2020) and drinking water (Kankanige and Babel, 2020; Obmann et al., 2018; Zuccarello et al., 2019) which poses a serious threat to human health. In the environment, MPs can be directly ingested and accumulated by living organisms due to their micrometric size (Cho et al., 2019; Setala et al., 2016), and transferred along of the trophic chain (Farrell and Nelson, 2013). The omnipresence of MPs hence influences environmental quality and is at risk to the ecosystems.

As the endpoint of the plastic waste cycle, marine environment is known to be affected by MP pollution (Asensio-Montesinos et al., 2022; Pavithran, 2021; Rios Mendoza et al., 2021). Many hazardous effects of their presence in marine ecosystems were reported on organisms (Palmer and Herat, 2021; Pirsahab et al., 2020).

The Association of Southeast Asian Nations (ASEAN) includes ten countries. Five out of them are estimated as top 20 countries mismanaging plastic wastes (Jambeck et al., 2015). The Vietnam, located at the East of Southeast Asia, is among these countries. It covers an area reaching more than 330,000  $\text{km}^2$ , owning a coastline long of 3,260 km. Maritime economics were defined as one of the most important, including many activities, such as aquaculture, tourism and fisheries. Plastic pollution has been recognized as a major problem for the quality of Vietnamese marine environments (Omeyer et al., 2022). Owing more than 98 million inhabitants in 2021 ([www.gso.gov.vn](http://www.gso.gov.vn)), Vietnam generated nearly 2 million tons of plastic wastes each year. It was classed as the fourth country responsible for plastic emissions to the oceans in a previous study (Jambeck et al., 2015) and the eighth in another one, more recent (Meijer et al., 2021). The country participates to the high levels of MPs in local marine environment (East Sea, Walker, 2021). However, studies about MP pollution have just been initiated in this region. It may be related to the lack of

adapted instruments in local laboratories, such as microscopy coupled to Fourier-transform infrared spectroscopy ( $\mu$ FTIR) or Raman spectroscopy allowing MP identification and quantification (Strady et al., 2021).

Among the published studies on plastic pollution in Southeast Asian region, three different methodologies were performed for the MP quantification. The first one relied on visual observation without polymer identification. With this method, plastic-like items were studied by eye or microscope observations. Hence, it focused usually on large items *i.e.* with a size bigger than 1 mm (Curren and Leong, 2019). The risk is the over-estimation of MP quantity, since up to 70% of the suspected plastic-like items couldn't be effectively characterized as plastic particles (Hidalgo-Ruz et al., 2012). Inversely, this method has some advantages as simple, fast and easy. Besides, it was known reliable on fiber MPs (Volgare et al., 2022). The second method involved the observation of sieves or filters under a microscope, then a manual sorting of the items suspected to be MPs were performed followed by a polymer identification by FTIR or Raman spectroscopy. Actually, the major limit of this method is the difficult manual sorting of the smallest items, balanced by the use of analytical instruments more spread in developing countries. From studies performed in Southeast Asian regions, only one performed a direct MP identification on the filter, which corresponds to the third type of MP quantification methods, using microscopy coupled to FTIR (Nam et al., 2019), while automated methods, *i.e.* FTIR-imaging or Laser Direct Infra-Red (LDIR) have not yet been employed (Ourgaud et al., 2022; Treilles et al., 2021).

The understanding of MP pollution in the marine environment of the North Vietnam coast is still lacking. Due to the non-controlled sources, high levels could be expected. The main novelty of this work is to propose the first insight of the MP distribution in abiotic compartments from different marine sites with various anthropogenic pressures and the assessment of the seasonal variability in one of the most important pollution hotspots worldwide. Hence, the different objectives of the study were: (i) to investigate the spatial variation of MP pollution in 4 locations in Tonkin Gulf, on the North part of the country, exhibiting different anthropogenic activities; (ii) to assess the influence of the season on the MP distribution and finally and (iii) to determine possible sources depending on the nature of plastic-like items found in the abiotic compartments studied.

## 2. Materials and Methods

### 2.1. Studied Locations

Samples were collected at 4 locations located in Tonkin Bay, Northern Vietnam (Fig. 1 and Table 1). They correspond to 4 sites approximately 50 km apart one by one, from the North to the South of the coastline: Van Don, Quang Ninh (QN), Cat Hai, Hai Phong (HP), Thai Thuy, Thai Binh (TB) and Giao Thuy, Nam Dinh (ND).

The sampling locations correspond to four important socio-economic centers which are located in the North of Vietnam. Sampling was performed in July 2020 and January 2021 corresponding to the rainy and dry season, respectively. Meteorological parameters such as air temperature, wind speed and humidity, as well as salinity of seawater were also reported (Table 1).

Table 1  
Sampling locations and environmental parameters measured during the sampling in the field

Location	Latitude	Longitude	Campaign	Temperature (°C)	Wind (km/h)	Humidity (%)	pH	Salinity (‰)
Quang Ninh (QN)	21°03'04.9"N	107°26'35.7"E	Jul-2020	37	8	69	7.76	30
			Jan-2021	18	6	82	8.07	31
Hai Phong (HP)	20°46'13.4"N	107°04'38.7"E	Jul-2020	36	12	61	7.76	30
			Jan-2021	15	7	46	8.01	30
Thai Binh (TB)	20°34'56.7"N	106°39'57.7"E	Jul-2020	37	8	70	7.40	25
			Jan-2021	14	17	26	7.20	30
Nam Dinh (ND)	20°14'03.4"N	106°31'38.7"E	Jul-2020	36	10	60	6.96	25
			Jan-2021	12	16	39	6.34	28

Quang Ninh province has a natural area of 6,178 km<sup>2</sup>. It has 1,337,600 inhabitants in 2020 ([www.gso.gov.vn](http://www.gso.gov.vn)). The province shares both land and sea borders with a coastline that stretches for 250 km. It owns the Ha Long Bay, a natural wonder of the world that has been recognized by UNESCO as a World Natural Heritage site. Tourism is an important contributor to Quang Ninh's economy, with the province attracting up to 8.8 million tourists in 2020 despite the COVID-19 pandemic ([www.vietnamtourism.gov.vn](http://www.vietnamtourism.gov.vn)). The sampling site is located in the coastal area of Van Don district, near Cai Rong port. It's an island district located in the northeastern outpost of Vietnam in Bai Tu Long bay. It covers 551 km<sup>2</sup> with 60% of forest land and 47,500 inhabitants in 2017.

Hai Phong is a coastal city and the second largest city in the Northern (after Hanoi capital). It has a natural area of 1,562 km<sup>2</sup> and a population of 2,053,500 inhabitants in 2020. Hai Phong port, which was established in the 1870s, is the largest seaport in Northern Vietnam. The volume of goods was estimated up to 142 million tons in 2020 (Nguyen, 2022). Samples were collected in Lan Ha Bay, located east of Cat Ba Island in Hai Phong, near Cat Ba National Park.

Thai Binh (1,586 km<sup>2</sup> of area and 1,870,300 inhabitants in 2020) is an important aquaculture center in the Northern. In 2022, it cultivated up to 182,000 tons of seafoods. Sampling site is located in an estuarine area of Thai Thuy district. It is near the Thai Binh estuary where the average water flow rate was around of 370 and 90 m<sup>3</sup>/s in January and in July 2016, respectively (Duong et al., 2022a).

Nam Dinh (1,668 km<sup>2</sup> of area and 1,780,300 inhabitants in 2020) was called the textile city of Vietnam. Today, it gathers more than 400 companies and 200,000 active workers in this domain. Aquaculture is also important with nearly 130,000 tons of seafoods produced in 2022. In this location, samples were collected in the estuarine zone of Giao Thuy district. This zone is strongly influenced by Ba Lat estuary (the largest estuary of Red River). The average annual water flow reached about 540 m<sup>3</sup>/s, with values from 247 to 841 m<sup>3</sup>/s for dry and rainy season, respectively (Le et al., 2023).

## 2.2. Sampling

Seawater was sampled using a Manta net with a mesh size of 80 µm and a diameter of 50 cm. The net was deployed during 120 seconds from a boat with a speed of 5 km/h. The volume of seawater and the surface covered by the deployment of the net were measured using a flow meter (2030R Flowmeter, General Oceanics). Items remained in the cod-end of the net were rinsed three times with filtered seawater (0.7 µm of porosity) in the field. Large items

such as wood and other vegetal debris were removed using a metallic clip. The remaining was transferred into a clean glass bottle. At each location, seawater was collected at three different sampling points with a distance higher than 200 m from each other. The items collected from the three sampling points (around 10 m<sup>3</sup> of filtered seawater) were then pooled to constitute a composite sample of the corresponding location with a final volume adjusted to 1 L.

Surface sediment samples, 0 to -5 cm of depth, were collected during low-tide periods using a spatula and doing squares of 25 x 25 cm in size. The samples were then preserved in aluminum folds. Five sampling points per site, with a distanced higher than 100 m from each other, were collected to achieve the representativity of the studied sites, as previously recommended (Frias et al., 2018). Sediments from the 5 sampling points were then pooled into an aluminum pot, handle mixed to ensure homogeneity and a volume of 1 L of wet sediment was transferred into a clean glass bottle, carefully closed.

Samples (seawater and sediments) were then placed into an isotherm box and kept cooled with dry-ice during the transport to the laboratory. At the laboratory, they were maintained at cold temperature (-20°C) until further analysis.

## **2.3. Sediment characterization**

Water content and granulometry of sediment samples were evaluated. For each parameter, 5 replicates of 20 g of wet sediment were used. Sediments were dried in an oven at 60°C until the mass unchanged in order to determine the humidity. In parallel, dried sediment was sieved through three mesh size (< 0.2 mm, 0.2 to 1 mm and > 1 mm) for grain size analysis. Results of both tests were expressed in percentage.

## **2.4. MP analysis**

### **2.4.1. Sample Pretreatment**

After taking out of freezer, the glass bottles containing either items in seawater or sediments were placed into a hood at room temperature (25 ± 1°C) until thawing. Seawater samples were homogenized by handing agitation for 2 minutes. Then, 20 mL were transferred into a 250 mL beaker. This step was performed 5 times to constitute 5 replicates of each location, with an agitation repeated between each transfer. Sediment samples were transferred to a glass crystallizer and handing mixed with metal spatula for 5 minutes. Approximately 20 g of wet sediment were transferred into a 500 mL glass beaker. The constitution of 5 replicates per sampling site was made using a quartering procedure. In detail, the sediment into the crystallizer was divided in 4 equivalent zones. One part of each zone was taken to constitute the 5 replicates.

### **2.4.2. Plastic-like Items Extraction Procedure**

Before MP analysis, the organic matter had to be removed from the samples. Thus, organic matter was digested by hydrogen peroxide solution (H<sub>2</sub>O<sub>2</sub> 30%, Xilong Scientific). Volumes of 20 mL and 100 mL of H<sub>2</sub>O<sub>2</sub> were added to the seawater and sediment samples, respectively. Both types of samples were agitated with a magnetic stirring (200 rpm) and heated at a controlled temperature of 40°C to avoid plastic-like item degradation. The digestion step was maintained until the end of the formation of foam in the beaker, corresponding to 24 to 72h depending on the organic matter quantity in the samples. Plastic-like items were then extracted from both seawater and sediment using a densimetric separation, with the addition of NaCl (Shanghai Zhanyun Chemical) saturated solution with density around of 1.20 g.cm<sup>-3</sup> (4:1; v/v, NaCl/sample). The separation time was 24h and repeated three times. Then, the surface layer of the hypersaline solution was filtered on glass filter (0.7 µm × 47 mm; Whatman). Finally, the filters were placed in closed glass petri dishes, at room temperature, and dried until further analysis.

### **2.4.3. Quality Assurance and Control**

Field and laboratory recommendations for MP analysis were strongly respected (GESAMP, 2019; Frias et al., 2018). The clothes and the gloves used for both for sampling and laboratory work were made of cotton and nitrile respectively. Materials were cleaned before the use, by rinsing them three times with double-distilled water. They were then preserved from MP contamination in aluminum foil. Before the use, the NaCl solution was prepared with double-distilled water and H<sub>2</sub>O<sub>2</sub> solution was filtered using a glass filter of 0.7- $\mu$ m porosity.

Cross contaminations were therefore investigated by the analysis of three different types of blanks. For each type, five replicates were performed. Firstly, clean filters were placed in a glass petri dish opened during the sampling, corresponding to field blanks. Secondly, the extraction protocol was performed without seawater and sediment samples. In this case, the same mass of double-distilled water than seawater or sediment was used instead of them, constituting the protocol blanks. Finally, clean filters in opened glass petri dish were placed near the microscope during the observations of the filters obtained from samples, representing observation blanks. No plastic-like item was found in the observation blanks. However, 0.38 ( $\pm$  0.49) plastic-like item/sample was found in field blanks (n = 40), as well as 0.60 ( $\pm$  0.49) plastic-like item /sample for protocol blanks (n = 5 for each sample type). Based on these values, cross contamination was rated as negligible for the sampling and analysis steps in the present study.

Procedure recovery was determined by spiking MPs in samples. Five MP types, including PE (polyethylene), PP (polypropylene), PS (polystyrene), PVC (polyvinyl chloride) and PET (polyethylene terephthalate) were prepared with a size from 100 to 500  $\mu$ m and different colors. Before spiking, they were photographed (Sony A7 Mark III). Ten items of each polymer were added into the samples of seawater and sediments in 5 replicates. After sample preparation and MP analysis, the recovery values (MPs found in the sample/MPs added in the sample) were about 83% for both matrices (details in Table S1, Supplementary Material). The highest recovery was determined for PE with 90.0  $\pm$  6.3% (n = 5) in seawater and 90.0  $\pm$  12.6% (n = 5) in sediment. The MPs made of the densest polymers, *i.e.* PET and PVC, showed the smallest recovery reaching approximately 80.0% in both matrices. These values were similar to others determined in a previous recent study (Duong et al., 2022b).

## 2.5. Plastic-like Item observation

Visual observation was performed in a clean room, input air being filtered at 0.3  $\mu$ m. Dried filters were observed under a microscope (Olympus SZX2-TR300) for the visualization of particles potentially made of plastic (Fig.S1, Supplementary Material). The size limit for observation using this instrument was evaluated at 10  $\mu$ m but only items larger than 50  $\mu$ m were counted to be up to the limit size needed for the manual sorting operation. Items were considered as plastic-like items when corresponding to the five criteria proposed by Nor and Obbard (2014), which are:

1. No cellular or organic structures are visible
2. Fibers are equally thick throughout their entire length and should not be tapered at the end
3. Particles are homogeneously colored
4. Fibers are not segmented, or appear as twisted flat ribbons
5. Particles are not shiny.

The observation of each filter was performed by two experimented and independent operators. On each filter, when an item was suspected to be a plastic-like item, its coordinates was indexed. All of them were photographed to identify physic characteristics. Quantitative (concentration) and qualitative data (form, size, color) were recorded from the visual observation by microscopy.

## 2.6. Polymer Determination by $\mu$ FTIR Imaging



The polymer determination was performed only on the particles and not on the fibers. In fact, the fibers are known to have a very small secondary dimension, usually, smaller than the size resolution of the  $\mu$ FTIR (Gasperi et al., 2018). Consequently, some fragments were randomly selected and removed from the sample filters, using one small clip and transferred into a glass vial (2 mL) filled with pure ethanol until the polymer determination step which has been carried out in France (GERS-LEE Université Gustave Eiffel). Before the polymer analysis, the items in ethanol were sonicated during 1 minute (ZETA sonic) and rinsed 3 times with pure ethanol, then filtered through an anodisc filter per type of sample (seawater or sediment) (0.2  $\mu$ m of porosity and 25 mm of diameter). A total of 32 selected fragments (26 and 6 for seawater and sediment, respectively) out of 39 fragments observed (27 in seawater and 12 in sediment) were analyzed using  $\mu$ FTIR spectrometer (NICOLET iN 10 MX, Thermo scientific).

Analysis method was performed by mapping mode with a resolution of  $25 \times 25 \mu\text{m}$  (Dhivert et al., 2022; Treilles et al., 2021). A spectrum was acquired for each particle in transmission mode and with an imaging detector. The background was performed on a clean anodisc filter. All spectra were recorded from  $4,000$  to  $1,200 \text{ cm}^{-1}$  with 1 scan and a resolution of  $8 \text{ cm}^{-1}$ . Due to the high signal of anodisc filters at wavenumbers lower than  $1,200 \text{ cm}^{-1}$ , this part of the spectra was not considered. The noise signal, *i.e.* water vapor and  $\text{CO}_2$  in the air, was automatically removed. The spectra obtained from each particle were compared to a bank of reference spectra present in the siMPle library (MP\_Library\_Extended\_Grouped\_1\_5.txt) of the siMPle software (version 1.1.β), recently developed by Aalborg University and Alfred Wegener Institute. The polymer composition of the particles was validated when the matching score was higher than 55%, 60% and 75% for PP, polystyrene and PE, respectively.

## 2.7. Statistical Analysis

According to the sampling method and the analytical procedure, the quantitative results of plastic-like items found in seawater were expressed in 2 different units, *i.e.* plastic-like item/ $\text{km}^2$  and plastic-like item/ $\text{m}^3$ , corresponding to the covered sampled surface area and the volume of seawater filtered through the Manta net. For sediment samples, the levels of plastic-like items were expressed as plastic-like item /kg dry weight (dw). These units allowed a better comparison of the results with values from the literature. The levels in samples were corrected with the subtraction of the mean value of plastic-like items found in the protocol blanks. Statistical differences of levels between the studied locations and seasons were determined by non-parametric tests (Kruskal-Wallis) with Dunn as the post hoc test and a *p-value* set up at 0.05.

## 3. Results

### 3.1 Sediment characteristics

Table 2 presented the sediment characteristics. Water content analysis demonstrated that sediment samples from 4 locations were similar. The humidity ranged around 20 to 30% in 7 out of 8 samples. The grain sizes of sediments at QN and HP sites are bigger than those from the two other sites. These results traduced the different environment of the sites: coastal type for QN and HP, estuarine type for TB and ND.

Table 2

Characteristics, *i.e.* water content and granulometry, of the sediments from the 4 locations (average  $\pm$  standard deviation; n = 5 for each).

Location	Sampling campaign	Water content (%)	Granulometry (mm)		
			< 0.2 (%)	0.2 to 1 (%)	> 1 (%)
Quang Ninh (QN)	Jul-2020	26.69 $\pm$ 0.81	54.52 $\pm$ 4.99	28.40 $\pm$ 3.64	17.08 $\pm$ 2.52
	Jan-2021	30.09 $\pm$ 0.89	42.66 $\pm$ 3.26	41.04 $\pm$ 7.91	16.30 $\pm$ 2.23
Hai Phong (HP)	Jul-2020	19.91 $\pm$ 1.08	98.59 $\pm$ 0.19	1.41 $\pm$ 0.21	0
	Jan-2021	9.98 $\pm$ 0.38	62.53 $\pm$ 8.49	29.14 $\pm$ 8.51	8.33 $\pm$ 2.86
Thai Binh (TB)	Jul-2020	31.95 $\pm$ 1.02	98.53 $\pm$ 2.54	1.47 $\pm$ 2.53	0
	Jan-2021	20.10 $\pm$ 0.21	95.72 $\pm$ 0.44	4.28 $\pm$ 0.44	0
Nam Dinh (ND)	Jul-2020	27.09 $\pm$ 0.88	99.55 $\pm$ 0.03	0.45 $\pm$ 0.03	0
	Jan-2021	31.10 $\pm$ 0.42	98.92 $\pm$ 0.82	1.08 $\pm$ 0.82	0

## 3.2. Plastic-like Items in Seawater

### 3.2.1 Quantitative Results

A total of 304 plastic-like items was counted from all the seawater samples. Plastic-like item concentrations in seawater ranged from 3.0 to 303.2 items/m<sup>3</sup> (Fig. 2). Assuming a homogenized distribution from the sea surface to 50 cm on depth (diameter of the net), these values corresponded to concentrations ranging from 2.47.10<sup>6</sup> to 6.62.10<sup>6</sup> plastic-like items/km<sup>2</sup>. The average values ranged from 5.86  $\pm$  3.53 to 168.61  $\pm$  96.03 plastic-like items/m<sup>3</sup> (at HP and ND, respectively) in the rainy season (*i.e.* July 2020) and from 8.27  $\pm$  3.70 to 19.29  $\pm$  3.39 plastic-like items/m<sup>3</sup> (at TB and ND, respectively) in the dry season (*i.e.* January 2021). The concentrations at ND in the rainy season were significantly higher than those found at HP (both seasons) and at TB (dry season). However, no significant difference was found between the two seasons at ND site.

### 3.2.2 Plastic-like Item Characteristics

The microscopic observation allowed to determine the color, the size and the form of the plastic-like items found in seawater samples from the Northern of Vietnam. Seven different colors were reported with purple (41.62%), red (22.62) and white (22.12%) as predominant. The other colors included blue (8.09%), black (2.48%), green (2.34%) and brown (0.72%). Regarding the size (length dimension), the greatest percentages were found for sizes ranging from 100 to 300  $\mu$ m (49.29%), followed by the fraction 300 to 500  $\mu$ m (21.72%), 500 to 1,000  $\mu$ m (13.35%), less than 100  $\mu$ m (9.23%) and larger than 1,000  $\mu$ m (6.41%). Finally, only two different forms were observed, with a large predominance of fibers *vs* fragments, *i.e.* 277 (91%) *vs* 27 (9%).

## 3.3. Plastic-like Items in Marine Sediment

### 3.3.1 Quantitative Results

A total of 163 plastic-like items were found from all the sediment samples. Plastic-like item abundance ranged from 63 to 955 items/kg dw (Fig. 3). Average values ranged from 175.22  $\pm$  81.59 to 463.78  $\pm$  310.31 items/kg dw at TB and

QN, both during rainy season, respectively. No significant difference was observed between the studied locations and seasons ( $p > 0.05$ ; Kruskal-Wallis test).

### 3.3.2 Plastic-like Item Characteristics

Purple and red plastic-like items were the predominant colors (47.34 and 32.51% respectively). Other colors are blue (8.39%), white (6.98%), green (3.49%), brown (0.89%) and yellow (0.39%). Most of item (72.85%) presented sizes lower than 300  $\mu\text{m}$ , with 55.99% belonging to the fraction 100 to 300  $\mu\text{m}$  and 16.86% less than 100  $\mu\text{m}$ , while the two other fractions sizes (300 to 500  $\mu\text{m}$  and 500 to 1,000  $\mu\text{m}$ ) showed similar percentages (14.48% and 11.39%, respectively). Item larger than 1 mm were rarely observed (only 1.27%) in marine sediment samples. As for the seawater, two forms of items were observed. The fibers were still predominated compared to the fragments, 151 (93%) vs 12 (7%).

### 3.4. Polymer Composition of Fragments

Table 3 presented the identification of fragments using  $\mu\text{FTIR}$  imaging. Among all the fragments sorted from the sample filters, the  $\mu\text{FTIR}$  analysis demonstrated the presence of three different kinds of polymers (PE, PP and PS) from 32 fragments.

Table 3  
Chemical composition of fragments in seawater and marine sediment samples

Sample	Analyzed fragment	MP identified			Other materials
		PE	PP	PS	
Seawater	26	2	16	1	7
Sediment	6	0	2	1	3

None of dense plastics (*i.e.* PET or PVC) were identified. Among these fragments, 3 were identified as plastic in the sediments and 19 in the seawater samples. Other particles were characterized as natural material (protein) or not-identified. A total of 18 fragments out of the 32 were made of PP (2 and 16 from sediment and seawater, respectively). It counted for about 82% of the total identified MPs, followed by PE and PS (9% for each polymer type).

## 4. Discussion

### 4.1 Spatial and Temporal Influence on Plastic-like Item Abundance

Plastic-like item abundance in seawater samples from the Northern Vietnam coast seemed lower than those found on Indonesia coast (Khoironi et al., 2020) but higher than in seawater from Malaysia coast (Khalid et al., 2018) and Indonesian seas (Yuan et al., 2023) (Table 4). The differences probably strongly depend on the sampling method employed. Khoironi et al. (2020) directly sampled the seawater, using a cup, while the other studies used different mesh sizes, *i.e.* 20, 80 and 330  $\mu\text{m}$  mesh size in the study of Khalik et al. (2018), Strady et al. (2021) and Yuan et al. (2023), respectively.

The highest concentration of plastic-like items was found in the seawater collected during the rainy season at ND location, which is an industrial zone of textile that could be an important source of fibers, as confirmed by Deng et al. (2020). These authors found that MP abundance in surface water in the textile industrial area was twice higher than other ones. The Red River flow is about 8 times higher in the rainy season than in the dry season, *i.e.* 8,600 and 1,085

m<sup>3</sup>/s, respectively (Dang et al., 2010). At Ba Lat estuary, the flow is 3 times higher in the rainy season compared to the dry one (841 and 247 m<sup>3</sup>/s, respectively, Le et al., 2023). The decrease of salinity measured at ND and TB during the sampling campaign (Table 1) also depicted the dilution of seawater by freshwater inputs. The change of flows is a probable explanation of the plastic-like item concentration increase during the rainy season, *e.g.* multiplied by 9 times at ND site. These results suggest the river discharge had a potential influence source on plastic-like items from the continental to the coastal zones. In the same way, Strady et al. (2020) estimated up to more than 10<sup>14</sup> items annually discharged by the Saigon River to downstream coastal zone. No influence of the types of marine environments was observed, the plastic-like item concentrations being not significantly different between estuarine zones (TB and ND in the dry season) and coastal areas (QN and HP). Except the textile industry impact on the plastic-like items in the rainy season discussed above (ND-1), no influence of other anthropogenic activities on the plastic-like item concentrations was highlighted, since there were no other significantly different results among the sites.

Table 4  
Studies about microplastics reported in seawater and marine sediment of Southeast Asia

Sample type	Location	Concentration <sup>(1), (2)</sup>	Size range predominant (µm)	Reference
Seawater	Kuala Nerus and Kuantan, Malaysia	0.13–0.69 items/L	200-5,000	Khalik et al., 2018
	Semarang, Indonesia	700 – 11,100 items/L	270-1,280	Khoironi et al., 2020
	Quang Ninh, Vietnam	0.4–28.4 items/m <sup>3</sup>	250-1,000	Strady et al., 2021
	Indonesia seas	0-0.17 items/m <sup>3</sup>	> 300	Yuan et al., 2023
	Northern coasts, Vietnam	3-303 items/m <sup>3</sup>	100–500	<b>This study</b>
Sediment	Sembawang and Changi, Singapore	31–60 items/kg ww	1,000–5,000	Curren and Leong, 2019
	Southern Vietnam	0-295 items/kg dw	500-5,000	Hien et al., 2020
	Semarang, Indonesia	8,000–49,000 items/kg dw	< 5,000	Khoironi et al., 2020
	Jakarta Bay, Indonesia	18,405 – 38,790 items/kg dw	30 – 5,000	Manalu et al., 2017
	Da Nang, Vietnam	5,100 – 11,000 items/kg dw	300-5,000	Nguyen et al., 2020
	Ho Chi Minh City, Vietnam	0–7 items/kg dw	500-5,000	Nhon et al., 2021
	Vung Tau, Vietnam	1542–2024 items/m <sup>3</sup>	250-1,000	Strady et al., 2021
	Northern coasts, Vietnam	63–955 items/kg dw	100–500	<b>This study</b>

(1): Concentrations of plastic-like items; (2): ww = wet weight, dw = dry weight.

Regarding marine sediment samples, the Table 4 shows that plastic-like item concentrations found in this study were lower than those observed in Indonesian coasts (Khoironi et al., 2020; Manalu et al., 2017) and higher than those found on Singapore coast (Curren and Leong, 2019). At the national level, the marine sediment levels measured in

this study were similar to those reported in Vung Tau (Strady et al., 2021), higher than those found at the Southern (Hien et al., 2020; Nhon et al., 2021) but lower than those reported at the center of Vietnam (Nguyen et al., 2020).

In comparison to the results obtained for the seawater, the river flows seemed to be less influent on the plastic-like item levels in sediments, at least over the studied period. No significant different results were found between the season, as well as between the studied sites.

MP contamination was known as linked to the granulometry in river (Dhivert et al., 2022) and marine sediment (Mendes et al., 2021). Sediments with fine grained size are usually more contaminated. The granulometry analysis of the sediments collected at QN showed larger grain size than the other sites, and similar on both studied seasons. However, plastic-like item concentrations at QN were not significantly lower than in the other sites. Since no significant difference of the plastic-like item levels was observed between the sites, the granulometry seemed to not be an influencing factor of item distribution.

## 4.2 Typology of Plastic-like Items found in water and sediments

The plastic-like item color seemed similar between seawater and marine sediment samples. Sum of three colors (purple, red and white) is 86.83 and 86.36% in seawater and marine sediments, respectively. Looking on the size, items found in seawater seemed slightly bigger than those ones in marine sediment. Smaller fractions (less than 300  $\mu\text{m}$ ) reached 72.85% in marine sediment compared to 58.52% in seawater. Especially, items with size higher than 1 mm were measured preferentially in seawater than in sediments, representing 6.41% compared to 1.27% of the total items, respectively. No difference of item forms was observed between seawater and sediment samples. There were both predominant fibers (> 90%) and some fragments. MP fibers were found predominant in marine environment as indicated by many studies (Cheang et al., 2018; Lots et al., 2017; Zhao et al., 2018).

The results about the identification of polymer from the fragments found in seawater and sediments of Vietnamese marine environment, *i.e.* PE, PP and PS, were consistent with the production, uses and also with other studies dealing with marine environment MP contamination (Abidli et al., 2018; Kazour et al., 2019; Saeed et al., 2020). The average size was  $728 \pm 733 \mu\text{m}$  ( $n = 19$ ) and  $1,108 \pm 1568 \mu\text{m}$  ( $n = 3$ ) in seawater and sediments, respectively. Interestingly, the size of MPs PS is smaller than other plastic types. It was  $140 \pm 17 \mu\text{m}$  for PS ( $n = 2$ ) compared to  $622 \pm 123 \mu\text{m}$  for PE ( $n = 2$ ) and  $869 \pm 905 \mu\text{m}$  for PP MPs ( $n = 18$ ). These polymers correspond to materials with a density lower than the seawater density ( $1.025 \text{ g/cm}^3$ ). Hence, their presence in marine sediment could be the result of bio-fouling process (micro-organisms colonization, digestion by marine animals) or particles aggregation. MP sedimentation process in marine environment is known depending on sounding environment and MP characteristics (density, size, surface area). As fiber form dominates in both matrices, our data are not enough to conclude on the relation between the form and sedimentation process. The particle size displayed an important role on the sinking capacity. We found that items size was slightly smaller in marine sediment samples which is in line with laboratory experiments. Indeed, Enders et al, (2015) experimented on PE and PP MPs in seawater with different sizes. The authors demonstrated that MPs larger than 1 mm were floating on the surface while smaller MPs were found as sinking (24 m of depth for 100- $\mu\text{m}$  MP).

## 5. Conclusion

MP contamination was evaluated in the marine environment from the Northern coast of Vietnam. Two matrices (seawater and sediment) were investigated at 4 locations and 2 seasons. Plastic-like items abundances and their characteristics in seawater and marine sediment have been shown. Quantitative results showed levels of 3 to 303 plastic-like items/ $\text{m}^3$  in seawater and 63 to 955 plastic-like items/kg dry weight in sediment samples. A seasonal

effect was demonstrated for seawater between the rainy and dry seasons, probably according to river discharges. Less variation was observed in sediments as no significant difference was found between seasons and the 4 studied locations. Fibers and colored MP were predominant in both studied matrices. Most of them are less than 300 µm in size. Three plastic types were identified for fragments, including PE, PP and PS. MP size highly varied in both of seawater and marine sediments, PS MPs appeared to be smaller than PE and PP MPs.

## Declarations

### Authors Contributions

Ngoc Nam Phuong: project administration, conceptualization, supervision, sampling and editing, Thi Thuy Duong: conceptualization, supervision, sampling and editing, Quoc Tuan Pham: methodology, experimental section, data analysis, sampling and original draft writing, Thi Xuan Thinh Ngo: methodology, experimental section, data analysis, sampling and original draft writing, Thi Minh Diep Nguyen: methodology, experimental section, data analysis, sampling and original draft writing, Ngoc Anh Phuong: methodology, experimental section, data analysis and original draft writing, Thi Phuong Quynh Le: methodology, experimental section, data analysis and original draft writing, Thanh Nghi Duong: methodology, experimental section, data analysis and original draft writing, Elie Dhivert: conceptualization, supervision and editing, Aurore Zalouk-Vergnoux: conceptualization, supervision and editing, Laurence Poirier: conceptualization, supervision and editing, Johnny Gasperi: conceptualization, supervision and editing. All authors have read and agreed to the published version of the manuscript.

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### Data Availability

All data are included within the article and the supplementary materials.

All authors have read, understood, and have complied as applicable with the statement on “Ethical responsibilities of Authors” as found in the Instructions for Authors.

### Ethics approval

Not applicable.

### Competing interests

The authors declare that there are no conflicts of interest.

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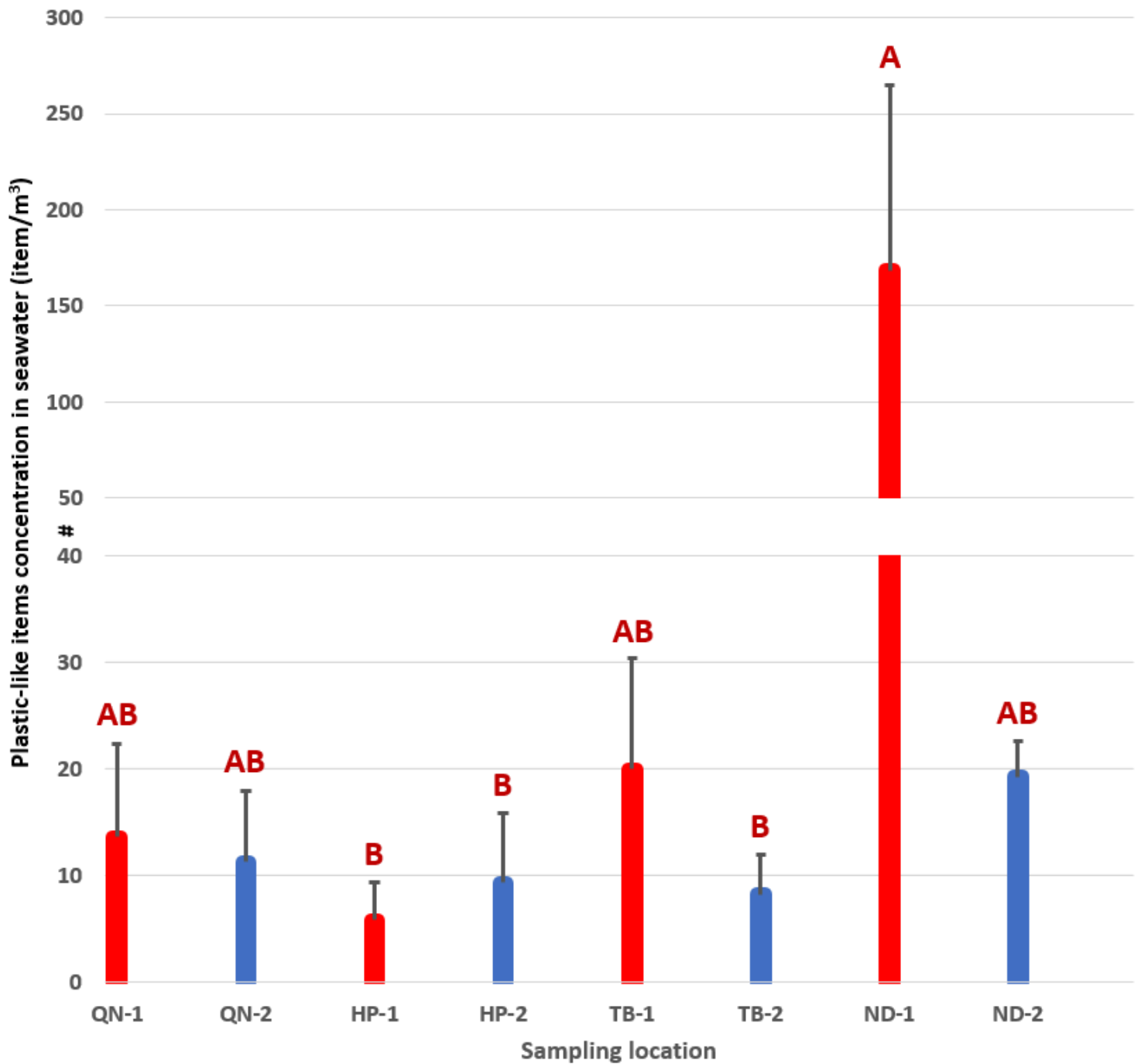
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## Figures



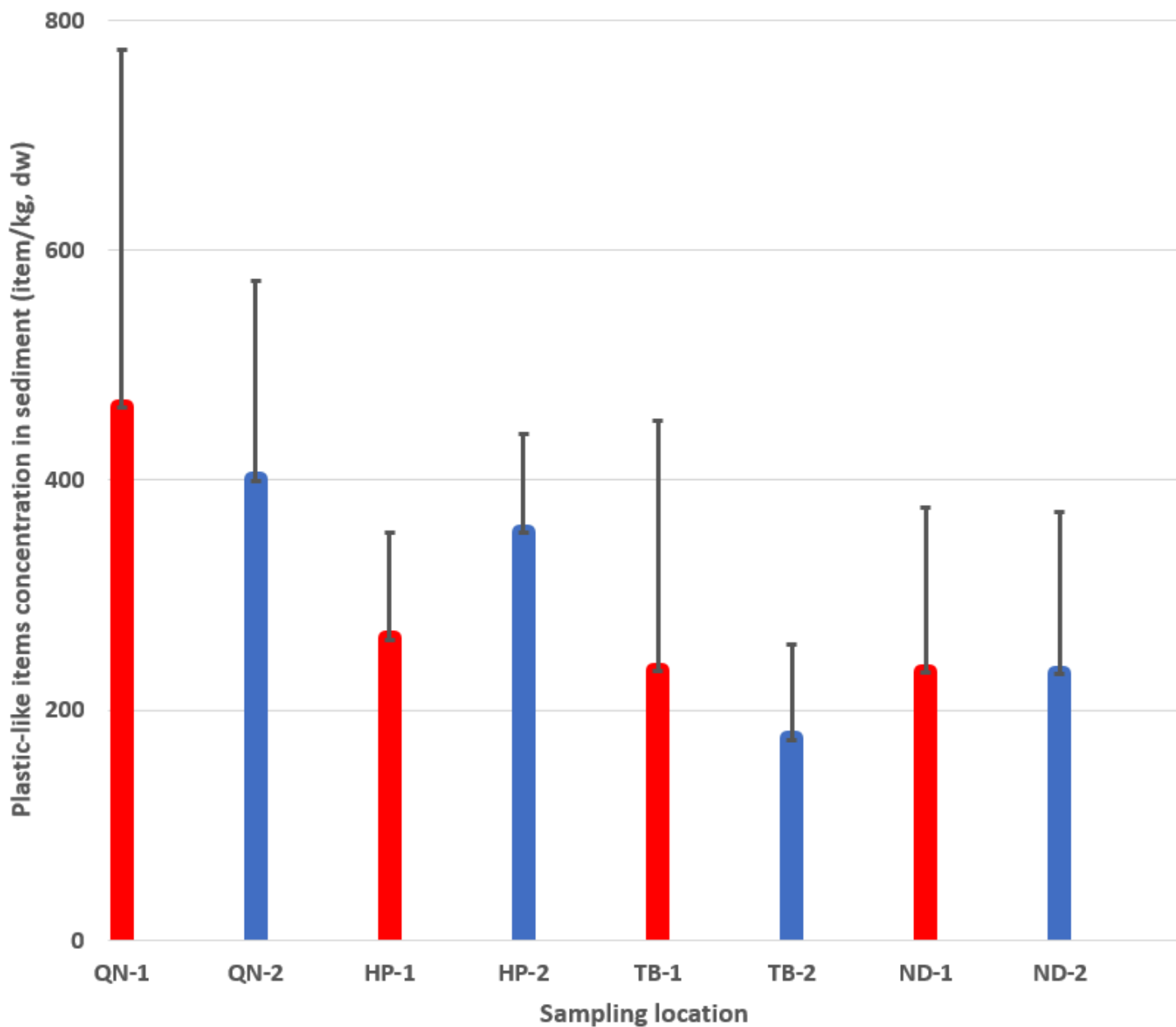
Figure 1

Four sampling locations in Northern Vietnam coast, Quang Ninh, Hai Phong, Thai Binh and Nam Dinh. Seawater and marine sediments were sampled in July 2020 and January 2021.



**Figure 2**

Average concentrations and standard deviations of plastic-like items found in seawater samples (items/m<sup>3</sup>) from the 4 studied locations: Quang Ninh (QN), Hai Phong (HP), Thai Binh (TB) and Nam Dinh (ND); n = 5 for each location. Seawater sampled in July 2020 and January 2021 are in red and blue, respectively as well as labelled with -1 and -2 in the sample labels. Significant differences between sampling locations and seasons are displayed with different capital letters ( $p < 0.05$ ; Kruskal-Wallis test).



**Figure 3**

Average concentrations and standard deviations of plastic-like items found in sediment samples (item/kg, dw) from the 4 studied locations: Quang Ninh (QN), Hai Phong (HP), Thai Binh (TB) and Nam Dinh (ND); n = 5 for each location. Sediments sampled in July 2020 and January 2021 are in red and blue, respectively, as well as labelled with -1 and -2 in the sample labels.

## Supplementary Files

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