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Abstract Microplastics (MPs) are small (< 5 mm) plastic particles that are widely found in marine, freshwater, terrestrial and atmospheric environments. Due to their prevalence and persistence, MPs are considered an emerging contaminant of environmental concern. The separation and quantitation of MPs from freshwater sediments is a challenging and critical issue. It is necessary to identify the fate and sources of MPs in the environment, minimise their release and adverse effects. Compared to marine sediments, standardised methods for extracting and estimating the amount of MPs in freshwater sediments are relatively limited. The present study focuses on MP recovery efficiency of four commonly used salt solutions (NaCl, NaI, $CaCl_2$ and $ZnCl_2$) for isolating MPs during the density separation step from freshwater sediment. Known combinations of artificial MP

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particles (PS, PE, PVC, PET, PP and HDPE) were spiked into standard river sediment. Extraction using NaI, $ZnCl_2$ and NaCl solutions resulted in higher recovery rates from 37 to 97% compared to the $CaCl_2$ solution (28–83%) and varied between polymer types. Low-density MPs (PE, HDPE, PP and PS) were more effectively recovered (> 87%) than the denser polymers (PET and PVC: 37 to 88.8%) using NaCl, NaI and $ZnCl_2$ solutions. However, the effective flotation of $ZnCl_2$ and NaI solutions is relatively expensive and unsafe to the environment, especially in the context of developing countries. Therefore, considering the efficiency, cost and environmental criteria, NaCl solution was selected. The protocol was then tested by extracting MPs from nine riverine sediment samples from the Red River Delta. Sediments collected from urban rivers were highly polluted by MPs (26,000 MPs items·kg^{-1} DW) compared to sediments located downstream. Using a NaCl solution was found to be effective in this case study and might also be used in long-term and large-scale MP monitoring programmes in Vietnam.

Keywords Microplastics · Density separation · Riverine sediment · Urban rivers · Vietnam

Introduction

Plastic, one of the revolutionary inventions of the twentieth century, has provided great convenience for domestic and industrial purposes and has gradually become ubiquitous. Plastics are long lasting, lightweight, inexpensive and easily moulded and manufactured. Consequently, the total annual global plastic production has increased approximately 233 times from 1.5 to 359 million tons over the past 68 years (Europe, 2018). At the global level, only 9% of all plastics produced are recycled and 12% are incinerated, representing a relatively small percentage compared to the amount that ends up in dumping sites or discarded in the environment (Geyer et al., 2017). Owing to their durability, low recycling rate and accidental loss due to poor waste management, a considerable amount of plastic waste has entered and persisted in both terrestrial and aquatic ecosystems. Plastic pollution has become a global environmental concern and plastic debris has been broadly reported worldwide, including in remote areas such as polar regions, where human activities are limited, and the Tibet Plateau (Jiang et al., 2019), Arctic (Peeken et al., 2018), Trindade Island (de Souza Petersen et al., 2016) and at the bottom of deep seas (Taylor et al., 2016; van Cauwenberghe et al., 2013). The plastic found in aquatic environments may have multiple sources related to anthropogenic activities via wastewater discharge, sewer system leaks and natural processes, including wind, rainfall, atmospheric transportation and water flow, although the plastic will ultimately end up in the ocean. At the global level, approximately 80% of ocean plastics come from rivers, ahind the remaining are from aquatic sources (Jambeck et al., 2015). Plastic waste enters ecosystems of all shapes and sizes. It can be broken down into smaller pieces under various physical, chemical and biological processes and is defined as macroplastics (> 25 mm), mesoplastics (5–25 mm), microplastics (MPs) (< 5 mm) and nanoplastics (< 0.1 μm) (Arthur et al., 2009; GESAMP, 2019). Based on their origin, MPs are divided into two subcategories: primary MPs that are manufactured as particulates and widely found in textiles, clothing fibres, cosmetics and medical products (Cole et al., 2011) and secondary MPs that are produced because of larger plastic degradation. Due to their small size, MPs can be easily ingested by a wide range of organisms in aquatic systems (Andrady, 2011), which causes them to accumulate throughout the aquatic food web (Batel et al., 2016). Moreover, MPs can absorb various pollutants from their nearby environments, such as heavy metals (Dong et al., 2020; Wang et al., 2020), dissolved organic matter (Abdurahman et al., 2020) and antibiotics (Li et al., 2018). These contaminants may endanger and negatively affect aquatic organisms and may affect human health (Paul-Pont et al., 2016; Rainieri et al., 2018).

MP studies in freshwater environments have received less attention than those in marine environments (Eerkes-Medrano et al., 2015; Lambert & Wagner, 2018; Thompson et al., 2009; Wagner et al., 2014). However, it has been suggested that MP pollution in freshwater is equal to or more serious than that in marine environments (Wu et al., 2018). As a part of freshwater systems, rivers are one of the main pathways for transporting inland macro- and MP debris to marine environments, accounting for 1.15 to 2.41 million tonnes per year (Lebreton et al., 2017). After being exposed to the environment, MPs can be found

floating and suspended in water columns, and can also sink and accumulate in riverbeds (Barnes et al., 2009; Kowalski et al., 2016) and pollute freshwater environments. To date, standardised methods for extracting and estimating the amount of MPs in sediment samples are limited, especially for river sediments enriched with high organic matter, despite efforts to report the occurrence of MPs in sediments from European (Scherer et al., 2020) or Chinese rivers (Zhang et al., 2020). Applying different protocols for MP extraction from sediments limits the comparison between studies. In general, the process used to determine MP concentrations in sediment samples comprises extraction, separation, identification and quantification (Shim et al., 2017). Several methods have been developed for MP extraction from marine and beach sediment samples, including filtration, sieving, density separation, flotation, chemical decomposition, electrostatic separation, elutriation column optimisation and magnetic extraction using coated Fe nanoparticles to magnetise plastics (Felsing et al., 2018; Grbic et al., 2019; Hengstmann et al., 2018; Hidalgo-Ruz et al., 2012; Müller et al., 2020; Nakajima et al., 2019; Phuong et al., 2021). However, density separation is the most commonly applied method for isolating MPs from sediment and is based on the specific density difference between MPs and sediments to separate less dense plastic polymers from denser sediment particles (Coppock et al., 2017; Phuong et al., 2021). Various density salt solutions have been used to extract MPs from marine sediment samples, and a wide range of recovery efficiencies have been found when using single or combined salt solutions for the extraction of MPs within the same environmental compartment (Claessens et al., 2013; Nuelle et al., 2014; Quinn et al., 2017).

Therefore, as MPs have become a global concern, a validated sample preparation for freshwater sediment is required to better understand the extent of MP pollution and its impact. Riverine sediments usually present higher organic matter content than marine sediment samples. The treatments used should thus include a chemical digestion step (such as oxidative, acidic and alkaline) before the density separation step to ensure an efficient extraction rate without degrading the polymers. One of the main priorities for MP quantification in natural environments is the selection of an affordable, easy and cost-effective protocol. Therefore, the purpose of this study was to investigate the most effective density separation treatment to extract MPs from riverine sediments according to the criteria described above. First, the extraction efficiency using four salt solutions (sodium chloride (NaCl, 1.2 $g \cdot cm^{-3}$); sodium iodide (NaI, 1.6 $g \cdot cm^{-3}$); calcium chloride ($CaCl_2$, 1.4 $g \cdot cm^{-3}$) and zinc chloride ($ZnCl_2$, 1.5 $g \cdot cm^{-3}$)) was evaluated on standard river sediment spiked with six MP polymer types. Then, the chosen density separation solution was applied to quantify MP contamination in riverine sediments sampled from the Red River Delta.

Material and methods

Analytical development

Preparation of a standard riverine sediment

Riverine sediment from the Red River, the largest river in northern Vietnam, was used as a standard sample for developing an analytical method for MP recovery tests. The sediment was collected at the Son Tay site located in the Red River (R) (longitude 21°15′ N, latitude 105°50′ E) using a grab sampler (0.1 m^2) in June 2020 (Fig. 1 and Table 1). To ensure that the sediment sample did not contain MPs and could be used as standard sediment, a sub-sediment sample was taken, and MPs were removed following the protocol of Strady et al. (2021). The sub-sample was homogenised and oven-dried at 40 °C for 72 h to a constant weight. Then, the dried sediment sample was sieved through a 1-mm mesh sieve to remove all the litter and macroscopic particles, such as shells, leaves and plastic (e.g. > 1 mm). Subsequently, 10 g of dried sediment was placed in a 250-mL beaker and digested using 30% H_2O_2 solution. The beaker was covered with aluminium foil and placed on a heating plate at 40 °C for 3 h for digestion. Saturated sodium chloride solution (NaCl, 1.2 $g \cdot cm^{-3}$) was used to isolate MPs. The oxidised sediments were transferred into a clean 250-mL glass beaker for conducting the density separation with filtered, saturated NaCl solution by the overflow producing technique. The separation step was repeated at least five times to ensure that all the plastic items were removed. All overflowed solutions were filtered with GF/A filters (pore sizes of 1.6 µm) using a glassware filtration unit. The density solution was examined under a microscope

Fig. 1 Map of nine sampling location in urban rivers, Vietnam

(Leica MZ12 stereomicroscope at 16–160-fold magnification) with a camera attached for the presence of MPs. When MPs were no longer present in the flotation solution, the oxidised sediment was oven-dried at 40 °C for 72 h to a constant weight. The "standard riverine sediment" or "clean sediment" was then stored in a glass bottle for further experiments.

Table 1 Information on samples sites and GPS position in the Red, To-Lich, Nhue and Day rivers

Site	Location	Latitude (°N)	Longitude (°E)
R	Son Tay, Red River	21.1561	105.5058
TL	HQV, To-Lich River	21.0400	105.8061
N1	CauDen, Nhue River	20.9709	105.7824
N2	MyHung, Nhue River	20.5444	105.4807
N3	Ba Da, Nhue River	20.5671	105.9219
D1	Phung, Day River	21.0752	105.6451
D2	CauQue, Day River	20.5745	105.8726
D3	CauDo, Day River	20.5158	105.9115
D4	DoThong, Day River	20.2174	106.0451

Preparation of density separation solutions

Four density separation solutions were investigated: sodium chloride (NaCl, 1.2 $g \cdot cm^{-3}$); sodium iodide (NaI, 1.6 $g \cdot cm^{-3}$); calcium chloride ($CaCl_2$, 1.4 $g \cdot cm^{-3}$) and zinc chloride ($ZnCl_2$, 1.5 $g \cdot cm^{-3}$). Saturated solutions of NaCl, NaI, $CaCl_2$ and $ZnCl_2$ were prepared under a fumehood by dissolving the salt powders in distilled water using a magnetic stirrer plate (Table 2). Each solution was then vacuum filtered using a 1.2-µm glass fibre (GF/A, Whatman) to remove any MP and remaining salt particles prior to use and was stored in a pre-cleaned glass bottle at room temperature (25 °C).

Table 2 Salt solutions used in microplastic recovery extraction test with their density, amount added, toxicity and price

Density solutions	Density (g cm −3)	Amount added to 1 L (g)	Toxicity/severity of hazard	Price (USD)
Sodium chloride (NaCl)	1.2	335	Low/none	26.1 for 1 kg
Sodium iodide (NaI)	1.6	1000	High/danger, very toxic to aquatic life, cause serious eye irritation and skin irritation	495.7 for 1 kg
Calcium chloride ($CaCl_2$)	1.4	1400	Warning, cause serious eye irritation	43 for 500 kg
Zinc chloride ($ZnCl_2$)	1.5	972	High/danger, very toxic to aquatic life, severe skin burns and eye damage	204.8 for 1 kg

*Prices from Merck in Sept 2021

Preparation of microplastic spiked samples

The standard riverine sediment prepared as mentioned above was spiked with different polymer particles to assess the MP recovery efficiency of the four saturated salt solutions. Six polymers widely used in daily life, including polystyrene (PS), polyethylene (PE), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polypropylene (PP) and high-density polyethylene (HDPE), were selected (Fig. 2 and Table 3). The colour of the MPs was chosen to ensure that they could be easily distinguished and counted for the test experiments. The tested polymer particles were carefully cut into fragments using a grinder. After cutting, all plastic particles were sieved through a metal sieve to ensure the size was less than 500 µm and then transferred to a sealed glass bottle. The selected MP particles in the range of 300–500 µm were kept and used for sample spiking. Each plastic fragment's dimensions were measured using a stereomicroscope (Leica MZ12 stereomicroscope at 16–160-fold magnification) equipped with image analysis software (LAS software®).

Preparation of spiked sediment sample

The MP-spiked samples were prepared from 10 g of standard riverine sediment sample to which a known number of prepared MP particles (20 particles of each MP type, 120 combined particles per sample) were

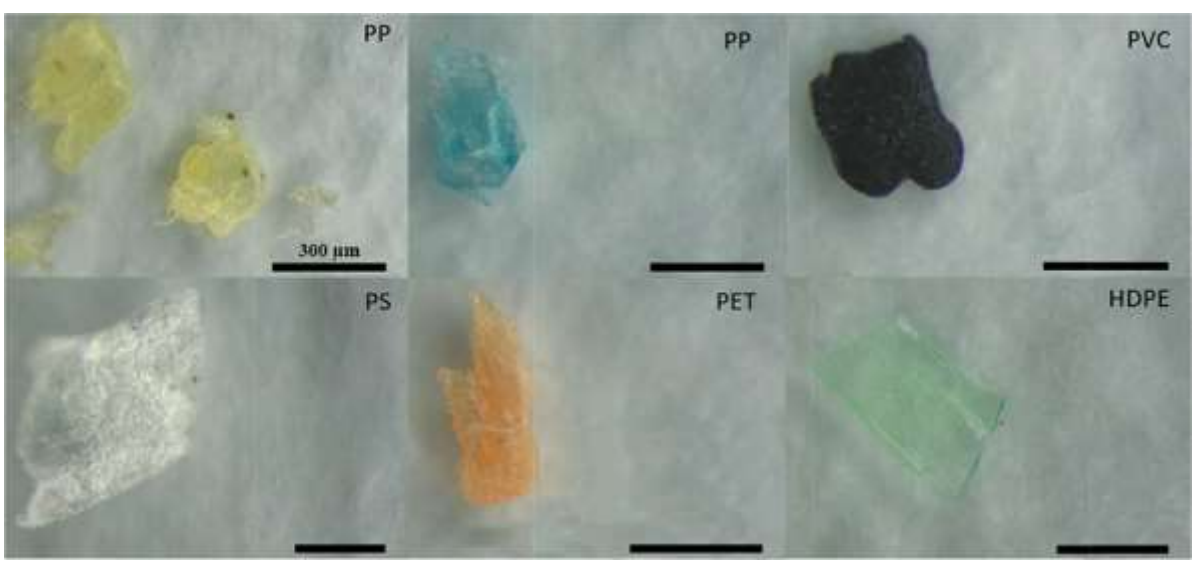

Fig. 2 Stereomicroscope images of polymer particles used in the density separation test including polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polystyrene (PS), polyethylene terephthalate (PET) and high-density polyethylene (HDPE)

Table 3 Properties of microplastic (polymer types, sources, colours) used in the present study

Polymers	Density ($g \cdot cm^{-3}$)	Colour	Source
Polyvinyl chloride (PVC)	1.1–1.35	Black	Electrical cable
Polyethylene terephthalate (PET)	1.38–1.41	Orange	Soft drink bottle
Polystyrene (PS)	0.96–1.04	White	Food container
High-density polyethylene (HDPE)	0.97	Green	Supermarket bag
Polyethylene (PE)	0.89–0.98	Yellow	Grinded beached yellow fish box (CRT 171, Carat GmbH)
Polypropylene (PP)	0.85–0.946	Blue	Plastic container

added or spiked. To remove the added MPs and calculate the removal efficiency, the MP-spiked sediment samples were treated based on the protocol described above (Strady et al., 2021) with changes at the separation density steps in which four different saturated salt solutions were used: NaCl (1.2 $g \cdot cm^{-3}$), NaI (1.6 $g \cdot cm^{-3}$), $ZnCl_2$ (1.5 $g \cdot cm^{-3}$) and $CaCl_2$ (1.4 $g \cdot cm^{-3}$). The number of MP particles detected after density separation was used to calculate the percentage of MP recovery.

MP recovery efficiencies = [number of particles extracted/number of particles spiked] × 100.

Three replicates were used for each density separation solution experiment.

Case study: sampling and analysis

The selected density separation solution was used to extract and analyse MPs from different riverine sediment samples. River sediment samples were collected from nine sampling sites in the Red, To-Lich, Nhue and Day rivers (located in the Red River Delta) in July 2020 using a grab sampler (0.1 m^2) (Fig. 1 and Table 1). The site sampled covers four urban rivers in Hanoi with different urbanisation levels and land uses. Site R (at Son Tay) is representative of the Red River section running through Hanoi City. The To-Lich River, with a length of 17 km and water flow of approximately 30 $m^3 \cdot s^{-1}$, has been considered as the wastewater collector system of Hanoi City (approximately 1,200,000 $m^3 \cdot day^{-1} \cdot night^{-1}$, of which approximately 20% could be treated). In this river basin, the population density is very high (2,279 inhabitants$\cdot km^{-2}$). The Nhue River, a peri-urban river with a total basin area of approximately 1,075 km^2 located in the Red River Delta, is supplied by water from the Red River through Lien Mac sluice and receives most of the untreated domestic and industrial wastewater from the Hanoi metropolitan area via the To-Lich River. The Day River, a distributary of the Red River, has a length of 240 km and runs through Hanoi, Ha Nam, Nam Dinh and Ninh Binh city/provinces and discharges to the sea at the Day River mouth. At each sampling site, sediment samples were collected in triplicate from the middle, left and right banks of the river using a grab sampler and then mixed to obtain approximately 3 kg of wet sediment per site (three replicates were conducted per site/three mix samples per site). The collected sediment samples were placed in an aluminium container and wrapped in aluminium foil. Sub-samples of wet sediment were dried at 40 °C for 72 h, followed by sieving with a mesh sieve of 1 mm. To remove natural organic materials, 10 g of dried sediment was digested with 30% hydrogen peroxide solution, and then the MPs were separated using saturated NaCl solution as described above. The MPs were categorised into three types of geometric shapes: fragments (including films and foams), fibres and pellets (Strady et al., 2021). LAS software® was used to measure the morphology of the collected MPs: areas were measured for fragments, diameter and length were measured for fibres and area and perimeter were measured for pellets. The size observation range was 300–5,000 µm and was derived from the protocol used and the recommendations of GESAMP (2019) when a systematic and/or representative analysis of the nature of the particles cannot be performed. More precisely, this size range considers fragment areas from 45,000 μm^2 (300 µm × 150 µm) to 25,000,000 μm^2 (5000 µm × 5000 µm). The MP concentration was expressed as particles per kilogramme of dry sediment (particles$\cdot kg^{-1}$ DW).

Polymer identification

The chemical composition of the particles extracted from river sediments that were suspected to be MP was determined by Raman spectroscopy (HORIBA XploRA Plus) coupled to the Raman Spectra Database Collection KnowItAll®. The laser power used was 632.8 nm, the wavelength ranged from 1700 to 700 cm^{-1} and the spectrum acquisition lasted for 60 s and two accumulations. Ten percent of the total suspected MPs were selected randomly and picked up (using a small metal tool and ethanol) for Raman spectroscopy.

Analytical conditions

All steps were carried out inside a flow cabinet to avoid MP contamination from the ambient environment during the experiments. The flow cabinet was kept in a clean condition and was wiped with alcohol. All the liquids used, such as deionised water (to make the solutions and clean the tools) and density solutions, were prefiltered through GF/A filters (1.6 µm) to avoid contamination and were kept in clean glassware. Control blanks (n = 3) were run in parallel to the sample process (Dehaut et al., 2019), and an average of two fibres per sample was found which can be considered negligible in comparison to the number of MPs detected in field sediment samples.

Results and discussion

MP recovery efficiencies using different salt solutions

The extraction efficiencies of the six spiked MP polymers (PS, PE, HDPE, PVC, PET and PP) performed with standard riverine sediment using the four different density separation solutions (i.e. $ZnCl_2$, NaI, NaCl and $CaCl_2$) ranged from 28.3% ± 3% to 97% ± 3% (Fig. 3). The results showed variations between polymers and between the density separation solutions. The average extraction efficiency was 83.9% for $ZnCl_2$, 82.2% for NaI, 80% for NaCl and 68% for $CaCl_2$, making $CaCl_2$ the least efficient solution (Supplementary Table 1). This result was unexpected as $CaCl_2$ has a higher density than NaCl, and we expected a higher separation and recovery efficiency with $CaCl_2$. The observed low recovery rate of the $CaCl_2$ solution could be attributed to its high viscosity and the remaining flocculation of calcium ions (Ca^{2+}) with organic matter after the oxidation

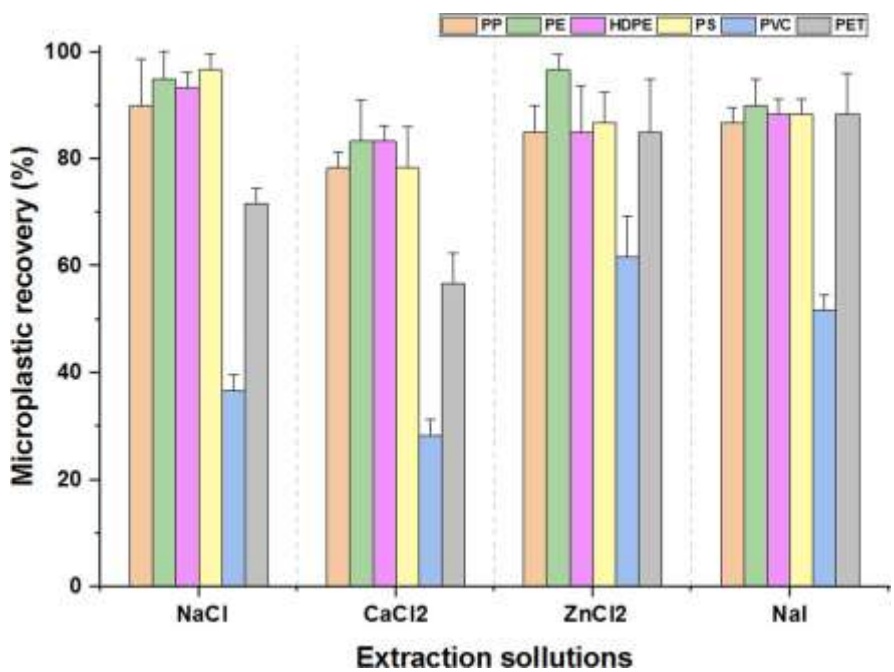

Fig. 3 Microplastic extraction recovery efficiencies of six polymers (PET, PP, PS, PVC, PE, HDPE) using four different density separation solutions ($CaCl_2$, NaCl, NaI and $ZnCl_2$) during the density separation step

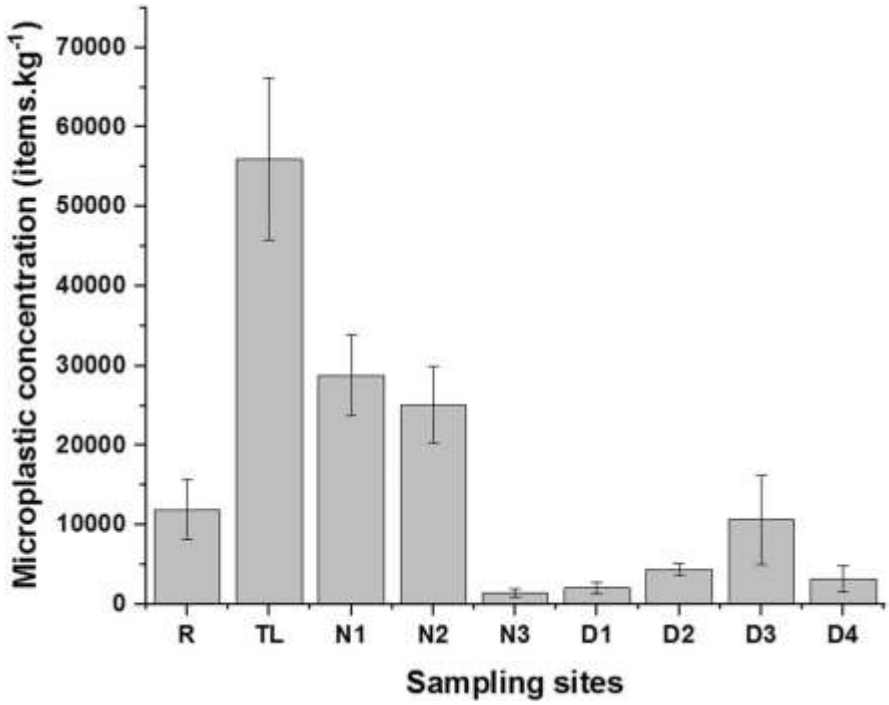

Fig. 4 MP concentrations in sediment collected at the nine sampling sites ($n = 3$)

process. The filtering of the $CaCl_2$ solution was extremely slow and time consuming compared to the filtration of the NaI, NaCl and $ZnCl_2$ solutions. A low extraction efficiency using the $CaCl_2$ solution (densities of 1.35–1.4 g·mL^{-1}) was also reported for PE particles (49–62%) by Stolte et al. (2015).

Table 4 MP concentrations in sediment samples from fresh aquatic environments reported worldwide

Freshwater systems	Sites	Density separation	MP abundance (items/particles, dry weight kg^{-1})	References
River	Andean rivers, Ecuador	NaCl	14.3–186.5	Donoso et al. (2020)
	Rhine and Main rivers, Germany	NaCl	Rhine: 228–3763 Main: 786–1368	Klein et al. (2015)
	Meuse River and canal, Netherland	NaCl	Meuse: 1400–4900 Canals: 68–10,500	Leslie et al. (2017)
	Ganga River, India	NaCl	17–36	Singh et al. (2021)
	Beijiang River, China	NaCl	178–544	Wang et al. (2017)
	Bloukrans River system, South Africa	NaCl	13.3–563.8	Nel et al. (2018)
	Qin River, China	NaCl	0–97	Zhang et al. (2020)
	Mersey/Irwell River, UK	NaCl-NaI	300–75,000	Hurley et al. (2018)
	Pearl River, China	NaCl	80–9597	Lin et al. (2018)
	Liangfeng River, China	NaCl	6,950–149,350	Xia e al. (2021)
	To-Lich, Nhue, Day rivers, Vietnam	NaCl	1,450–55,950	This study
Lake	Bizerte lagoon, northern Tunisia	NaCl	3,000–18,000	Abidli et al. (2017)
	Lake Bolsena and Chiusi, Italy	NaCl	Bolsena: 109–117 Chiusi: 205–266	Fischer et al. (2016)
	Taihu Lake, China	NaCl	11–234.6	Su et al. (2016)
	Poyang Lake, China	NaCl	54–506	Yuan et al. (2019)

Among the other three density separation solutions, NaCl, NaI and $ZnCl_2$, no difference in recovery efficiency was observed ($p > 0.05$; Supplementary Table 2), and our results confirmed the previous good recovery efficiency observed (Claessens et al., 2013; Coppock et al., 2017; Fries et al., 2013; Imhof et al., 2012). The NaCl solution with a density of 1.2 $g \cdot cm^{-3}$ was reported to have an 80 to 100% MP recovery rate (Fries et al., 2013), while the recovery ranged from 55 to 90% for a saturated NaCl solution of 1.17 $g \cdot cm^{-3}$ (Quinn et al., 2017). These last authors noted MP recovery efficiencies of 91% using a sodium iodide (NaI) saturated solution of 1.57 $g \cdot cm^{-3}$ and 99% using a saturated $ZnBr_2$ solution of 1.71 $g \cdot cm^{-3}$ (Quinn et al., 2017). Some authors observed that using special devices such as the Munich Plastic Sediment Separator (Imhof et al., 2012) and the Sediment Microplastic Isolation Unit (Coppock et al., 2017) improved the extraction efficiency of NaI and $ZnCl_2$ solutions, while an elutriation device allowed a recovery efficiency with 1.6 $g \cdot cm^{-3}$ saturated NaI of 65.8 to 100% (Claessens et al., 2013).

It is important to note that the recovery efficiencies differed for the MP types and density separation solutions used (Fig. 3). In the cases of HDPE (0.97 $g \cdot cm^{-3}$), PE (0.89–0.98 $g \cdot cm^{-3}$), PS (0.96–1.04 $g \cdot cm^{-3}$) and PP (0.85–0.946 $g \cdot cm^{-3}$), all considered as light MPs, their fragments were easily and effectively extracted (e.g. 75 to 97%, Fig. 3), due to their ability to float, with all tested solutions: $ZnCl_2$ (1.5 $g \cdot cm^{-3}$), NaI (1.6 $g \cdot cm^{-3}$), NaCl (1.2 $g \cdot cm^{-3}$) and $CaCl_2$ (1.4 $g \cdot cm^{-3}$). In contrast, the denser polymers PVC (1.1–1.35 $g \cdot cm^{-3}$) and PET (1.38–1.41 $g \cdot cm^{-3}$) presented a lower recovery efficiency (Fig. 3). PVC achieved the lowest extraction efficiency, accounting for 28.3% ± 2.5% for $CaCl_2$, 36.7% ± 3% for NaCl, 52% ± 2.8% for NaI and 61.7% ± 7.5% for $ZnCl_2$. The recovery efficiency of the PET polymer was 57% ± 5.5 for $CaCl_2$, 72% ± 3 for NaCl, 88.3% ± 7.5 for NaI and 85% ± 9 for $ZnCl_2$, with an average value of 75.4%. Both $ZnCl_2$ and NaI recovered significantly more PVC and PET than did the NaCl and $CaCl_2$ solutions ($p < 0.05$), which was not the case for PE, HDPE, PP and PS ($p > 0.05$). These results are consistent with previous observations of a high recovery rate (85–95%) observed on 200–400 µm size PE, HDPE and PS particles using NaCl solutions, which are less effective for PET and PVC (Quinn et al., 2017), and with higher recoveries using several solutions (NaCl, $CaCl_2$, NaBr, $ZnBr_2$, $ZnCl_2$, NaI, NaCl-NaI) for PE, PP, PA and PS rather than PVC, POM

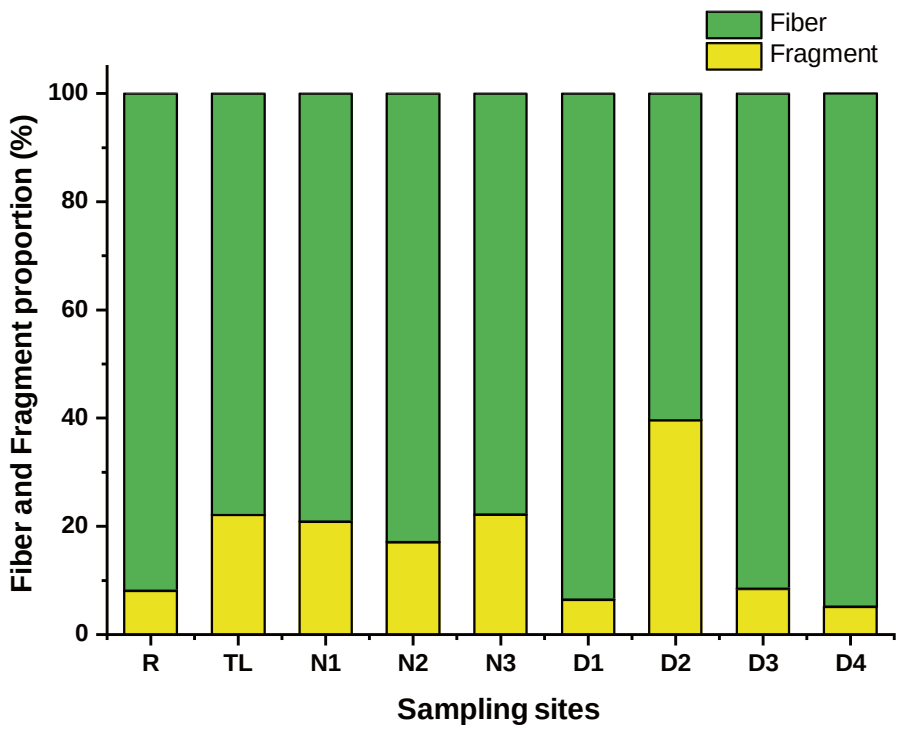

Fig. 5 Proportion of fibre and fragment forms observed in sediment collected at the nine sampling sites

(polyacetal and polyoxymethylene), PET and poly methyl methacrylate (PMMA) (He et al., 2021).

Choosing a solution adapted to the local context

The choice of solution to perform the density separation step depends on its effectiveness in recovering MPs and on other criteria, making the protocol dependent on the studied environment, that is a developing country in this case. The choice should take into account the long-term approach, meaning it should consider the cost of the protocol and sourcing availability of the reagent used, as well as the environmental-friendly status of the reagents. In developing countries, the treatment of hazardous chemicals is limited, as is the protection and treatment of technicians, especially for accidental vapour inhalation or dermal contact. In the present study and in scientific literature (Claessens et al., 2013; Quinn et al., 2017; Radford et al., 2021), despite NaI and $ZnCl_2$ solutions showing higher PVC and PET recovery rates than NaCl solution, the choice of NaI or $ZnCl_2$ was not systematic due to our specific criteria. NaI and $ZnCl_2$ are relatively expensive salts compared to NaCl (Table 2), especially in developing countries where import taxes must be added to the overall cost. Therefore, the cost of the salts will be high when applied to a large number of samples, such as for snapshot campaigns and long-term and large-scale MP monitoring programmes. To reduce the overall cost of using NaI salt, a re-use protocol was developed by Kedzierski et al. (2017) that maintains its high efficiency recovery even after 10 re-uses. However, NaI solution presents the disadvantage of reacting with the filter's cellulose, causing a blackish membrane colour, making it difficult to observe and identify the potential MPs, which will then require a further clean filter transfer procedure (Quinn et al., 2017). Finally, NaI and $ZnCl_2$ are known to be toxic to aquatic life due to their long-lasting effects, and the absence of treating these chemicals could be an issue due to the large volume required for the separation process. $ZnCl_2$ is also harmful for technicians in cases of inhalation and dermal contact that require specific care treatments.

Being affordable and harmless to the environment (Fig. 3), with recovery efficiencies of 36.7 ± 3% for PVC, 71.7 ± 3% for PET, 90 ± 7% for PP, 93.3 ± 2% for HDPE, 95 ± 5% for PE and 96.7 ± 3% for PS, NaCl satisfies our criteria and can be considered suitable for use in the density separation step protocols for developing countries (Strady et al., 2021). Recently, NaCl salt has become increasingly used worldwide for extracting MPs from riverine sediment samples (Cutroneo et al., 2021) and marine sediments (Phuong et al., 2021). However, repeating the density separation step at least three times on the same sample has been shown to improve the MP recovery efficiency, even for denser ones, such as PVC (Horton et al., 2017). Recently, Bellasi et al. (2021) suggested

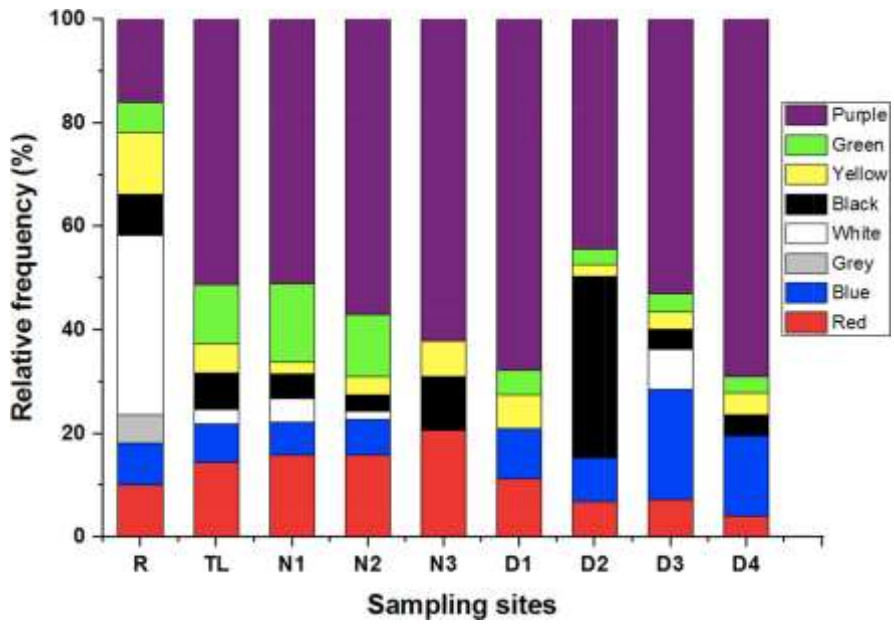

Fig. 6 Proportion of observed MP colour in sediment collected at the nine sampling sites

that using a NaCl/sucrose solution increased its final density and allowed a better recovery of denser polymers such as PVC and PET.

Microplastic recovery using NaCl solution for density separation steps: the case study of the Red, To-Lich, Nhue and Day rivers

Based on the recovery efficiency obtained, MPs in riverine sediments from the Red, To-Lich, Nhue and Day rivers were analysed using the NaCl density separation solution. The MP concentrations (particles kg^{-1} DW) measured at the nine sampling sites are presented in Fig. 4. MP particles were observed at all sampling sites, showing the global MP contamination of riverine sediment, with concentrations ranging from 1,450 ± 494 particles·kg^{-1} at the N3 site (e.g. Ba Da, Nhue River) to 55,950 ± 10,111 particles·kg^{-1} at the TL site (e.g. To-Lich River). In comparison to riverine sediments worldwide extracted using the NaCl density separation solution, the observed MP concentrations in this study were higher than those reported in Europe, America, Africa and Asia (Table 4). In the Red River Delta system, fibres were the most dominant shape found (e.g. 60 to 95%) compared to fragments (e.g. 5 to 22%), as reported worldwide (Alam et al., 2019; Barrows et al., 2018), while pellet forms were not observed (Figs. 5 and

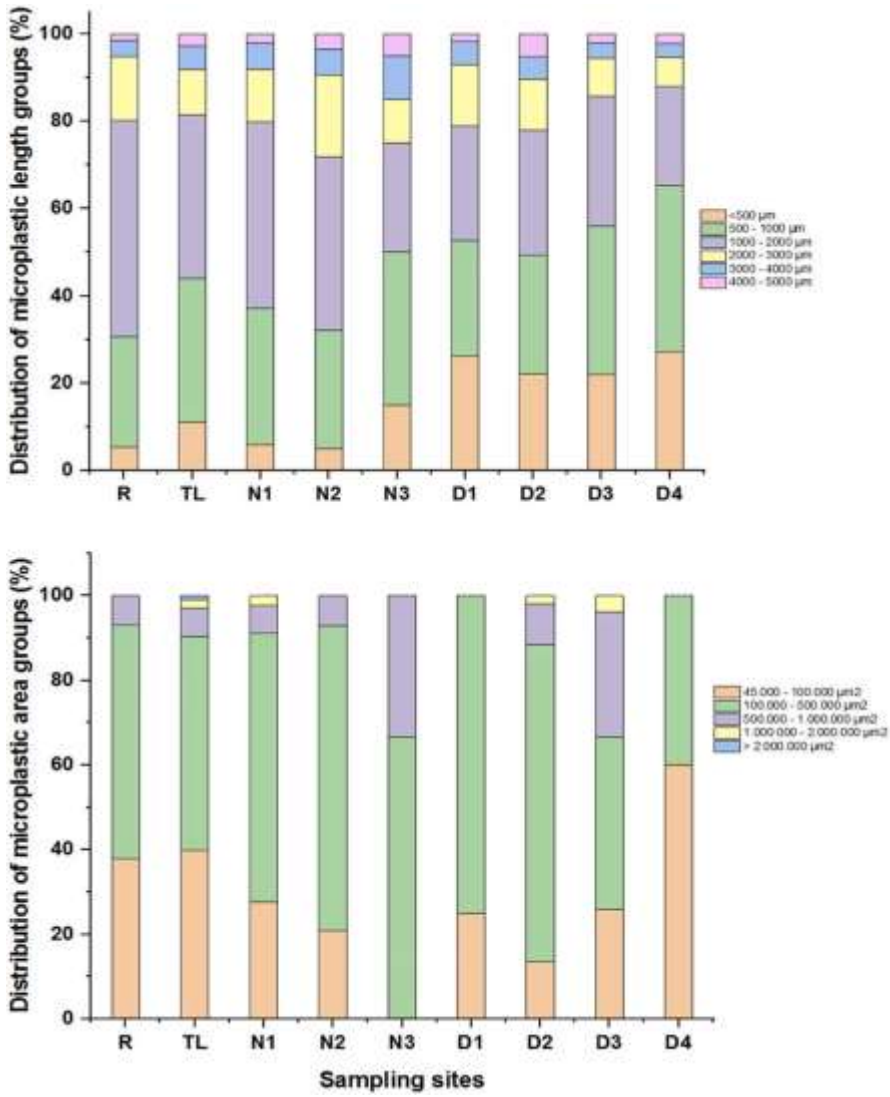

Fig. 7 Proportion distribution of fibre and fragment size range of MPs in sediment collected at the nine sampling sites

6). A variety of colours was observed, with purple and red being the predominant ones (Fig. 7). Based on the size measured under a stereomicroscope, the MP fibre size (1,000–2,000 µm) was predominant and accounted for 34% of the total MPs found. MPs with sizes of 300–500 µm and 500–1,000 µm contributed 16% and 31%, respectively. Larger sizes such as 3,000–4,000 µm and 4,000–5,000 µm were less common. Regarding the fragment form, MPs with a size range of 500,000–1,000,000 μm^2 dominated and accounted for 60% of the total identified MPs (Fig. 6). Raman spectrometry analysis (n = 44) identified the presence of PE (34%), PP (59%), PET (4%) and EVA (2%) (Figs. 8 and 9). Interestingly, several spectra showed a high match to pigments and dyes (Hostaperm Blue, Mortoperm Blue, Cobalt phthalocyanine), known as plastic components or additives used during manufacturing, and were later degraded as MPs in the field (Lots et al., 2017; van Cauwenberghe & Janssen, 2014).

MPs enter aquatic ecosystems mainly via untreated domestic and industrial wastewater release, recreational activities, agriculture and industry (Anderson et al., 2016; Wagner et al., 2014). In Hanoi, 80% of wastewater effluents are discharged without treatment (approximately 1,200,000 m^3 per day) into urban rivers (MONRE, 2019). Thus, wastewater or sewage from the city could be the major source of MPs in these rivers. Direct discharge of untreated wastewater from various sources (domestic waste, untreated sewage drain, washing clothes, solid commercial waste, etc.) could be responsible for the remarkably high MP concentrations measured in the sediments of the To-Lich and Nhue rivers (TL, N1, Fig. 4). In these two river surface waters, MP concentrations are critical; the MP concentrations were 109 ± 47 particles·m^{-3} and 1,589 ± 1,313 particles·m^{-3} in the Nhue and To-Lich rivers, respectively, with a predominance of fibres over fragments (Strady et al., 2021). The abundance of MPs measured in the sediments of the To-Lich River (up to 55,950 particles·kg^{-1} DW) was as high as those observed at the contamination hot spots in the Mersey/Irwell River catchment, UK (Hurley et al., 2018) and the Liangfeng River (6,950–149,350 particles·kg^{-1}). The decreasing concentrations observed in urban rivers that act as an open sewer until the more downstream sites (D4, DoThong, Day River) are raising several scientific questions that will need to be addressed in the future, such as the following. (i) What is the role of riverine water discharge in

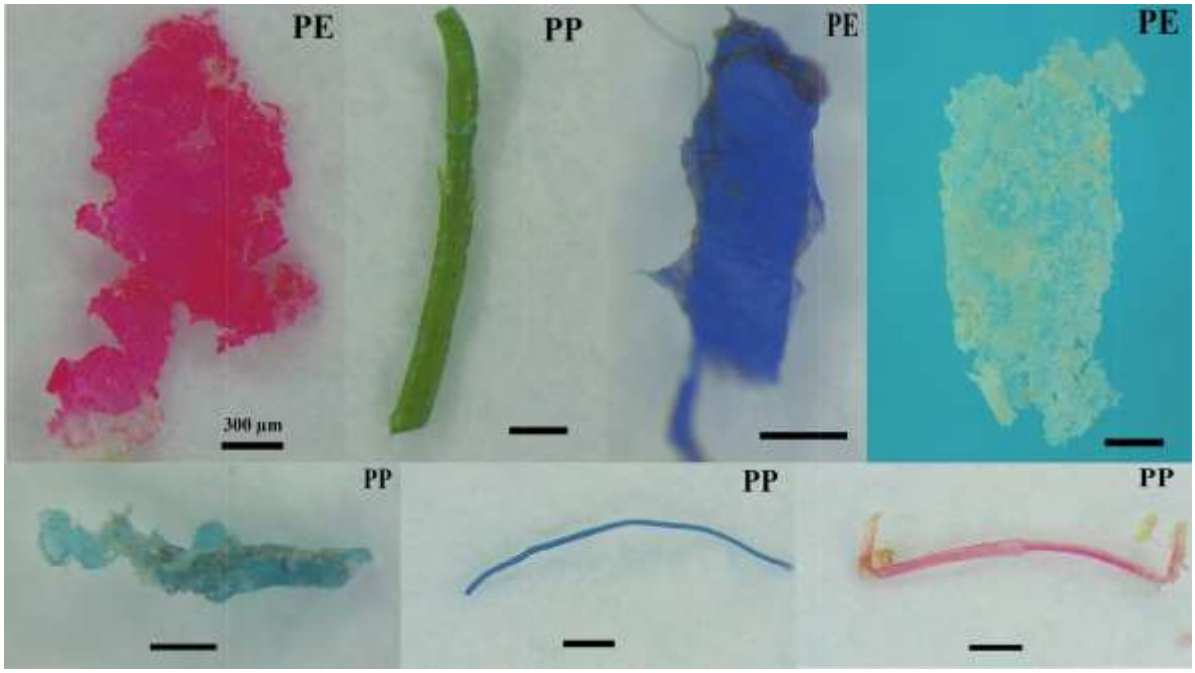

Fig. 8 Examples of observed MP forms in river sediment samples at nine sampling sites

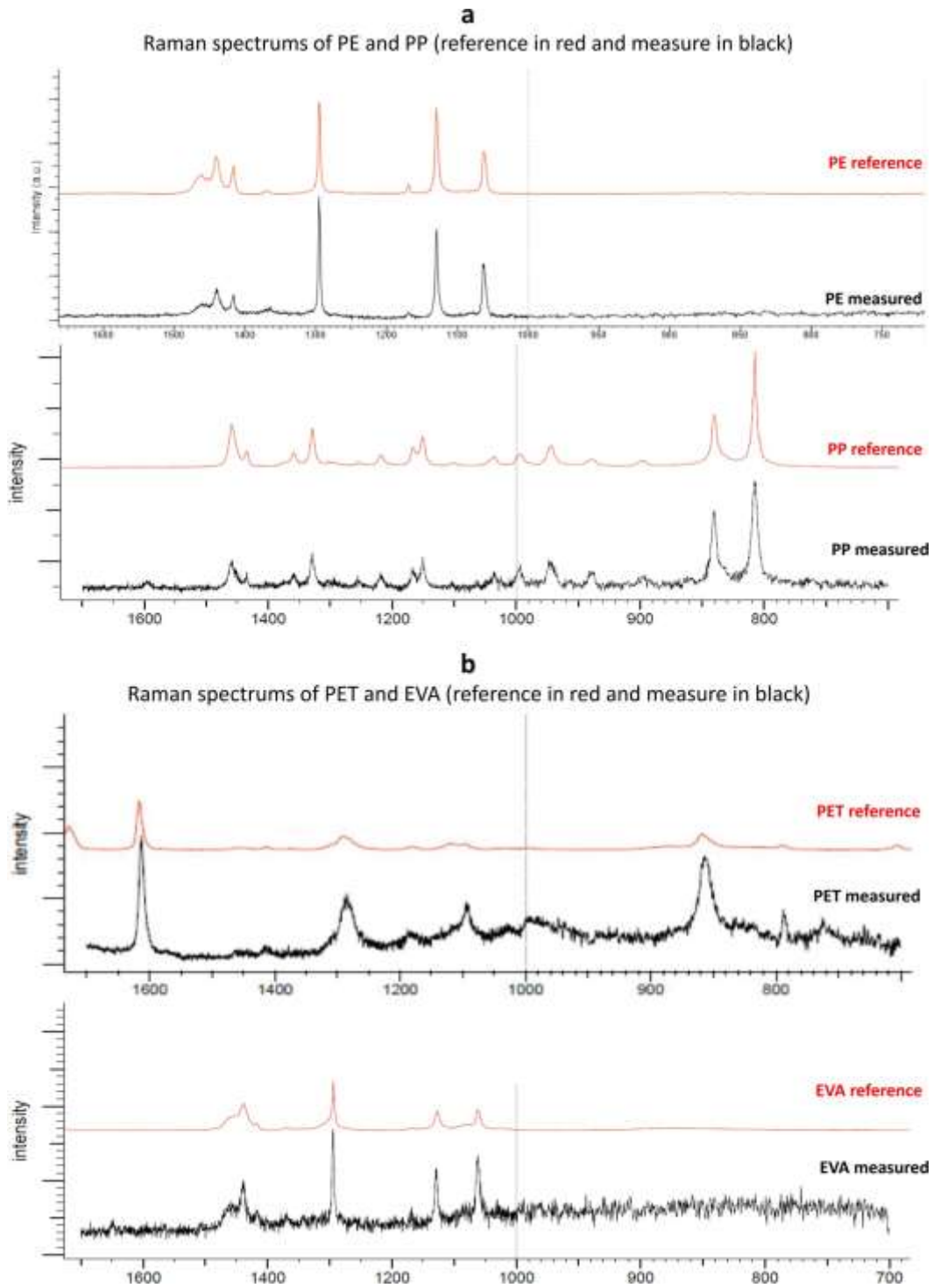

Fig. 9 **a** Raman spectrums of PE and PP (reference in red and measure in black). **b** Raman spectrums of PET and EVA (reference in red and measure in black)

MP sediment content? Are the concentrations higher in rivers with lower water discharges? (ii) Can the resuspension of the bottom sediment affect the transport of MPs into the water volume and then their settlement? (iii) What is the role of the estuarine mixing zone and biogeochemical processes involved in the fate and transport of MPs? (iv) Are sediments considered as a source or sink of MPs to the sea? These questions need to be clarified at the scientific community level but also in this environment, as the Red River Delta is flowing towards the Gulf of Tonkin in Vietnam, an important zone of fishing and aquaculture activities.

Conclusions

Globally, MP pollution is widespread and is becoming an important environmental concern. A robust and easy-to-use protocol is thus important for enhancing the measurement and understanding of MP pollution in water and sediments. In developing countries, criteria such as low cost, sourcing and environmentally friendly status are more important than the effectiveness of setting up a robust protocol. In this context, we demonstrated that NaCl solution for the density separation step was more appropriate than NaI, $ZnCl_2$ and $CaCl_2$. We then applied the protocol to riverine sediments from the Red River Delta and observed decreasing concentrations in the urban rivers receiving wastewater effluents to the estuarine mixing zone of the continuum. This tendency highlights the need to study the fate of MPs in estuaries, but also between sediments and the water column, to understand the inputs to the coastal zone and oceans.

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Data availability The main part of the research data is included in the article. Other data can be made available from the corresponding author upon request.

Declarations

Conflict of interest The authors declare no competing interests.

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