Microplastic abundance and characteristics in French Atlantic coastal sediments using a new extraction method☆

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ABSTRACT

The ubiquitous presence of microplastics (MPs) has been demonstrated in all environmental compartments in the recent years. They are detected in air, freshwater, soil, organisms and particularly in marine ecosystems. Since sediments are known to be the major sink of many organic and inorganic pollutants, the aim of this study was to develop and validate a fast and cheap methodology to assess the MP contamination in intertidal sediments from the Gulf of Biscay (Pays de la Loire region, France). Sediments were sampled at three locations (Pays de la Loire region, France) and during two seasons: October 2015 and March 2016. The analytical protocol involved MP extraction from dried sediments using milliQ water and a centrifugation technique. After a filtration step of supernatants, MPs were detected and directly identified on the membrane filters using μFTIR spectroscopy in reflection mode. For the first time, the number of replicates allowing to obtain a satisfying representativeness of the whole sampled sediment was also evaluated at 10 replicates of 25 g each. The average number of MPs in sediments was 67 ($\pm$76) MPs/kg dw (N $=$ 60) with no significant difference between sites and seasons. Ten different compositions of MPs were defined by μFT-IR with a high proportion of polypropylene (PP) and polyethylene (PE), 38 and 24%, respectively. Among MPs, mainly fragments (84%) were observed with main size classes corresponding to [>100 μm] and [50—100 μm] but no particles > 1 mm could be found suggesting that mainly small microplastics (<1 mm) were subject to vertical transport.

1. Introduction

Applications and societal benefits of plastics have augmented due to their advantages (Andrady and Neal, 2009). More than 300 million tons of plastics are produced every year and the production is increasing annually since 2013 (PlasticsEurope, 2016). Most of them are used for single-use (Hopewell et al., 2009) and an estimation of 10 percent ends up in the ocean (Barnes et al., 2009). The plastic waste is assumed to be a major source of microplastics (MPs, $\leq$ 5 mm in size) by fragmentation of plastic debris due to mechanical, chemical and biological factors (Costa et al., 2010; Andrady, 2011; Zettler et al., 2013). Indeed, damages in the marine environment due to the presence of MPs were reported (Andrady,

2011; Cole et al., 2011; Gall and Thompson, 2015). Hence the assessment of MP levels in marine environments (waters, sand/sediment and animals) has been the focus of scientists for the last decade (Mathalon and Hill, 2014; Desforges et al., 2014, 2015; Besseling et al., 2015; Cozar et al., 2015; Phuong et al., 2018a, 2018b).

Among different environmental compartments, numerous studies concentrated on the sediments (Van Cauwenberghe et al., 2015b) as they are known to be a major sink of contamination of marine ecosystems as dense MPs can sink directly. However, most studies demonstrated that floating MPs (e.g. polyethylene and polypropylene, with a density lower than water) were also found in sediments (Carson et al., 2011; Dekiff et al., 2014; Frere et al., 2017). The sedimentation of these MPs could be explained by the change of their density due to biofouling action (Zettler et al., 2013; Lagarde et al., 2016) and/or sorption of organic matter (Teuten et al., 2007; Bakir et al., 2012, 2014; Lee et al., 2014). Rocha-Santos and Duarte

(2015) highlighted the lack of standardized protocols to assess MP contamination in 2015. A current challenge in researching MP pollution is the lack of standardized protocols for sampling, extracting, identifying and characterizing MPs. This lack of standardization leads to a difficult comparison of results from several studies using different protocols. For example, the use of a digestion step or not, different sizes of sieves, different spectroscopy methods (Raman *vs* FT-IR), are all sources of result variations between studies. Table 1 shows the wide variety of protocols used in studies about MPs in sediments. Beyond the different analytical protocols, the location of studied areas seems to be a major factor influencing MP distribution in the field (Alomar et al., 2016; Ballent et al., 2016) and there are also many other reported influencing factors (Hanvey et al., 2017) such as the seasons, the sea current, the tide, etc. Matsuguma et al. (2017) found that the MP abundance in sediments depended on the sampling depth in Japan, Thailand, Malaysia and South Africa. Regarding Table 1, all protocols included digestion, extraction and identification steps. Hydrogen peroxide was used for digestion in 6 out of 28 studies. For MP extraction from sediments, most studies used dense solutions such as saturated NaCl, NaI, $CaCl_2$ or $ZnCl_2$ with or without centrifugation, whereas only 6 of 28 studies digested organic matter with hydrogen peroxide. Recently, several studies used the physical and chemical properties of MPs such as their hydrophobicity using Colza oil for isolation or their adsorption capacity using Nile Red to facilitate microscopic observation (Crichton et al., 2017; Maes et al., 2017a). Nevertheless, the

Table 1
Sources, procedures and references corresponding to a MP contamination assessment in marine sediments (classification done by identification techniques).

Sampling area	Digestion	Extraction		Identification	Procedure recovery (%)	References
		Reagents	Method			
Spain	No	H_2O	Shaking 15min	Observation	Nd[a]	Alomar et al., 2016
Italy	No	NaCl	—	Observation	Nd	Guerranti et al., 2017
Italy	No	NaCl	—	Observation	Nd	Cannas et al., 2017
USA	No	NaI	Stirring vigorously	Observation	Nd	Graham and Thompson, 2009
Belgium	No	NaI	Centrifugation (3500 g × 5 min)	Observation	98–100% of PVC[b]	Van Cauwenberghe et al., 2013
Germany	No	$CaCl_2$	Settle overnight	Observation	20–100% of PE spiked depend on color	Stolte et al., 2015
Adriatic Sea	No	NaCl	Shaking vigorously	Observation	Not reported	Blaskovic et al., 2017
North Sea	H_2O_2	$ZnCl_2$	—	Observation	Nd	Liebezeit and Dubaish, 2012
Canada	H_2O_2	NaCl	Stirring 2min	Observation	Nd	Mathalon and Hill, 2014
South Africa	No	NaCl saturate	Stirring vigorously	Observation	Nd	Nel and Froneman, 2015
Canada	No	Na_2WO_4	Shaking vigorously	Observation (verified with Raman)	Nd	Ballent et al., 2016
Baltic Sea	No	NaCl	Shaking 2min	Observation (verified with μFTIR)	Nd	Graca et al., 2017
Portugal	No	NaCl	Stirring vigorously	Observation (verified with μFTIR)	Nd	Martins and Sobral, 2011
North Sea	No	NaCl	Stirring vigorously	Observation (verified with FTIR)	Nd	Leslie et al., 2017
China	H_2O_2	NaCl	Stirring manually	Observation (verified with μFTIR)	Nd	Peng et al., 2017
Hong Kong	H_2O_2	NaCl	Shaking vigorously	Observation (verified with FTIR)	3.3% of PVC and 100% of PP	Tsang et al., 2017
Germany	No	NaCl-NaI	—	TD-PYR-GC/MS	Nd	Dekiff et al., 2014
Germany	H_2O_2	NaCl-NaI	—	TD-PYR-GC/MS	68–99% depend on MP type	Nuelle et al., 2014
Western Europe	No	NaI	Centrifugation (3500 g × 5 min)	Raman spectroscopy	98–100% of PVC[b]	Van Cauwenberghe et al., 2015a
Italy	No	$ZnCl_2$	—	Raman spectroscopy	Nd	Imhof et al., 2013
France	No	$NaCl-Na_2WO_4$	—	Raman spectroscopy	Nd	Frere et al., 2017
USA	No	NaCl	Stirring	FTIR spectroscopy	Nd	Carson et al., 2011
Canada	No	Canola oil	Settle 2min	FTIR spectroscopy	92–99% depend on MP form	Crichton et al., 2017
North Sea	No	Nile red, $ZnCl_2$	Centrifugation (100 g × 60 min)	FTIR – Fluorescence	85–98% depend on sediment sample	Maes et al., 2017a
England	No	NaCl saturate	Stirring 30s	FTIR spectroscopy	Nd	Blumenroder et al., 2017
England	No	$ZnCl_2$	Stirring 3min	FTIR spectroscopy	92–98% depend on MP type	Coppock et al., 2017
Singapore	No	NaCl, Tween-80	Centrifugation (200 cycles/2min)	FTIR spectroscopy (ATR)	55–72%	Nor and Obbard, 2014
Eastern Asia South Africa	H_2O_2	NaI	Centrifugation (2000 × 10min)	FTIR spectroscopy (ATR)	93%	Matsuguma et al., 2017
England	No	NaCl saturate	Stirring 30s	FTIR spectroscopy (ATR)	Nd	Browne et al., 2010
Arctic	No	$ZnCl_2$	Stirring 35–60min	μFTIR spectroscopy (ATR)	Nd	Bergmann et al., 2017
England	No	NaCl saturate	Stirring 30s	FTIR spectroscopy (transmission)	Nd	Thompson et al., 2004
India	No	NaCl saturate	Stirring 1–2 h	FTIR spectroscopy (transmission)	Nd	Reddy et al., 2006
Atlantic Ocean	No	NaCl	Stirring 30s	FTIR spectroscopy (transmission)	Nd	Woodall et al., 2014
Singapore	No	NaCl saturate	Centrifugation (200 cycles/1min)	FTIR spectroscopy (reflection)	Not reported	Ng and Obbard, 2006
Belgium	No	NaCl saturate	Stirring 2min	FTIR spectroscopy (reflection)	69–98% depend on MP type	Claessens et al., 2011
Italy	No	NaCl	Stirring 1.5min	μFTIR spectroscopy (reflection)	Nd	Vianello et al., 2013

[a] Nd (not determined).
[b] Results adapted from Claessens et al. (2013).

colonization of micro-organisms or the sorption of amphiphilic/hydrophilic compounds on the surface of MPs could lead to a more limited performance of these methods. As reported in Table 1, in many studies MPs identification was only performed using microscopic observation without any spectroscopic method. It was demonstrated as not totally sufficient to assess the environmental MP contamination (Hidalgo-Ruz et al., 2012). Some other studies extrapolated the number of MPs by spectroscopically analyzing only a part of the particles which were previously observed by microscopy, but this method seemed to be not precise enough since the determined number could be very different from the real number of MPs in the sample. Each technique displayed different advantages and disadvantages mainly according to the limited size of analyzed MPs and time consumption (Kappler et al., 2016). The analytical procedure developed in the present work presents many advantages compared to the others, i.e. affordable, simple and environmentally friendly. Besides, as the MP distribution in sediment samples is assumed to be not homogeneous, the number of sample replicates is also a parameter to examine to ensure the representativeness of the data. The aim of this study was to develop and validate a fast and cheap methodology to assess the MP contamination in intertidal sediments from the Bay of Biscay coast (Pays de la Loire region, France). This littoral region gathers significant areas of aquaculture at the national level, for which the characterization of MP contamination is of great concern. For example, the production of mussels and oysters reaches 16 thousand tons annually for national consumption. For the first time the representativeness of the data obtained according to the number of analyzed replicates was investigated. Finally, a comparison of our results with previous data obtained along the French North-East Atlantic coast in seawater, sediments and marine organisms was considered, to highlight relationships between the contamination in both environmental compartments, i.e. physical and biotic.

2. Materials and methods

2.1. Studied sites and sediment sampling

In order to assess the MP contamination in the shellfish habitats, sediments were sampled in the production zones. Three locations were selected in the Pays de la Loire region: Pen-Bé (N 47°25′33″ W 2°27′46″), Coupelasse (N 47°01′31″ W 2°01′99″) and Aiguillon Bay (N 46°16′26″ W 1°14′14″) (Fig. 1). They are three important spots of shellfish production at the regional and national scales leading to important socio-economic concerns. The strategy and the description of sampling areas are detailed in a previous study (Phuong et al., 2018b). For each sampling site, sediments were collected on intertidal mudflats close to farming areas, at two different seasons: October 2015 (beginning of autumn) and March 2016 (beginning of spring). In autumn at Aiguillon Bay, the surface sediments (20 cm of depth) were sampled using a box-corer from a boat because of high tide. For all the other samplings, the surface sediments (0–10 cm of depth) were collected with a spatula at low tide on three 50 cm length squares randomly selected but distanced by at least 20 m. Then, all sub-samples, representing 3 kg of sediments, were pooled in glass-boxes before being conducted to the laboratory in a refrigerated enclosure. Nitrile gloves were used for all the sampling duration. In the laboratory, the sediments were kept in a freezer at −20 °C until analysis.

2.2. Practices for reducing contamination risks

All experiments were carefully performed with the aim of preventing MP contamination. Materials were previously rinsed three times with MilliQ water (PUBLAB, Option R-7/15) before their use. Laboratory coats in cotton and nitrile gloves were worn all the time. Sample handling was performed in a clean hood.

2.3. Sample treatment

The sediments were taken out of the freezer and thawed just before their preparation for analysis. Sub-samples of 25 g wet sediments were placed in a glass-beaker under aluminum foil and dried during 24 h, at 80 °C, in an oven. Then, they were sieved with 1 mm stainless steel metal.

2.4. MP extraction

The extraction protocol was performed on dried and sieved sediments (fraction < 1 mm). The remaining matter on the 1 mm stainless steel metal sieve was also analyzed to evaluate the presence of MPs in this fraction.

Sediment matters were introduced into centrifuge tubes of 50 mL made of PTFE (polytetrafluoroethylene) plastic (Nalgene tube − Thermo Fisher Scientific).

A preliminary set-up including a single-step of digestion before centrifugation was considered using KOH (10%) or HNO_3 (65%). These reagents were added in ratio 2:1 v/m (20 mL of reagent for 10 g of dried sediment). The results obtained during these tests were not concluding and the digestion step was abandoned.

Then, a simple extraction method using either 20 mL of demineralized water or 20 mL of 50% KI was tested. The hypothesis was that with 50% KI, the recoveries of MPs made of polymer denser than water would be better than with water. After a careful stirring using a stainless-steel spoon, the samples were centrifuged. Different combinations of temperature (15 °C and 18 °C), duration (2, 5 and 10 min) and speed of centrifugation (200, 500 and 1000 cycles/min) were tested. The surface of supernatants was then collected using Pasteur pipettes and filtered on cellulose nitrate membranes with pore diameters of 12 μm and a size of 25 or 47 mm. Eventually, filters were dried at room temperature in glass Petri dishes remained closed until analysis.

2.5. MP identification

The identification and characterization of MP was directly performed on the filters by using a Fourier transform infrared microscopy system (μFT-IR; Spotlight 200i FT-IR microscopy system, PerkinElmer) in reflection mode. The size, the color and the form of MPs were determined. The color and size were recorded according to Galgani et al. (2013). The MPs were then categorized in 3 ranges of size: 20–50 μm; 50–100 μm and >100 μm. The chemical identification was only possible for particle size >20 μm due to the limited focalization on measurement point using μFT-IR in reflection mode. The fibers were not systematically excluded from the analysis but they were not counted as MPs when they were not identified because of a width smaller than 20 μm. For that reason, they could be underrepresented. About form, MPs were classified as fragments or fibers because no other forms were observed. The whole surface of each filter was inspected and for each observed particle, a measurement was performed with 8 accumulations ranging from 4000 to 600 cm^{-1}. The microscope had a magnification of about 300×. For the first measure, an aperture of 60×60 μm was used and then changed depending on the size of interesting particles. The spectrum of the particle was then compared to the polymer database (PerkinElmer library about 8000 reference spectrums) and the type of polymer was determined when the research score was higher than 60%.
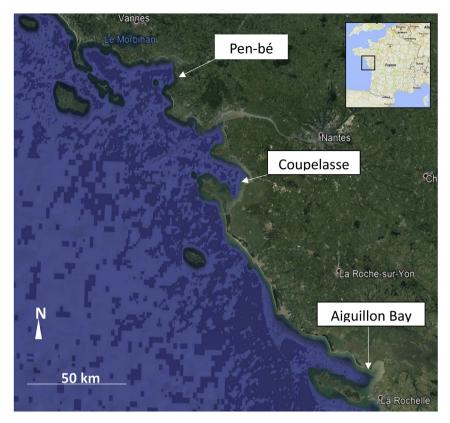
Fig. 1. Sampling locations on French Atlantic Coast (Google Earth picture).

2.6. Protocol validation

The protocol of analysis was validated by spiking sediment samples with MPs. The MPs were employed as references and had to represent the different possible densities of MPs commonly found in environmental samples. MPs with densities lower than that of water such as PE ($d = 0.91–0.94\,g\,cm^{-3}$) and PP ($d = 0.83–0.85\,g\,cm^{-3}$), as well as those with densities higher than water such as polyvinyl chloride (PVC; $d = 1.38\,g\,cm^{-3}$) and polyethylene terephthalate (PET; $d = 1.37\,g\,cm^{-3}$) were used in the protocol validation. These MPs were generated from the cryomilling of commercial polymers in a laboratory. MPs of PE, PP, PVC and PET were respectively made from a cable, a bag, a pipe and a water bottle. All of these MPs ranged from 50 to 400 μm after a separation step using metallic sieves. Ten fragments of each polymer were used for spiking the same sediment sample (25 g wet weight) in triplicates (n = 3), leading to 40 particles in each sample. By visual observation of the sediments and MPs in the tubes during the validation if the protocol, the better conditions of centrifugation were determined as 18 °C, during 5 min at 500 cycles/min (Centrifuge, Jouan MR23i). After analysis in these conditions, the recoveries were calculated for each polymer. Negative blanks were also performed concurrently in ten replicates, following the same protocol by using 25 mL of demineralized water instead of 25 g of sediments. These experiments allowed the evaluation of the cross-contamination due to airborne, manipulation, etc. Once validated, the protocol was applied to the sediment samples (25 g wet weight), in ten replicates per site and season. Then, the results were expressed as the number of MPs (average ± standard deviation) per kg of sediment dry weight (dw).

2.7. Representativeness of the sample

The aim was to determine the number of replicates needed to give results representative of the whole sediment sample. For this test, the sediment taken from one site and representing one season was randomly selected. The validated protocol of MP analysis was applied to 20 sub-samples of 25 g each, called replicates, and the MP abundance was measured for each replicate. The average number of MPs in the sediment was calculated according to the number of replicates, randomly added one by one. The minimum of replicates leading to a good representativeness of the whole sediment was graphically determined.

2.8. Statistical analysis

The data were analyzed using the XLSTAT software. Non-parametric Kruskal-Wallis (KW) tests were used in order to highlight significant differences of MP contents in sediments collected at different sites and seasons. Differences between sediment types were relevant when $p < 0.05$. The KW test was followed by a post hoc test Multiple Comparisons of p-value (MCP).

3. Results and discussion

3.1. Protocol set-up and validation

For the protocol set-up, a digestion step before the extraction was first considered using KOH, which is known as a good reagent for the digestion of organic matter (Dehaut et al., 2016; Phuong et al., 2018a). However, these tests were not conclusive because

precipitation appeared in the solution even after centrifugation. The substitution of KOH by HNO$_3$ was tested for this digestion step. Recoveries from 66 to 100% were found for PE and PP but in the best case, only 3.3% of PVC MPs were detected in spiked samples, respectively with and without sediments. HNO$_3$ probably reacted with the surface of PVC MPs leading to changes of their surface properties. Moreover, this acid was reported as damaging nylon (Claessens et al., 2013) and discoloring PE (Phuong et al., 2018a). Eventually, no step of digestion was considered as the marine sediments which were analyzed contained little organic debris. Thus, the following tests were only based on flotation separation technique. Tests using 50% KI for the centrifugation step were performed, but a lot of matter in suspension and at the surface of the solution was observed after the centrifugation. As a consequence, larger filters (47 mm) had to be used leading to a longer duration of μFT-IR analysis, without any improvement of the spiked MPs recovery. Eventually, the optimized procedure of MP extraction involved 20 mL of demineralized water added to 25 g of 1 mm-sieved sediments and a centrifugation step followed by a filtration step on a 12 μm pore-size filter of 25 mm. This procedure could be characterized as cheap, "green" and rapid which is valuable for the assessment of the environmental contamination.

The results obtained after the analysis of sediments spiked with MPs and extracted according to the optimized procedure showed good recoveries for the 4 MPs tested (PE, PP, PVC and PET), whatever their relative density to the water. The recoveries with sediments (107 ± 6, 83 ± 6, 93 ± 6 and $83 \pm 6\%$ respectively for PE, PP, PVC and PET; n = 3) were not significantly different to recoveries without sediments (respectively 97 ± 6, 87 ± 15, 87 ± 6 and $93 \pm 6\%$ for PE, PP, PVC and PET; n = 3). The flotation of plastics is not completely based on the density and this process was shown to be influenced by other factors such as the size, the shape, the surface chemicals (Shen et al., 2001, 2002; Wang et al., 2014). The flotation of high density MPs was already observed and discussed for MPs made of PVC (1.14–1.38 g cm^{-3}), polyurethane (PU, 1.20–1.26 g cm^{-3}), polyamide (PA, 1.12–1.15 g cm^{-3}) in the seawater of the Atlantic Ocean (Enders et al., 2015), or PET (1.38–1.41 g cm^{-3}) in the sea surface microlayer of the Korean Coast (Song et al., 2014).

Regarding identification, three out of the four MP types (PE, PP and PVC) provided satisfactory identification scores through FT-IR analysis, about 80% for PE and PP and 65% for PVC. For MPs made of PET, the first given identification comparing spectra to the library was "polyester" with a research score around 94%. The identification as PET was only reaching 65%. Nevertheless, it is important to consider that polyester corresponds to a large group of polymers which include PET. The low evidence for PET identification may be due to the possible presence of additives in the PET MPs.

Globally, for spiked or raw sediment samples, only about 10% of the particles visualized on filters were confirmed as made from plastic by μFT-IR, as highlighted in previous studies (Hidalgo-Ruz et al., 2012; Phuong et al., 2018a). To complete the validation, 10 negative blanks were performed by using 25 mL of demineralized water instead of 25 g of sediments. After drying, followed by centrifugation and filtration steps, the microscopy allowed the quantification of an average of 1 (± 1) item per filter. Three out of the fourteen items detected in these blanks were small filaments with a thickness inferior to 15 μm. They were consequently impossible to identify because the chemical identification was only possible for a particle size >20 μm due to the limited focalization on a measurement point using μFT-IR in reflection mode. When considering particles >20 μm, the identification of items concluded to no particles made from plastic. This experiment allowed to ensure no cross-contamination by MPs considering a size superior

to 20 μm. The difficulty to identify filaments, since they have a small size, was also reported in the work of Wesch et al. (2017).

3.2. Representativeness of the sample

After the analysis of 20 sub-samples of 25 g of the same wet sediment (replicate), the results were expressed for each replicate as the number of MPs per kg of dry sediments. Then, the average number of MPs was calculated after taking into account the addition of another replicate result randomly selected. Fig. 2 represents the average number of MPs per kg of dry sediments related to the number of replicates considered for the calculation of the average.

Although the sediments were mixed before analysis, Fig. 2 demonstrates that the number of MPs in replicates was highly variable. When the number of replicates was low, i.e. 2 to 9, the average number of MPs was rising from 0 to 80 MP/kg of dry sediments. Then, when the number of replicates was between 10 and 20, the average number of MPs reached a plateau with values ranging from 75 to 100 MP/kg of dry sediments. Fig. 2 highlights that it was essential to analyze a minimum of 10 replicates, in the case of this study, to ensure the representativeness of the whole sediment sample. In other studies, the number of replicates was lower than 10 (Blaskovic et al., 2017; Carson et al., 2011; Claessens et al., 2011) but the mass of sediments analyzed was also higher (up to 1 kg). In fact, if the distribution of MPs in sediments was homogenous, the representativeness would be ensured with a lower number of replicates. According to Fig. 2, ten replicates of 25 g of wet sediments were analyzed for each site and season.

3.3. MPs in the sediments

3.3.1. Quantitative results

No plastic item was observed in the >1 mm sediment fractions from each site and season. This observation highlighted that the accumulation of large MPs (>1 mm) in superficial sediments of intertidal mudflats was limited. It seemed to be consistent with the model describing the MP distribution in seawater (Enders et al., 2015). These authors demonstrated that buoyant polymers like PE and PP of sizes $\geq$ 1 mm were floating on the surface in a similar manner as it is expected for the macroplastic debris. The small MPs (10 and 100 μm) are expected to be found deeper in the water column (average of 24 m for 100 μm MP, and 33 m for 10 μm MP) and more likely in sediment. The residence time of large MPs in the surface ocean is then considered to be longer than for small MPs and a transport of these particles to distant areas than coastal mudflats could be expected.

In the sieved sediment samples (<1 mm), the average concentration for all the considered samples was 67 (± 76) MPs per kg of dry sediments (N = 60). Moreover, all sediment samples contained MPs. This result corroborates the ubiquity of MPs in the marine environment (Browne et al., 2011; Eriksen et al., 2014).

Fig. 3 shows the results of MP abundance in sediments according to the site and the season. The data obtained in autumn at Aiguillon Bay must be compared carefully to those for other sites and season as the sampling was done differently (using a box-corer during high tide). The medians ranged from 28 to 88 MPs per kg of dry sediments. Fig. 3 shows a great variability of the results as the MP number in sediments is a discontinuous variable as explained above. Thus, no significant difference was highlighted between sediments from the different sampling sites collected at both seasons (p value < 0.05). The anthropogenic pressures of the different sites, the maximum distance of 140 km between them and their oceanic and terrigenous influences did not lead to different MP contaminations as already shown in bivalves from the same sampling sites (Phuong et al., 2018b). This observation could traduce a
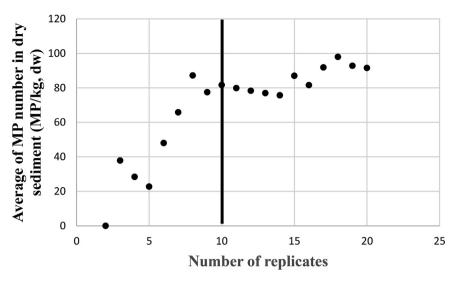
Fig. 2. Average number of MPs found in sediments related to the number of replicates (dry sediment sub-sample of 25 g each). The vertical line corresponds to the limit number of replicates needed for a good representativeness of the sample.
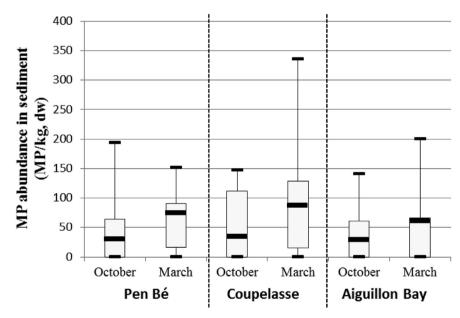

Fig. 3. MP abundance in sediments related to the sampling site and season expressed as number of particle/kg of dry sediments. N = 10 per season and per site. Box-plots depicted minimum, first quartile, median, third quartile and maximum values.

very diffuse distribution of MPs in the environment at the scale of this coastal zone.

In order to compare the results of the present study to those reported in the literature, Table 2 reports MP abundance found in sediments from different sampling locations around the world. When the results were not expressed as the number of particles per kg of dried sediments, the calculations were done to transform the unit using an average sediment density of 1600 kg m^{-3} (Fettweis et al., 2007) and an average wet sediment/dry sediment ratio of 1.25. The present results appeared to be of the same order of magnitude than those depicted in numerous previous studies (Thompson et al., 2004; Coppock et al., 2017; Peng et al., 2017). However, huge differences were also observed compared to other ones, with present values higher in some cases (Dekiff et al., 2014;

Stolte et al., 2015) or lower (Nel and Froneman, 2015; Matsuguma et al., 2017). As it was already discussed in the previous article about MP contamination of bivalves from this area (Phuong et al., 2018b), the similarities and differences between results could be due to spatial variations of the MP distribution depending on different factors, *i.e.* anthropogenic pressures or water currents. But the analytical procedures of MP analysis could also be an explanation of the variations leading, theoretically, to an impossible comparison of results. As an example, the lowest size of the MPs analyzed with the different analytical procedures is not systematically mentioned in works presented in Table 2 while it is an important criteria to take into account for comparisons of quantitative results of MPs.

Table 2
MPs with a size inferior to 5 mm per kg of dried sediments from different sampling locations around the world.

Continent	Sampling area	MPs		References
		Quantity in sediments (number of particles/kg dw)	Characteristics	
America	USA	79–165	Fragments	Graham and Thompson, 2009.[a]
	Canada	83–161.8	Fibers (77%)	Crichton et al., 2017
	Canada	760	PE, PS	Ballent et al., 2016
	Canada	2000–8000	Fibers	Mathalon and Hill, 2014
Africa	South Africa	400–1750	PE, copolymer	Matsuguma et al., 2017
	South Africa	161–759	Blue/black fibers	Nel and Froneman, 2015 [a]
Antarctica	Arctic Ocean	42–6595	<25 μm (80%)	Bergmann et al., 2017
Asia	China	121 ± 9	Fibers	Peng et al., 2017
	Hong Kong	47–279	PE, PP	Tsang et al., 2017
	Singapore	36.8 ± 23.6	PE, PP, PVC	Nor and Obbard, 2014
	Eastern Asia	100–1900	PE, PP (fragments)	Matsuguma et al., 2017
	Singapore	0–16	PE, PS	Ng and Obbard, 2006
Europe	Baltic Sea	25–53	Polyester, fibers	Graca et al., 2017
	North Sea	100–3600	Spheres	Leslie et al., 2017
	Germany	1.3–2.3	PE, PP	Dekiff et al., 2014
	Western Europe	6.0 ± 5.7	PE, PS	Van Cauwenberghe et al., 2015a, 2015b
	France	0.97 ± 2.08	PE, PP	Frere et al., 2017
	Belgium	7.2–20.4	Fibers, granules	Van Cauwenberghe et al., 2013
	Germany	0–7	Fibers	Stolte et al., 2015
	England	3030	Fibers, blue, PTFE	Blumenroder et al., 2017
	England	67.4 ± 13.2	PE and PE copolymer	Coppock et al., 2017
	North Sea	210–461	Granule, fibers	Liebezeit and Dubaish, 2012
	England	322	PVC, polyester	Browne et al., 2010.[a]
	England	86	Fibers, 9 natures	Thompson et al., 2004
	Atlantic Ocean	200	Polyester (fibers)	Woodall et al., 2014 [a]
	Belgium	97.2 ± 18.6	Fibers	Claessens et al., 2011
	Italy	672 ± 2175	PE, PP	Vianello et al., 2013
	Spain	900 ± 100	Black, blue	Alomar et al., 2016 [a]
	Italy	45–1069	Filament	Guerranti et al., 2017
	Italy	62–1069	Black, blue	Cannas et al., 2017
	France	38–102	PP, PE (fragment)	This study

PE: Polyethylene; PP: Polypropylene; PVC: Polyvinyl chloride; PTFE: Polytetrafluoro ethylene and PS: Polystyrene.
[a] Quantities in sediments were recalculated with an average sediment density of 1600 kg m^{-3} (Fettweis et al., 2007) and an average wet sediment/dry sediment ratio of 1.25.

3.3.2. MP characteristics

• General characteristics

MPs found in all sediments whatever the site and the season were mainly fragments (84%) and some were filaments (16%), but neither granule nor pellet. They were made of 10 different polymers (PP, PE, polystyrene (PS), PVC, acrylonitrile butadiene styrene (ABS), polyvinylpyrrolidone (PVP), polyacrylonitrile (PAN), polyamide (PA), Polyester, Copolymer of PE and PP), with a majority of PP, PE with respective proportions of 38% and 24%. Five polymers (PP, PE, PS, PVC, and polyester) represented more than 90% of MPs (respectively 38, 24, 9, 9 and 7%). The compositions of the MPs found in the sediments seemed to be a good reflection of both worldwide and European production of plastic (PlasticsEurope, 2016). The two predominant polymers (PP and PE) are usually used in cars, toys, housewares and food packaging with a high demand at the European level which corresponds to 19.3 and 29.8% for PP and PE polymer types (PlasticsEurope, 2017). Besides, both of them have a short usage lifetime (Hopewell et al., 2009) contributing to their presence in the environment and notably in marine compartments. PP and PE have a lower density than water and thus they should be buoyant. Their detection in sediments is the consequence of sedimentation processes as described by Enders et al. (2015) for small buoyant MPs, as discussed hereafter. Besides, the colonization of MPs by micro-organisms/algae (Zettler et al., 2013), the sorption of organic matter (Teuten et al., 2007), the aggregation of MPs (Lagarde et al., 2016) or their integration in marine snow (Long et al., 2015) could lead to an increase of their density and their surface hydrophobicity. The phenomenon of sedimentation probably also occurs for polystyrene (PS) which

presents a broad range of density (0.16–1.05 g cm^{-3}; Engler, 2012). This polymer is mainly used for drinking cups, packing materials, and electronics because of its insulation properties. Concerning PVC, its presence in sediments could be expected due to its higher density compared to water. PVC is used in pipes (40%), cables, and food packings. Considering polyester, it represents a group of polymers, including the polyacrylate, the polyglycolide and especially the PET, mainly used as textile yarn. Thereby, according to Song et al. (2014), 75% of polyester MPs found in the sediments were filaments probably coming from textiles. The rest of the polyester MPs (25%) was made of fragments maybe coming from the fragmentation of drink bottles or industrial paints. Other MPs found in a small proportion in the sediments were copolymers including PP or PE, ABS, PA, PAN and PVP.

The MP size distribution is presented in figure S1 (supplementary material). The smallest particle found was 40 μm in length and the longest was a fiber of 2000 μm (n = 55). The majority, *i.e.,* 44%, of particles are between 100 and 250 μm. To allow comparison with previous results obtained in bivalves (Phuong et al., 2018b), only three size ranges [>100 μm], [50–100 μm] and [<20–50 μm] were considered in the following work. According to these three ranges, the MP abundances reached 47%, 45% and 7%, respectively.

Regarding colors, the predominant ones were grey (60%) and white (13%). This result could be in line with the hypothesis of a long time spent in the environment. A total of 8 colors (grey, red, white, green, black, blue, pink and yellow) were observed.

• Spatial and temporal variations of MP contamination in sediments

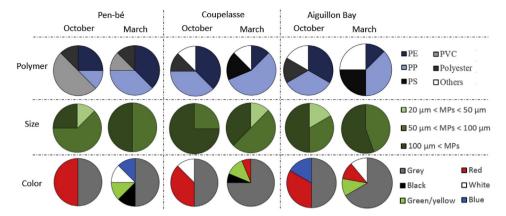Characteristics of MPs (type of polymer, size and color) found in

Fig. 4. Distribution of MPs according to the polymer, the size and the color, identified in sediments from different sites of the French Atlantic Coast and at two seasons (N = 10 per site and per season).

the sediments from the different sampling sites collected at both seasons are given in Fig. 4.

Although MP contamination varied according to site and season, some trends in polymer type emerge. PP and PE were present in all sediment samples. However, PVC was only found at Pen-Be, along with PE, PP and polyester. Whereas PS was only found at Coupelasse and Aiguillon Bay in March, and no polyester. No trends in size classes were observed and few MPs smaller than 50 μm were found.

3.4. Comparison of MPs found in different environmental compartments from the French North-East Atlantic coast

The MP contamination of the French North-East Atlantic Coast was assessed in a few publications on water, sediment or biota compartments as shown in Table S2 (supplementary material).

Results of MP contamination are really different depending on the study, *i.e.*, the studied site or the performed analytical procedure. For seawater, the variations could be explained by differences in sampling locations and methods. Frere et al. (2017) sampled in the bay of Brest using a standard Malta trawl with a 335 μm mesh net while Van Cauwenberghe et al. (2015a) sampled in the North Sea using a bucket. As a result, the MP size found in these studies was very different with MPs ranging from 30 to 300 μm in Van Cauwenberghe et al. (2015a) study and from 335 to 5000 μm in Frere et al. (2017).

About sediments, the variability of the results is great too, with levels from 0.97 to 481 items/kg of dried sediments. The results of this work were higher than those reported by Frere et al. (2017) and Van Cauwenberghe et al. (2015a) for the English Channel and the North Sea respectively, but lower than those of Lots et al. (2017) and Maes et al. (2017b) for the English Channel. It may be a consequence of spatial variations between study spots but also the result of different identification techniques (observation in Maes et al., 2017b; Raman spectroscopy in Frere et al., 2017, Lots et al., 2017, Van Cauwenberghe et al., 2015a and μFT-IR in this study). The predominant shape found in the present study was a fragment while it was a sphere in the study of Maes et al. (2017b) showing a probable difference of contamination source. Although the sediments sampled in both studies were superficial, other factors could influence the results obtained such as the sampling date, the exposure to ocean currents (Alomar et al., 2016) and the side distance (Vianello et al., 2013; Graca et al., 2017).

A few studies were performed on bivalves from the French North-East Atlantic Coast (Vandermeersch et al., 2015; Van Cauwenberghe et al., 2015a; Phuong et al., 2018b). A higher homogeneity of MP levels could be observed for bivalves, compared to

sediments. However, some differences could be highlighted in the methods used for sample treatment and MP identification (Phuong et al., 2018a). This last study concerns bivalves collected at the same sampling sites and at the same dates. The MPs found were identified with the same method than those used for the sediments analyzed in the present study. As oysters and mussels are two filter-feeding organisms, their MP content should be the result of the filtration of MPs suspended or floated in the seawater column. However, it also seems interesting to compare the MP contents in bivalves with the results obtained in sediments from the same area, as the MPs were likely to be present in the seawater column before sedimentation. This sedimentation process was observed to occur naturally for all MP particles with a size under 100 μm (Enders et al., 2015), and also transformed by MP colonization with microorganisms or by adsorption of organic matter (Teuten et al., 2007).

In Phuong et al. (2018b), the whole abundance of MPs reached 0.23 ± 0.20 and 0.18 ± 0.16 MPs/g of wet weight of soft tissues, in mussels and oysters, respectively. By considering a water level of about 70% in soft tissues of mussels and oysters, the contamination could be evaluated at 0.77 ± 0.67 and 0.60 ± 0.53 MPs/g dry weight of mussels and oysters, respectively. In the present study, the MP content in sediments was evaluated at 0.067 ± 0.076 MPs/g dw. The MP content in bivalves from the same location and season was ten times higher than those with sediments showing an accumulation of MPs by filter-feeding species. Karlsson et al. (2017) also found an accumulation of MPs by mussels in a larger proportion as MP content in mussels was approximately a thousand fold higher than those in sediments and seawater from the North Sea coast. About the MP particle size distribution, the proportion of 20—50 μm MP found in sediments was twice and five times lower than in oysters and mussels, respectively. The proportion of MPs ranging from 50 to 100 μm was relatively similar *i.e.*, 53, 52 and 45% for oysters, mussels and sediments, respectively. Thus, the lower proportion of small MPs (20—50 μm) in sediments was balanced with a higher proportion of MPs with a size equal or superior to 100 μm, compared to bivalves. These results could suggest a potential selective filtration by the bivalves according to the MP size and in favor of particles with sizes ranging from 20 to 50 μm. Regarding the quality of MPs, PE and PP are highly predominant in both matrices (sediments and bivalves) sampled on the same locations. These results seem to be in agreement with previous studies such as Karlsson et al. (2017) who also found PP and PE as predominant polymers in sediments and mussels. Furthermore Frere et al. (2017) and Van Cauwenberghe et al. (2015a) also observed that PVC polymer was only found in sediment samples.

3.5. Conclusions

A cheap, green and fast analytical procedure for MP extraction and identification from sediments was optimized. This procedure was validated by spiking experiments with 4 different polymers (PP, PE, PVC and PET) representing a large range of densities and with a size ranging from 50 to 400 μm. The protocol corresponded to 4 successive steps: drying of sediments, centrifugation with milliQ water, filtration through nitrate cellulose (12 μm) and direct observation/identification using μFT-IR spectroscopy. The optimum number of sediment replicates to achieve the representativeness of the sample was also determined to be 10 replicates of 25 g each. Quantitative and qualitative results about MP contamination were provided for sediments from the French Atlantic coast at 3 sites and 2 seasons. The average number of MPs in sediments was 67 (±76) MPs/kg dw (N = 60) with no significant differences between sites and seasons. Among MPs, mainly fragments (84%) were observed, filaments represented 16%. MPs were made of 10 different polymers, with a majority of PP, PE. The main size classes of MPs were [>100 μm] and [50—100 μm] with the predominant colors of grey (60%). These observations highlighted the limit of the accumulation of large MPs (>1 mm) in intertidal mudflats. MP contents in seawater, sediments and bivalves from the French Atlantic coast sampled at the same location were discussed showing a potential selective filtration of small MPs (20—50 μm) by the bivalves which should be confirmed by laboratory experiments. Once again, this study highlights the ubiquity of MPs in the marine environment.

Acknowledgments

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.08.032.

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