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Extraction of microplastics from sediment matrices: experimental comparative analysis

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1. Introduction

Anthropogenic litter is a widespread, ubiquitous, and global threat for marine and terrestrial ecosystems. Plastics are the most abundant litter by number in most environments, e.g., >80% at the surface of European rivers (González Fernández *et al.*, 2018). Plastics have low degradability, especially under low UV-radiation, oxygen concentration, and temperature, such as on the sea floor (Andrady, 2017). The degradation of large plastic pieces creates small fragments called microplastics (MPs; <5 mm; GESAMP, 2015), which are more numerous than macroplastics (>5 mm). MPs can also be manufactured directly at micrometric size by plastic industries (GESAMP, 2015). MPs have been found in most biotopes worldwide, even within remote and/or pristine areas (Allen *et al.*, 2019; Bergmann *et al.*, 2017). Due to their small size, they can be ingested by a wide range of organisms, which can be associated with potential harmful toxicology (Wright *et al.*, 2013). In addition, MPs can adsorb a broad range of pollutants from the surrounding environment (Rochman, 2015). They can also desorb pollutants which were used during their fabrications, such as bisphenol A and phthalates (Hirai *et al.*, 2011; Teuten *et al.*, 2009).

The quantification of MPs can be divided into four main steps: sampling, sample treatment, counting, and analysis. The ideal protocol should enable the quantification of all MPs, regardless of length or density. It should also be convenient and cheap to enable large-scale investigations. This challenge is particularly important for studies on sediment matrices (e.g., beach, riverbed, and deep sea) where MPs must be extracted from sediments before the identification step. Most extraction protocols are based on the difference of volumetric mass density between sediments and plastics (He *et al.*, 2021). Recently, oil has been proposed as an interesting alternative for this separation (Mani *et al.*, 2019). Plastics are lipophilic, and therefore, attracted by oil. In addition, plastic and oil are hydrophobic, so they tend to join each other in a water environment. Electrostatic separation (Felsing *et al.*, 2018), magnetic separation (Grbic *et al.*, 2019), solvent extraction (Corti *et al.*, 2020;

Fuller and Gautam, 2016), and depolymerization with phase transfer catalysis (Castelvetto *et al.*, 2020) have also been investigated for this purpose. However, they require expensive equipment, which restrict their uses, despite their remarkable results (Bellasi *et al.*, 2021).

In density separation methods, the use of an extracting solution (ES) with an appropriate volumetric mass density (i.e., less dense than the sediment and denser than the plastic) enables the recovery of MPs at the solution surface. Zinc chloride (ZnCl_2) and sodium iodide (NaI) are the most common high-density solutions used for this purpose ($>1.6 \text{ g/mL}$), and they can be applied to approximately 93% of the plastics produced in Europe (Kedzierski *et al.*, 2017b). However, they are both toxic to the environment and humans, and expensive. Therefore, a hypersaline solution (NaCl) is often preferred because it is non-toxic and cheap. Nevertheless, NaCl only allows the extraction of low-density polymers (<1.2), which represents only half of the European plastic production (Kedzierski *et al.*, 2017b).

This study aims to compare several protocols for the extraction of MPs from sediments. Several ES (oil, water, oil-in-water, NaCl, oil-in-NaCl, and NaI) were tested in combination with three isolation methods (IM) (hand stirring, centrifugation, and aeration). The results are expected to help future studies to solve the challenge of extracting MPs from sediments and soils. Our findings do not emphasize one unique protocol. Instead, they provide information for the selection of the most suitable process, by providing the basis to avoid common mistakes and enabling a more accurate comparison between the methods.

2. Methods

2.1 Environmental sample collection

Sediments were sampled in May 2019 within a dredging disposal area belonging to *Voies Navigables de France* (VNF, Navigable Waterways of France), along the Aa River (St-Omer,

France; 50.773479N, 2.265114E). Two sub-sites with contrasting granulometry were selected. In the first one, the sediments were coarse and heterogeneous, whereas in the second one, they were fine and homogeneous (Table S1). At each sub-site, three samples of approximately 2 kg of wet sediment were collected using a metal trowel just below the surface and between 10 and 20 cm depth. The 2-kg samples were transferred into glass bottles and stored at 4 °C before the tests. Environmental MPs trapped within those sediments have been quantified and characterized in a concomitant study and cannot be confused with added pristine MP used in this work (See details in Fig S1; Constant et al., 2021).

2.2 Recovery experiments

2.2.1 Spiked pristine microplastics

For each experiment, approximately 50 g of wet sediments were transferred to a 100 mL glass container, to which 50 mL of Milli-Q water was added to liquefy the sediment. After one week, the water was removed and discarded using a glass pipette. Subsequently, pristine MPs were added to the container (Table S2). A mixture of low and high-density polymers were used: 10 pellets (2 ± 0.2 mm long) of polyethylene (PE; 0.9 g/cm^3) and polyethylene terephthalate (PET; 1.4 g/cm^3), 10 fragments (1 ± 0.3 mm long) of PE and PET, 10 foams (1 ± 0.2 mm long) comprising polyethylene and acrylonitrile butadiene styrene (PE-ABS; 1.0 g/cm^3), and 10 fibers (4 ± 0.5 mm long) of polyamide (PA; density: 1.2 g/cm^3) and polyester (PES; density: 1.4 g/cm^3). In addition, 10 pieces of cotton fiber were added to represent fibers of heavy density (1.5 g/cm^3). Pellets were purchased from a plastic manufacturer (Acordis©). Fragments were obtained by cryo-crushing the pellets, and fibers were obtained by cutting bobbin threads. The added MPs were easily recognizable from the other particles already present in the samples.

2.2.2 Separation methods

81 He *et al.* (2021) reviewed 144 publications extracting MPs within sediment, soil, sand, sludge, and
82 other complex environments. They noticed that MPs are mostly separated from sediment matrices
83 by gravity. As most synthetic polymers are lighter than sediments, MPs can be extracted with a
84 solution of appropriate density. In density-based and lipophilicity-based separation methods, an ES
85 is added to the sediment, mixed, left to stand for a separation period, and the supernatant is
86 collected from the top or the sediment is removed from the bottom. A wide range of IMs and ESs
87 have already been proposed. In the present study, three different IMs were tested:

- 88 • Hand stirring: manual agitation for 2 min followed by 5 h of decantation, adapted from
89 Thompson *et al.* (2004)
- 90 • Centrifugation: manual agitation for 30 s followed by 5-min centrifugation at 500 rpm, as
91 performed by Phuong *et al.* (2018)
- 92 • Aeration: airflow input for 2 min at 100 mL/s followed by 5 h of decantation, adapted from
93 Nuelle *et al.* (2014)

94 For each IM, six ESs were tested:

- 95 • Water (1 g/mL; Phuong *et al.*, 2018)
- 96 • Canola oil (0.9 g/mL; Rodrigues *et al.*, 2018)
- 97 • Water and a thin layer of canola oil (Crichton *et al.*, 2017)
- 98 • Concentrated NaCl solution (1.2 g/mL; Thompson *et al.*, 2004)
- 99 • Concentrated NaCl solution with a thin layer of canola oil (Karlsson *et al.*, 2017)
- 100 • Concentrated NaI solution (1.6 g/mL; Claessens *et al.*, 2013)

101 For all tests, 100 ± 1 mL of solution was added to 50 ± 5 g of sediment in a glass container. After each
102 density separation process, the supernatant was filtered by GF/F Whatman filters (47 mm diameter;
103 $0.7 \mu\text{m}$ porosity). This extraction procedure was repeated 3 times. Filters were dried at 50°C
104 overnight and then examined under a Leica MZ12 dissecting stereo-microscope ($\times 10$

magnification). First, 36 tests were performed ($3 \text{ IMs} \times 6 \text{ ESs} \times 2 \text{ sediments} \times 1 \text{ replicate}$). Subsequently, the three ES with the best extraction yield (i.e., oil-in-water, concentrated NaCl and NaI solutions) were tested two additional times ($3 \text{ IMs} \times 3 \text{ ESs} \times 2 \text{ sediments} \times 2 \text{ replicates}$), resulting in additional set of 36 tests, for a total of 72 tests for the study (Table 1).

2.3 Data analysis

The extraction rate (%) was calculated for each pristine MP based on the number of MP recovered compared to the number of spiked MP. Statistical analyses were performed using R software (R Core Team, 2018), including the caption package “captioner” (Alathea, 2015), manuscript package “rmarkdown” (Allaire *et al.*, 2018), data manipulation package “dplyr” (Wickham *et al.*, 2017), and graphical package “ggpubr” (Kassambara, 2017). As normality of distribution was not observed (Shapiro-Wilk test), four non-parametric tests were used: the Wilcoxon-Man-Whitney test to compare two groups, Kruskal-Wallis test to compare more than two groups, Scheirer-Ray-Hare test to investigate the influence of two different factors and the interaction among factors, and post hoc Dunn's test to compare differences of all possible pairs and pinpoint specific medians that are significantly different from the others.

3. Results

The recovery rates for 3 IMs and 6 ESs were determined by spiking pristine MPs in fine and coarse-grained sediments. All results (i.e., for every polymer and shape in both sediment types) are shown in Fig. S2. Some trends were observed after the first set of 36 tests (i.e., no replicate). Overall, the recovery rates for low-density (PE and PE-ABS pooled) and high-density polymers (PA, PES, and PET pooled) were significantly different for the various ES (Scheirer-Ray-Hare (SHR) test, both $p < 0.01$; Table S3) but not for different IM (SHR test, $p = 0.11$ and 0.16 , respectively; Table S3). Interactions between ES and IM also created significant differences (SHR test, $p = 0.02 < 0.01$;

128 Table S3). Extraction efficiencies were heterogeneous for every ES, ranging from 0% to 100% (Fig.
 129 1). The recovery rate upon use of oil alone was $37\pm 32\%$ (0–83%; $n = 6$), and it was significantly
 130 lower for low-density polymers (Kruskal-Wallis test, $p < 0.01$; Table S3). Excluding oil, $84\pm 17\%$
 131 (7–100%; $n = 66$) of low-density MPs were recovered, and more than two thirds (68%) of the tests
 132 presented an extraction rate greater than 80%. Excluding the lowest extraction rate of oil-in-water,
 133 the results for ESs (except oil) varied within the range of 50%–100% with the relative standard
 134 deviation (ratio of standard deviation to the mean) varying between 0.11 and 0.21.
 135 For high-density polymers (PA, PES, PET), extraction efficiencies were also heterogeneous.
 136 Excluding the highest extraction rate (1/6 value), results for water and NaCl were all lower than 3%.
 137 The results of other ES varied substantially, and their relative standard deviation ranged between
 138 0.23 and 0.73. Recovery efficiencies for water and oil were $6\pm 11\%$ (0–27%; $n = 6$) and $6\pm 10\%$ (0–
 139 28%; $n = 6$) respectively, which are significantly lower than those for the NaI solution and the oil-
 140 in-NaCl solution (Kruskal-Wallis test, $p < 0.01$; Table S3), at $71\pm 17\%$ (42–90%; $n = 18$) and
 141 $46\pm 26\%$ (0–82%; $n = 18$), respectively. The oil-in-water and NaCl solutions had significantly lower
 142 efficiencies than the NaI solution (Kruskal-Wallis test, $p < 0.01$; Table S3), at $26\pm 20\%$ (10–65%, n
 143 = 18) and $29\pm 22\%$ (2–60%, $n = 6$), respectively, but these results were not significantly different
 144 from those of other ESs. In addition, the NaI solution presented a significantly higher efficiency
 145 than other ESs.
 146 The replicate experiments performed with oil (in water and NaCl solution) and NaI solution
 147 confirmed the previously observed high heterogeneity (Fig. 2). For PA, PET, and PES MPs, the
 148 extraction rates were widely dispersed between 0% and 100% for each of the three selected ES,
 149 with mean values ranging between 13% and 84%, and relative standard deviation ranging between
 150 0.22 and 1.36. NaI was significantly more efficient than oil for PA fibers, PET pellets, and PET
 151 fragments, but not for PES fibers (Kruskal-Wallis tests; Fig. 2). For this last ES, no significant
 152 differences were observed between different IM (i.e., hand stirring, centrifugation, and fluidization;

Kruskal-Wallis tests; Fig. 3) for PA, PET, and PES MPs. For fine sediments, the recovery rate was slightly better with a centrifugation step than other IM for PET, but not for fibers. Extraction rates were slightly better for coarse than fine sediments, but the differences were not significant (SHR test, $p = 0.09$; Table S3).

For each test, the extraction process was repeated three times. The first extraction recovered $67 \pm 35\%$ (0–100%; $n = 72$) of the total recovered MP (i.e., after three extractions). The second and third extractions represented $19 \pm 27\%$ (0%–100%) and $14 \pm 23\%$ (0%–100%) of the total recovered MP, respectively. The extraction efficiency of NaI after only one extraction was $45 \pm 28\%$ (17%–100%; $n = 18$) for low-density polymers and $47 \pm 23\%$ (5%–85%) for high-density polymers. After three extractions, this rate doubled ($89 \pm 11\%$; 67%–100%) and increased one and a half times ($71 \pm 17\%$; 42%–90%) for low- and high-density polymers, respectively.

4. Discussion

The extraction rates varied highly within and among the 18 separation protocols tested (Fig. S1). Regardless of MP and sediment types, extraction efficiencies changed remarkably between the replicates. Such variability implies a low repeatability, which was under the minimum requirements for a reliable MP analysis (Cadiou *et al.*, 2020). Nevertheless, the NaI-centrifugation protocol presented a lower variability (<10%), which was more acceptable, particularly for coarse sediments.

The extraction rates within a protocol did not change significantly for different IM (hand stirring, centrifugation, and aeration) or sediment feature (coarse or fine). Therefore, the most convenient IM can be chosen without influencing the recovery efficiency. Hand stirring was the simplest, easiest, and fastest method. Centrifugation resulted in a solid sediment pellet at the bottom of the container, which limited the resuspension of fine sediments, but it did not require a settling step (5 h). Aeration was the smoothest method, as it limited the fragmentation of weak MP, and the bubbles promoted the separation of MPs adhered to soil or sediments (Nuelle *et al.*, 2014). The different size fractions

177 of sediment grains may lead to different physical properties (Konechnaya *et al.*, 2020). For instance,
178 fine particles can adhere to the surface of MPs, increasing their densities and decreasing the
179 efficiency of density separation methods (Radford *et al.*, 2021). To address this issue, some authors
180 have proposed the use of ultrasonic waves to separate particles (Wang *et al.*, 2018; Zhang *et al.*,
181 2018). The absence of significant differences between fine and coarse sediments in this study may
182 have occurred due to the use of pristine MPs, whereas the surfaces of weathered MPs may react
183 differently (Andrady, 2017).

184 Fragments, pellets, and foams of low-density polymers (PE and PE-ABS) were well extracted
185 (>80%) by all ESs tested, except oil alone. Contrary to Rodrigues *et al.* (2018), which obtained a
186 mean weight recovery of 65% for both low and high-density polymers (PE, PET, PP, PS, and PVC;
187 results not provided for each individual polymer), less than half of MPs spiked were recovered
188 using only oil. Rodrigues *et al.* (2018) mentioned that the recoveries may be overestimated because
189 oil can stay attached to particles even after filtration and drying, thereby increasing the final weight
190 values.

191 To a large extent, extraction rates of high-density polymers (PET, PA, and PES) were lower than
192 those obtained in previous studies (Fig. 4 and Table S4). Part of this relatively lower efficiency
193 might be attributed to the difficulty in collecting MPs from the supernatant. We observed MPs
194 adhered to the glass walls of the container used for extraction, which likely occurred because the
195 MPs used were pristine, and therefore, more prone to electrostatic interactions. Repeated extractions
196 increase the probability to catch those particles. Increasing the number of separation steps may also
197 improve the separation of MPs and sediment matrices. In the literature, several interesting
198 alternative methods have been investigated. Nuelle *et al.* (2014) reported an overflow recovery
199 process that prevents the adhesion and/or removes the MPs adhered to glass. Nakajima *et al.* (2019)
200 proposed a glass device for an easier supernatant separation. Mani *et al.*, (2019) proposed the

201 removal of sediments from the bottom, instead of collecting the supernatant at the top, with the risk
202 of resuspension of fine particles.

203 Denser polymers were poorly extracted when NaCl and oil-in-water were used. NaCl has a low
204 density and has been reported by many studies to be less efficient than other substances, as shown
205 in Figure 4 (Han *et al.*, 2019; Li *et al.*, 2019; Zhang *et al.*, 2020). Oil-in-water has been
206 recommended as a good ES as it is neither harmful nor expensive. The separation of sediments and
207 MPs is not based on densities, but on the hydrophobicity of plastics and oil. Nevertheless, lipophilic
208 properties of plastics may be altered by contaminants on MP surfaces (He *et al.*, 2021). Several
209 authors obtained recovery rates >90% for dense polymers in sediments and wastewater effluents
210 (Crichton *et al.*, 2017; Mani *et al.*, 2019; Scopetani *et al.*, 2020). As previously mentioned, oil is
211 difficult to remove and interferes with the Fourier transform infrared spectroscopy and Raman
212 spectra, but the use of aggressive detergents can be efficient for the elimination of oil (Mani *et al.*,
213 2019). In this study, the addition of few milliliters of oil (oil-in-water and in saline solutions) did
214 not significantly change the recovery rates of water and saline solutions alone. Finally, NaI
215 presented the best extraction efficiency for high-density fragments, pellets, and fibers. Most studies
216 using ES with a density higher than 1.5 achieved quantitative extraction rates (>90%; Table S4).
217 This indicates that the ES (i.e., chemicals) probably exerts less influence than density. NaI has been
218 commonly used because it can reach a density of 1.8, and the solution can be reused several times
219 (Kedzierski *et al.*, 2017b). In addition, some methods can significantly reduce the amount of
220 sediment required for the process. For example, the elutriation method developed by Kedzierski *et*
221 *al.* (2016) is based on particle-size subdivision and can extract >90% of dense plastics, with a sand
222 recovery <1%, which considerably reduces the amount of NaI required. However, this method was
223 developed and improved only for sand sediments (Kedzierski *et al.*, 2018, 2017a). Its efficiency
224 would likely be lower for fine and/or organic matter-rich sediments, which can aggregate and/or

225 react with plastics (Zhang and Liu, 2018). Organic matter can be removed using an oxidant such as
226 hydrogen peroxide, and aggregation can be prevented using a dispersal agent.

227 Currently, the methodologies are relatively similar to extract MPs from the different complex solid
228 matrices (He et al., 2021; Junhao et al., 2021). However, matrices features (organic matter, particle-
229 size, etc.) may severely influence the extracting rate, promoting one approach rather than the others.
230 Some methods, recently developed, are less reliant on matrices characteristics. Solvent extractions
231 ensure high recoveries, but dissolve the MP items (La Nasa et al., 2021; Stile et al., 2021; Wen et al.,
232 2021). Tötzke et al., (2021) proposed to use complementary neutron and X-ray tomography to
233 identify MPs in sediment, without extracting them. Finally, current methods cannot accurately extract
234 all types of plastics (size, shape, and density). Therefore, monitoring studies should choose and
235 optimize their protocols according to their scientific requirements, and associate different methods
236 to widen the extractable range of plastics.

237 **5. Conclusions**

238 This study investigated a major challenge for the quantification of MPs within sediment matrices:
239 the separation between sediments and MPs. Eighteen combinations of IM and ES were tested for
240 light and dense pristine polymers on fine and coarse sediments. The main conclusions based on 72
241 tests are as follows:

- 242 (1) The IMs (hand stirring, centrifugation, and aeration) and sediment features (coarse or fine) did
243 not affect extraction efficiencies;
- 244 (2) Fragments, pellets, and foams of low-density polymers (PE and PE-ABS) were well extracted
245 (>80%) for all tested ESs, excepted pure oil;
- 246 (3) Overall, NaI presented the best extraction efficiency for the extraction of fragments, pellets, and
247 fibers of high-density polymers;

248 (4) The addition of few milliliters of oil to the aqueous solutions did not improve the recovery of
249 pristine MPs. Nevertheless, the oil-in-saline solution presented a slight better extraction than water
250 and oil alone;

251 (5) Repeated extractions improved recovery efficiency.

252 This work should be continued using MPs artificially altered in laboratory and MPs directly
253 recovered from the environment so that possible variations in electrostatic effects and other
254 adsorption mechanisms can be evaluated. The interactions between MPs and sedimentary particles
255 (such as clays and organic matter), which can modify densities and probably their extractability
256 rates, should also be investigated.

257

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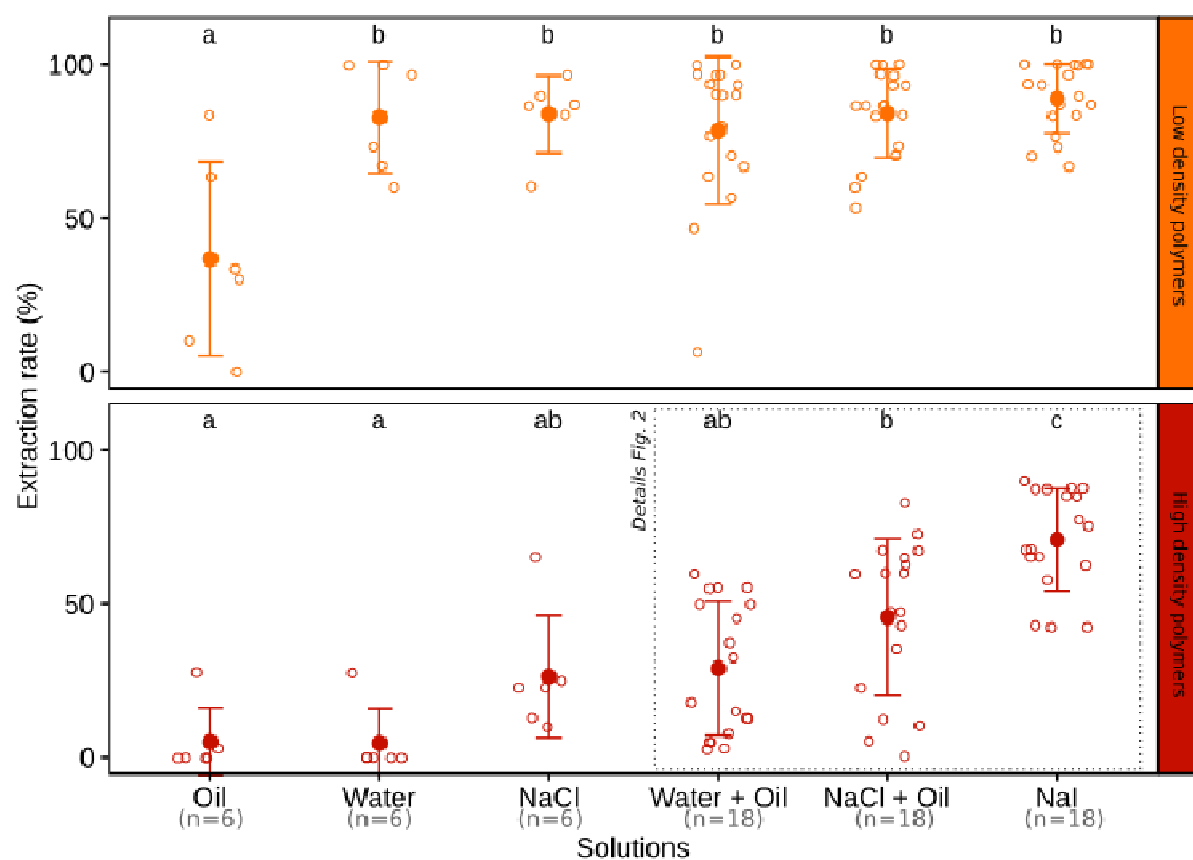


Fig. 1. Extraction rates (%) of low (orange) and high (red) density polymers for each extracting solutions. Small empty circles show results of each tests (i.e. 3 agitation methods \times 2 sediment type \times 1 or 3 replicates). Large solid circles and bars represent means and standard deviations, respectively. Compact letters (a, b and c) indicate groups with significant differences. Extraction rate for each 7 types of spiked MPs were pooled by polymer density. Low density: PE and PE-ABS. High density: PET, PA and PES. See details about statistical tests in section 2.3.

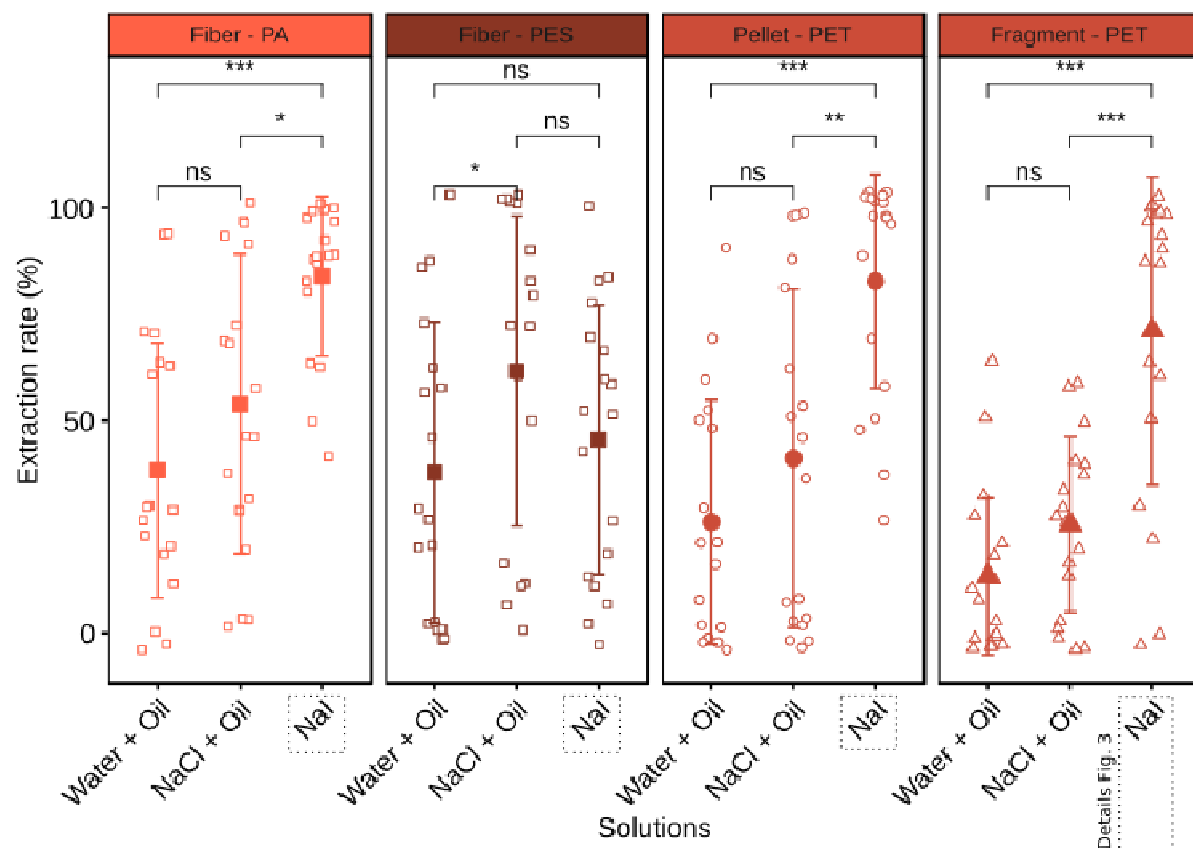


Fig. 2. Extraction rates (%) of each high density polymers for three extracting solutions. Small empty circles, triangles and squares show results for 10 pellets, fragments and fibers, respectively. Large solid circles and bars represent means and standard deviations, respectively. ns: Non-significant (p-value > 0.05). *: Significant (p-value < 0.05). **: Highly significant (p-value < 0.01). ***: Very highly significant (p-value < 0.001). See details about statistical tests in section 2.3.

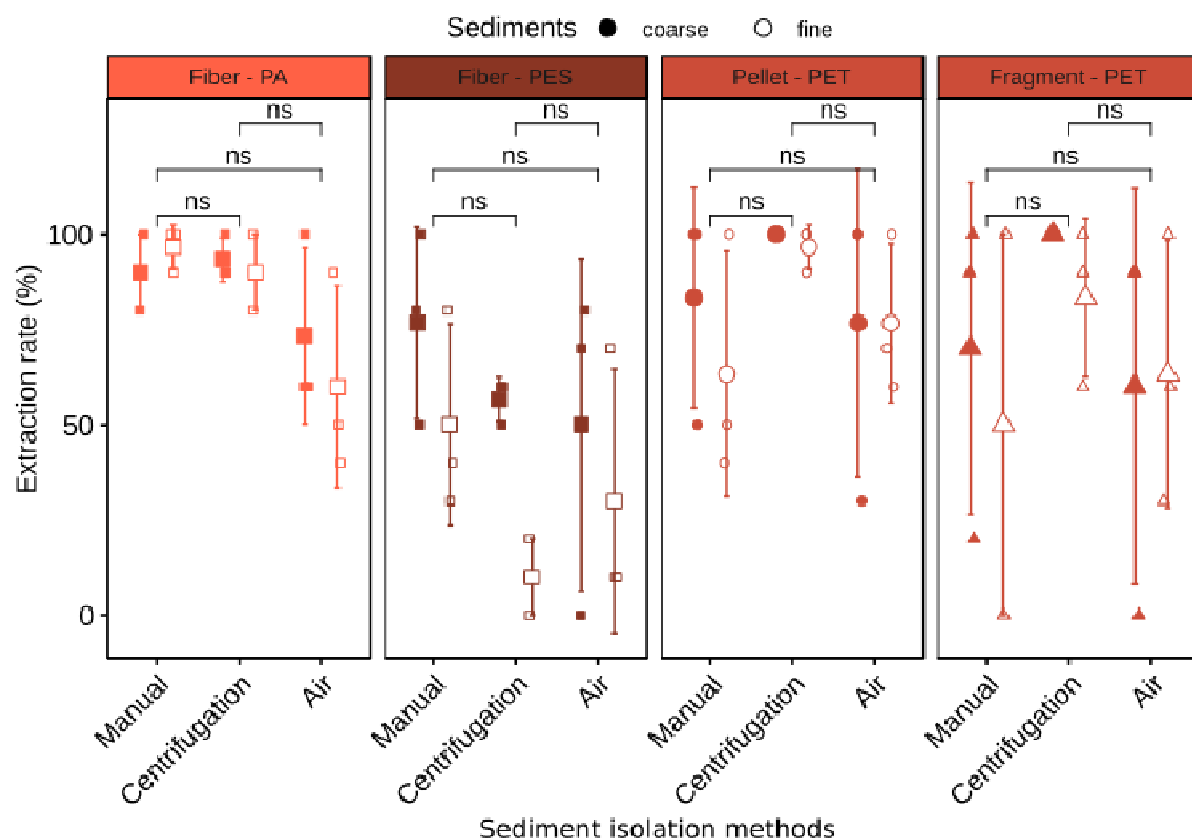


Fig. 3. Extraction rates (%) of each high density polymers obtained with the NaI solution for each methods and both sediment types. Small circles, triangles and squares show results for 10 pellets, fragments and fibers, respectively. Large icons and bars represent means and standard deviations, respectively. Coarse and fine sediments were pooled for statistical tests (*i.e.* 6 replicates per group). ns: Non-significant (p-value > 0.05). *: Significant (p-value < 0.05). **: Highly significant (p-value < 0.01). ***: Very highly significant (p-value < 0.001). See details about statistical tests in section 2.3

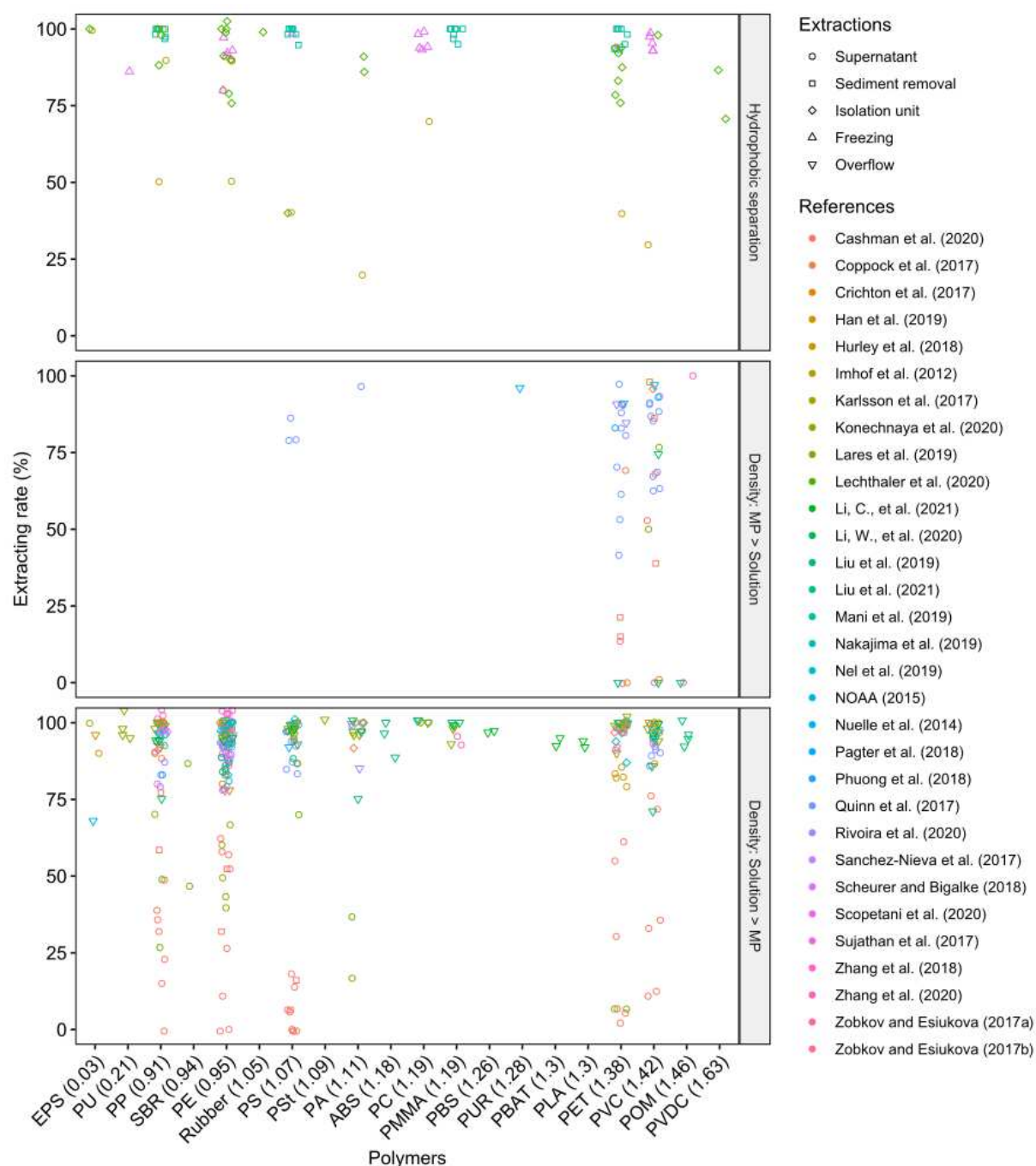


Fig. 4. Extraction rates (%) retrieved from 30 studies. Extractions: methods to collect MPs after being isolated from sediment.

Table 1. Procedures performed for extracting spiked microplastics in fine and coarse sediments. 18 combinations with 3 stirring methods and 6 solutions were tested with 7 types of pristine microplastics. Each combination was tested for coarse and fine sediments (2), tests for 3 solutions (Oil in water and in NaCl, and NaI) were repeated 3 times ($\times 3$).

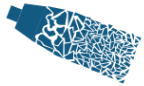
Solution	Manual	Centrifugation	Air	Total
Oil	2	2	2	6
Water	2	2	2	6
Water + Oil	2×3	2×3	2×3	18
NaCl	2	2	2	6
NaCl + Oil	2×3	2×3	2×3	18
NaI	2×3	2×3	2×3	18
Total	24	24	24	72

Materials



Sediments

+



Microplastics

+

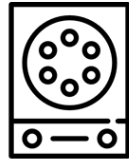


Extracting
solutions

Separation methods



Air insufflation



Centrifugation



Hand-stirring

Extraction rate (%)

