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# 1                   **Extraction of microplastics from sediment matrices:** 2                   **experimental comparative analysis**

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10

# 11 **1. Introduction**

12 Anthropogenic litter is a widespread, ubiquitous, and global threat for marine and terrestrial  
13 ecosystems. Plastics are the most abundant litter by number in most environments, e.g., >80% at the  
14 surface of European rivers (González Fernández *et al.*, 2018). Plastics have low degradability,  
15 especially under low UV-radiation, oxygen concentration, and temperature, such as on the sea floor  
16 (Andrady, 2017). The degradation of large plastic pieces creates small fragments called  
17 microplastics (MPs; <5 mm; GESAMP, 2015), which are more numerous than macroplastics (>5  
18 mm). MPs can also be manufactured directly at micrometric size by plastic industries (GESAMP,  
19 2015). MPs have been found in most biotopes worldwide, even within remote and/or pristine areas  
20 (Allen *et al.*, 2019; Bergmann *et al.*, 2017). Due to their small size, they can be ingested by a wide  
21 range of organisms, which can be associated with potential harmful toxicology (Wright *et al.*, 2013).  
22 In addition, MPs can adsorb a broad range of pollutants from the surrounding environment  
23 (Rochman, 2015). They can also desorb pollutants which were used during their fabrications, such  
24 as bisphenol A and phthalates (Hirai *et al.*, 2011; Teuten *et al.*, 2009).

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

35 Fuller and Gautam, 2016), and depolymerization with phase transfer catalysis (Castelvetto *et al.*,  
36 2020) have also been investigated for this purpose. However, they require expensive equipment,  
37 which restrict their uses, despite their remarkable results (Bellasi *et al.*, 2021).

38 In density separation methods, the use of an extracting solution (ES) with an appropriate volumetric  
39 mass density (i.e., less dense than the sediment and denser than the plastic) enables the recovery of  
40 MPs at the solution surface. Zinc chloride ( $ZnCl_2$ ) and sodium iodide (NaI) are the most common  
41 high-density solutions used for this purpose ( $>1.6$  g/mL), and they can be applied to approximately  
42 93% of the plastics produced in Europe (Kedzierski *et al.*, 2017b). However, they are both toxic to  
43 the environment and humans, and expensive. Therefore, a hypersaline solution (NaCl) is often  
44 preferred because it is non-toxic and cheap. Nevertheless, NaCl only allows the extraction of low-  
45 density polymers ( $<1.2$ ), which represents only half of the European plastic production (Kedzierski  
46 *et al.*, 2017b).

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

## 54 **2. Methods**

### 55 **2.1 Environmental sample collection**

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

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

## 66 **2.2 Recovery experiments**

### 67 **2.2.1 Spiked pristine microplastics**

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

### 80 **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,  $100\pm 1$  mL of solution was added to  $50\pm 5$  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  $50\ ^\circ\text{C}$   
104 overnight and then examined under a Leica MZ12 dissecting stereo-microscope ( $\times 10$

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

## 109 **2.3 Data analysis**

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

## 120 **3. Results**

121 The recovery rates for 3 IMs and 6 ESs were determined by spiking pristine MPs in fine and coarse-  
122 grained sediments. All results (i.e., for every polymer and shape in both sediment types) are shown  
123 in Fig. S2. Some trends were observed after the first set of 36 tests (i.e., no replicate). Overall, the  
124 recovery rates for low-density (PE and PE-ABS pooled) and high-density polymers (PA, PES, and  
125 PET pooled) were significantly different for the various ES (Scheirer-Ray-Hare (SHR) test, both  $p <$   
126  $0.01$ ; Table S3) but not for different IM (SHR test,  $p = 0.11$  and  $0.16$ , respectively; Table S3).  
127 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;



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

157 For each test, the extraction process was repeated three times. The first extraction recovered  
158  $67 \pm 35\%$  (0–100%;  $n = 72$ ) of the total recovered MP (i.e., after three extractions). The second and  
159 third extractions represented  $19 \pm 27\%$  (0%–100%) and  $14 \pm 23\%$  (0%–100%) of the total recovered  
160 MP, respectively. The extraction efficiency of NaI after only one extraction was  $45 \pm 28\%$  (17%–  
161 100%;  $n = 18$ ) for low-density polymers and  $47 \pm 23\%$  (5%–85%) for high-density polymers. After  
162 three extractions, this rate doubled ( $89 \pm 11\%$ ; 67%–100%) and increased one and a half times  
163 ( $71 \pm 17\%$ ; 42%–90%) for low- and high-density polymers, respectively.

## 164 **4. Discussion**

165 The extraction rates varied highly within and among the 18 separation protocols tested (Fig. S1).  
166 Regardless of MP and sediment types, extraction efficiencies changed remarkably between the  
167 replicates. Such variability implies a low repeatability, which was under the minimum requirements  
168 for a reliable MP analysis (Cadiou *et al.*, 2020). Nevertheless, the NaI-centrifugation protocol  
169 presented a lower variability (<10%), which was more acceptable, particularly for coarse sediments.  
170 The extraction rates within a protocol did not change significantly for different IM (hand stirring,  
171 centrifugation, and aeration) or sediment feature (coarse or fine). Therefore, the most convenient IM  
172 can be chosen without influencing the recovery efficiency. Hand stirring was the simplest, easiest,  
173 and fastest method. Centrifugation resulted in a solid sediment pellet at the bottom of the container,  
174 which limited the resuspension of fine sediments, but it did not require a settling step (5 h). Aeration  
175 was the smoothest method, as it limited the fragmentation of weak MP, and the bubbles promoted  
176 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, matrix 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 matrix 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

## 258 **References**

- 259 Alatheia, L., 2015. Captioner: numbers figures and creates simple captions. [https://CRAN.R-](https://CRAN.R-project.org/package=captioner)  
260 [project.org/package=captioner](https://CRAN.R-project.org/package=captioner)
- 261 Allaire, J., Xie, Y., McPherson, J., Luraschi, J., Ushey, K., Atkins, A., Wickham, H., Cheng, J.,  
262 Chang, W., 2018. Rmarkdown: dynamic documents for R. [https://CRAN.R-](https://CRAN.R-project.org/package=rmarkdown)  
263 [project.org/package=rmarkdown](https://CRAN.R-project.org/package=rmarkdown)
- 264 Allen, S., Allen, D., Phoenix, V.R., Le Roux, G., Durántez Jiménez, P., Simonneau, A., Binet, S.,  
265 Galop, D., 2019. Atmospheric transport and deposition of microplastics in a remote mountain  
266 catchment. *Nat. Geosci.* 12, 339–344. <https://doi.org/10.1038/s41561-019-0335-5>
- 267 Andrady, A.L., 2017. The plastic in microplastics: A review. *Mar. Pollut. Bull.* 119, 12–22.  
268 <https://doi.org/10.1016/j.marpolbul.2017.01.082>
- 269 Bellasi, A., Binda, G., Pozzi, A., Boldrocchi, G., Bettinetti, R., 2021. The extraction of microplastics  
270 from sediments: An overview of existing methods and the proposal of a new and green  
271 alternative. *Chemosphere* 278, 130357. <https://doi.org/10.1016/j.chemosphere.2021.130357>
- 272 Bergmann, M., Wirzberger, V., Krumpfen, T., Lorenz, C., Primpke, S., Tekman, M.B., Gerdts, G.,  
273 2017. High Quantities of Microplastic in Arctic Deep-Sea Sediments from the  
274 HAUSGARTEN Observatory. *Environ. Sci. Technol.* 51, 11000–11010.  
275 <https://doi.org/10.1021/acs.est.7b03331>
- 276 Cadiou, J.-F., Gerigny, O., Koren, Š., Zeri, C., Kaberi, H., Alomar, C., Panti, C., Fossi, M.C.,  
277 Adamopoulou, A., Digka, N., Deudero, S., Concato, M., Carbonell, A., Baini, M., Galli, M.,  
278 Galgani, F., 2020. Lessons learned from an intercalibration exercise on the quantification and  
279 characterisation of microplastic particles in sediment and water samples. *Mar. Pollut. Bull.*  
280 154, 111097. <https://doi.org/10.1016/j.marpolbul.2020.111097>
- 281 Castelvetro, V., Corti, A., Bianchi, S., Ceccarini, A., Manariti, A., Vinciguerra, V., 2020.  
282 Quantification of poly(ethylene terephthalate) micro- and nanoparticle contaminants in marine  
283 sediments and other environmental matrices. *J. Hazard. Mater.* 385, 121517.  
284 <https://doi.org/10.1016/j.jhazmat.2019.121517>
- 285 Claessens, M., Van Cauwenberghe, L., Vandegehuchte, M.B., Janssen, C.R., 2013. New techniques  
286 for the detection of microplastics in sediments and field collected organisms. *Mar. Pollut. Bull.*  
287 70, 227–233. <https://doi.org/10.1016/j.marpolbul.2013.03.009>
- 288 Constant, M., Alary, C., De Waele, I., Dumoulin, D., Breton, N., Billon, G., 2021. To What Extent  
289 Can Micro- and Macroplastics Be Trapped in Sedimentary Particles? A Case Study  
290 Investigating Dredged Sediments. *Environ. Sci. Technol.* 55, 5898–5905.  
291 <https://doi.org/10.1021/acs.est.0c08386>
- 292 Corti, A., Vinciguerra, V., Iannilli, V., Pietrelli, L., Manariti, A., Bianchi, S., Petri, A., Cifelli, M.,  
293 Domenici, V., Castelvetro, V., 2020. Thorough Multianalytical Characterization and  
294 Quantification of Micro- and Nanoplastics from Bracciano Lake’s Sediments. *Sustainability*  
295 12, 878. <https://doi.org/10.3390/su12030878>
- 296 Crichton, E.M., Noël, M., Gies, E.A., Ross, P.S., 2017. A novel, density-independent and FTIR-  
297 compatible approach for the rapid extraction of microplastics from aquatic sediments. *Anal.*  
298 *Methods* 9, 1419–1428. <https://doi.org/10.1039/C6AY02733D>
- 299 Felsing, S., Kochleus, C., Buchinger, S., Brennholt, N., Stock, F., Reifferscheid, G., 2018. A new  
300 approach in separating microplastics from environmental samples based on their electrostatic  
301 behavior. *Environ. Pollut.* 234, 20–28. <https://doi.org/10.1016/j.envpol.2017.11.013>
- 302 Fuller, S., Gautam, A., 2016. A Procedure for Measuring Microplastics using Pressurized Fluid  
303 Extraction. *Environ. Sci. Technol.* 50, 5774–5780. <https://doi.org/10.1021/acs.est.6b00816>

304 GESAMP, 2015. Sources, fate and effects of microplastics in the marine environment: a global  
305 assessment, IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP Joint Group  
306 of Experts on the Scientific Aspects of Marine Environmental Protection). ed, Rep. Stud.  
307 GESAMP. <https://doi.org/10.13140/RG.2.1.3803.7925>

308 González Fernández, D., Hanke, G., Kideys, A., Alicia, N., Sanchez-Vidal, A., Bruge, A., Bayram,  
309 öztürk, Palma, C., Santinelli, C., Daan, D., Damia, B., Elias, D., Rojo-Nieto, E., Fernando, F.,  
310 Bessa, F., Suaria, G., Siedlewicz, G., Castro-Jiménez, J., Joana, G., Yuri, G., 2018. Floating  
311 Macro Litter in European Rivers-Top Items. <https://doi.org/10.2760/316058>

312 Grbic, J., Nguyen, B., Guo, E., You, J.B., Sinton, D., Rochman, C.M., 2019. Magnetic Extraction of  
313 Microplastics from Environmental Samples. *Environ. Sci. Technol. Lett.* 6, 68–72.  
314 <https://doi.org/10.1021/acs.estlett.8b00671>

315 Han, X., Lu, X., Vogt, R.D., 2019. An optimized density-based approach for extracting  
316 microplastics from soil and sediment samples. *Environ. Pollut.* 254, 113009.  
317 <https://doi.org/10.1016/j.envpol.2019.113009>

318 He, D., Zhang, X., Hu, J., 2021. Methods for separating microplastics from complex solid matrices:  
319 Comparative analysis. *J. Hazard. Mater.* 409, 124640.  
320 <https://doi.org/10.1016/j.jhazmat.2020.124640>

321 Hirai, H., Takada, H., Ogata, Y., Yamashita, R., Mizukawa, K., Saha, M., Kwan, C., Moore, C.,  
322 Gray, H., Laursen, D., Zettler, E.R., Farrington, J.W., Reddy, C.M., Peacock, E.E., Ward,  
323 M.W., 2011. Organic micropollutants in marine plastics debris from the open ocean and  
324 remote and urban beaches. *Mar. Pollut. Bull.* 62, 1683–1692.  
325 <https://doi.org/10.1016/j.marpolbul.2011.06.004>

326 Junhao, C., Xining, Z., Xiaodong, G., Li, Z., Qi, H., Siddique, K.H.M., 2021. Extraction and  
327 identification methods of microplastics and nanoplastics in agricultural soil: A review. *J.*  
328 *Environ. Manage.* 294, 112997. <https://doi.org/10.1016/j.jenvman.2021.112997>

329 Karlsson, T.M., Vethaak, A.D., Almroth, B.C., Ariese, F., van Velzen, M., Hassellöv, M., Leslie,  
330 H.A., 2017. Screening for microplastics in sediment, water, marine invertebrates and fish:  
331 Method development and microplastic accumulation. *Mar. Pollut. Bull.* 122, 403–408.  
332 <https://doi.org/10.1016/j.marpolbul.2017.06.081>

333 Kassambara, A., 2017. Ggpubr: ‘Ggplot2’ based publication ready plots. [https://doi.org/  
334 https://CRAN.R-project.org/package=ggpubr](https://doi.org/https://CRAN.R-project.org/package=ggpubr)

335 Kedzierski, M., Le Tilly, V., Bourseau, P., Bellegou, H., César, G., Sire, O., Bruzaud, S., 2017a.  
336 Microplastics elutriation system. Part A: Numerical modeling. *Mar. Pollut. Bull.* 119, 151–  
337 161. <https://doi.org/10.1016/j.marpolbul.2017.04.060>

338 Kedzierski, M., Le Tilly, V., Bourseau, P., Bellegou, H., César, G., Sire, O., Bruzaud, S., 2016.  
339 Microplastics elutriation from sandy sediments: A granulometric approach. *Mar. Pollut. Bull.*  
340 107, 315–323. <https://doi.org/10.1016/j.marpolbul.2016.03.041>

341 Kedzierski, M., Le Tilly, V., Bourseau, P., César, G., Sire, O., Bruzaud, S., 2018. Microplastics  
342 elutriation system: Part B: Insight of the next generation. *Mar. Pollut. Bull.* 133, 9–17.  
343 <https://doi.org/10.1016/j.marpolbul.2018.05.011>

344 Kedzierski, M., Le Tilly, V., César, G., Sire, O., Bruzaud, S., 2017b. Efficient microplastics  
345 extraction from sand. A cost effective methodology based on sodium iodide recycling. *Mar.*  
346 *Pollut. Bull.* 115, 120–129. <https://doi.org/10.1016/j.marpolbul.2016.12.002>

347 Konechnaya, O., Luchtrath, S., Dsikowitzky, L., Schwarzbauer, J., 2020. Optimized microplastic  
348 analysis based on size fractionation, density separation and  $\mu$ -FTIR. *Water Sci. Technol.* 81,  
349 834–844. <https://doi.org/10.2166/wst.2020.173>

350 La Nasa, J., Biale, G., Mattonai, M., Modugno, F., 2021. Microwave-assisted solvent extraction and  
351 double-shot analytical pyrolysis for the quali-quantitation of plasticizers and microplastics in  
352 beach sand samples. *J. Hazard. Mater.* 401, 123287.  
353 <https://doi.org/10.1016/j.jhazmat.2020.123287>

354 Li, Q., Wu, J., Zhao, X., Gu, X., Ji, R., 2019. Separation and identification of microplastics from  
355 soil and sewage sludge. *Environ. Pollut.* 254, 113076.  
356 <https://doi.org/10.1016/j.envpol.2019.113076>

357 Mani, T., Frehland, S., Kalberer, A., Burkhardt-Holm, P., 2019. Using castor oil to separate  
358 microplastics from four different environmental matrices. *Anal. Methods* 11, 1788–1794.  
359 <https://doi.org/10.1039/C8AY02559B>

360 Nakajima, R., Tsuchiya, M., Lindsay, D.J., Kitahashi, T., Fujikura, K., Fukushima, T., 2019. A new  
361 small device made of glass for separating microplastics from marine and freshwater sediments.  
362 *PeerJ* 7, e7915. <https://doi.org/10.7717/peerj.7915>

363 Nuelle, M.-T., Dekiff, J.H., Remy, D., Fries, E., 2014. A new analytical approach for monitoring  
364 microplastics in marine sediments. *Environ. Pollut.* 184, 161–169.  
365 <https://doi.org/10.1016/j.envpol.2013.07.027>

366 Phuong, N.N., Poirier, L., Lagarde, F., Kamari, A., Zalouk-Vergnoux, A., 2018. Microplastic  
367 abundance and characteristics in French Atlantic coastal sediments using a new extraction  
368 method. *Environ. Pollut.* 243, 228–237. <https://doi.org/10.1016/j.envpol.2018.08.032>

369 R Core Team, 2018. R: A Language and Environment for Statistical Computing. R Foundation for  
370 Statistical Computing, Vienna, Austria. <https://www.R-project.org/>

371 Radford, F., Zapata-Restrepo, L.M., Horton, A.A., Hudson, M.D., Shaw, P.J., Williams, I.D., 2021.  
372 Developing a systematic method for extraction of microplastics in soils. *Anal. Methods*.  
373 <https://doi.org/10.1039/D0AY02086A>

374 Rochman, C.M., 2015. The Complex Mixture, Fate and Toxicity of Chemicals Associated with  
375 Plastic Debris in the Marine Environment, in: Bergmann, M., Gutow, L., Klages, M. (Eds.),  
376 *Marine Anthropogenic Litter*. Springer International Publishing, Cham, pp. 117–140.  
377 [https://doi.org/10.1007/978-3-319-16510-3\\_5](https://doi.org/10.1007/978-3-319-16510-3_5)

378 Rodrigues, M.O., Gonçalves, A.M.M., Gonçalves, F.J.M., Nogueira, H., Marques, J.C., Abrantes, N.,  
379 2018. Effectiveness of a methodology of microplastics isolation for environmental monitoring  
380 in freshwater systems. *Ecol. Indic.* 89, 488–495. <https://doi.org/10.1016/j.ecolind.2018.02.038>

381 Scopetani, C., Chelazzi, D., Mikola, J., Leiniö, V., Heikkinen, R., Cincinelli, A., Pellinen, J., 2020.  
382 Olive oil-based method for the extraction, quantification and identification of microplastics in  
383 soil and compost samples. *Sci. Total Environ.* 733, 139338.  
384 <https://doi.org/10.1016/j.scitotenv.2020.139338>

385 Stile, N., Raguso, C., Pedruzzi, A., Cetojevic, E., Lasagni, M., Sanchez-Vidal, A., Saliu, F., 2021.  
386 Extraction of microplastic from marine sediments: A comparison between pressurized solvent  
387 extraction and density separation. *Mar. Pollut. Bull.* 168, 112436.  
388 <https://doi.org/10.1016/j.marpolbul.2021.112436>

389 Teuten, E.L., Saquing, J.M., Knappe, D.R.U., Barlaz, M.A., Jonsson, S., Björn, A., Rowland, S.J.,  
390 Thompson, R.C., Galloway, T.S., Yamashita, R., Ochi, D., Watanuki, Y., Moore, C., Viet, P.H.,  
391 Tana, T.S., Prudente, M., Boonyatumanond, R., Zakaria, M.P., Akkhavong, K., Ogata, Y.,  
392 Hirai, H., Iwasa, S., Mizukawa, K., Hagino, Y., Imamura, A., Saha, M., Takada, H., 2009.  
393 Transport and release of chemicals from plastics to the environment and to wildlife. *Philos.*  
394 *Trans. R. Soc. B Biol. Sci.* 364, 2027–2045. <https://doi.org/10.1098/rstb.2008.0284>

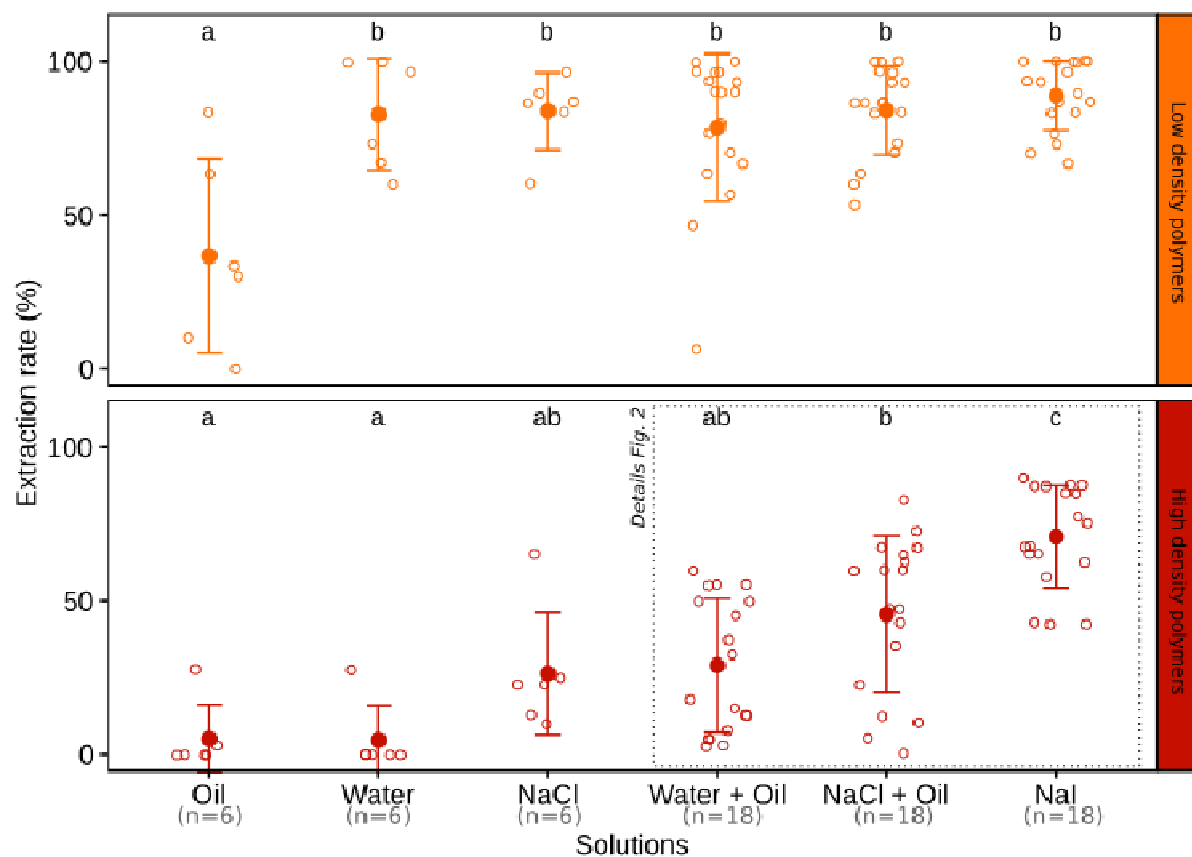
395 Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D.,  
396 Russell, A.E., 2004. Lost at Sea: Where Is All the Plastic? *Science* 304, 838–838.  
397 <https://doi.org/10.1126/science.1094559>

398 Tötze, C., Oswald, S.E., Hilger, A., Kardjilov, N., 2021. Non-invasive detection and localization of  
399 microplastic particles in a sandy sediment by complementary neutron and X-ray tomography.  
400 *J. Soils Sediments*. <https://doi.org/10.1007/s11368-021-02882-6>

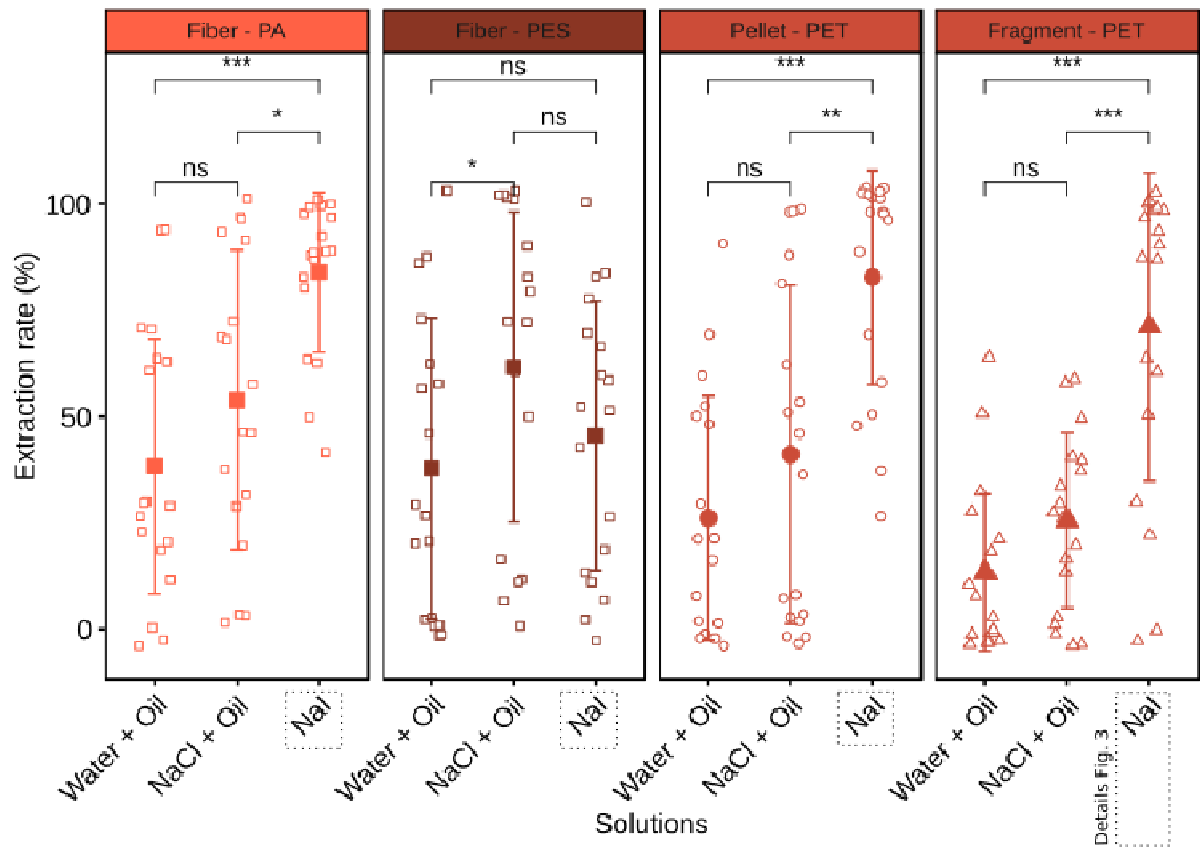
401 Wang, Z., Taylor, S.E., Sharma, P., Flury, M., 2018. Poor extraction efficiencies of polystyrene  
402 nano- and microplastics from biosolids and soil. *Plos One* 13, e0208009.  
403 <https://doi.org/10.1371/journal.pone.0208009>



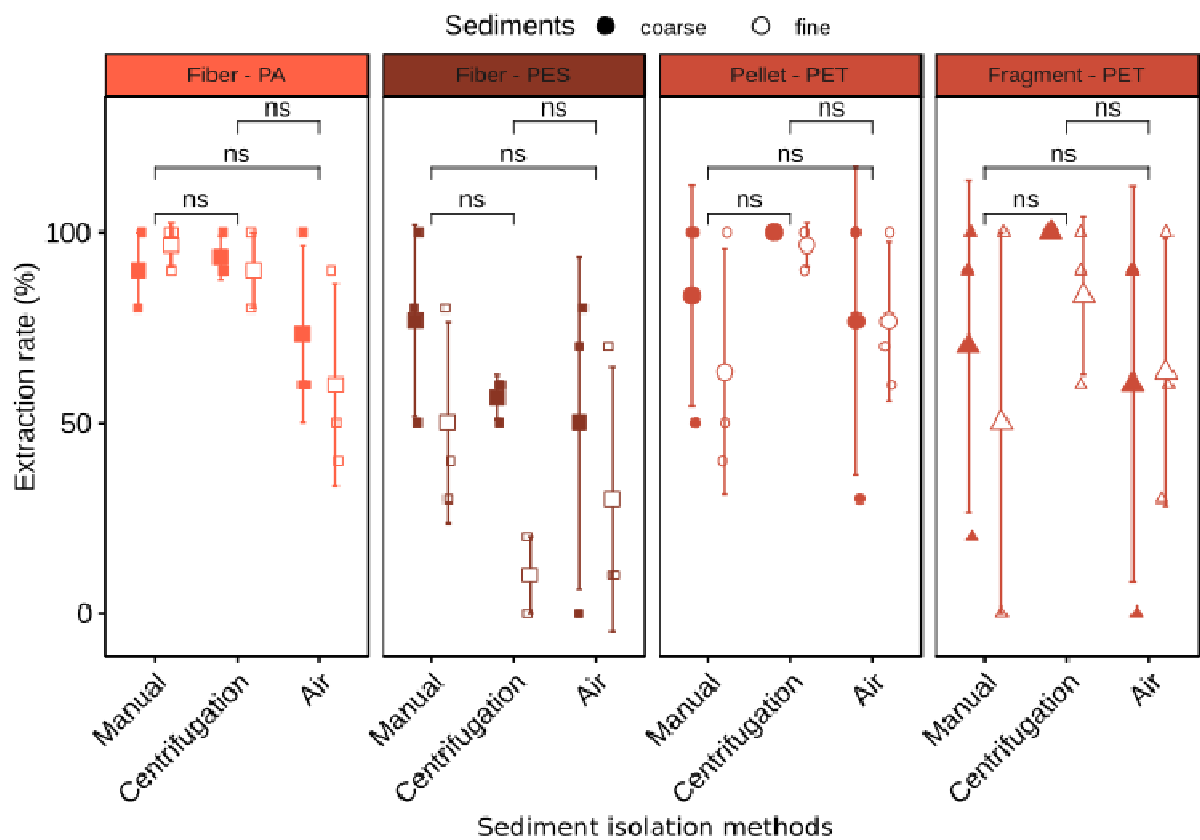
404 Wen, D., Chen, Y., Tong, Y., Wang, H., Zhang, H., Luo, Y., 2021. Quantification of Microplastics in  
405 Soils Using Accelerated Solvent Extraction: Comparison with a Visual Sorting Method. *Bull.*  
406 *Environ. Contam. Toxicol.* <https://doi.org/10.1007/s00128-021-03200-z>  
407 Wickham, H., François, R., Henry, L., Müller, K., 2017. dplyr: a grammar of data ma-nipulation.  
408 <https://CRAN.R-project.org/package=dplyr>  
409 Wright, S.L., Thompson, R.C., Galloway, T.S., 2013. The physical impacts of microplastics on  
410 marine organisms: A review. *Environ. Pollut.* 178, 483–492.  
411 <https://doi.org/10.1016/j.envpol.2013.02.031>  
412 Zhang, G.S., Liu, Y.F., 2018. The distribution of microplastics in soil aggregate fractions in  
413 southwestern China. *Sci. Total Environ.* 642, 12–20.  
414 <https://doi.org/10.1016/j.scitotenv.2018.06.004>  
415 Zhang, S., Yang, X., Gertsen, H., Peters, P., Salanki, T., Geissen, V., 2018. A simple method for the  
416 extraction and identification of light density microplastics from soil. *Sci. Total Environ.* 616,  
417 1056–1065. <https://doi.org/10.1016/j.scitotenv.2017.10.213>  
418 Zhang, X., Yu, K., Zhang, H., Liu, Y., He, J., Liu, X., Jiang, J., 2020. A novel heating-assisted  
419 density separation method for extracting microplastics from sediments. *Chemosphere* 256,  
420 127039. <https://doi.org/10.1016/j.chemosphere.2020.127039>  
421



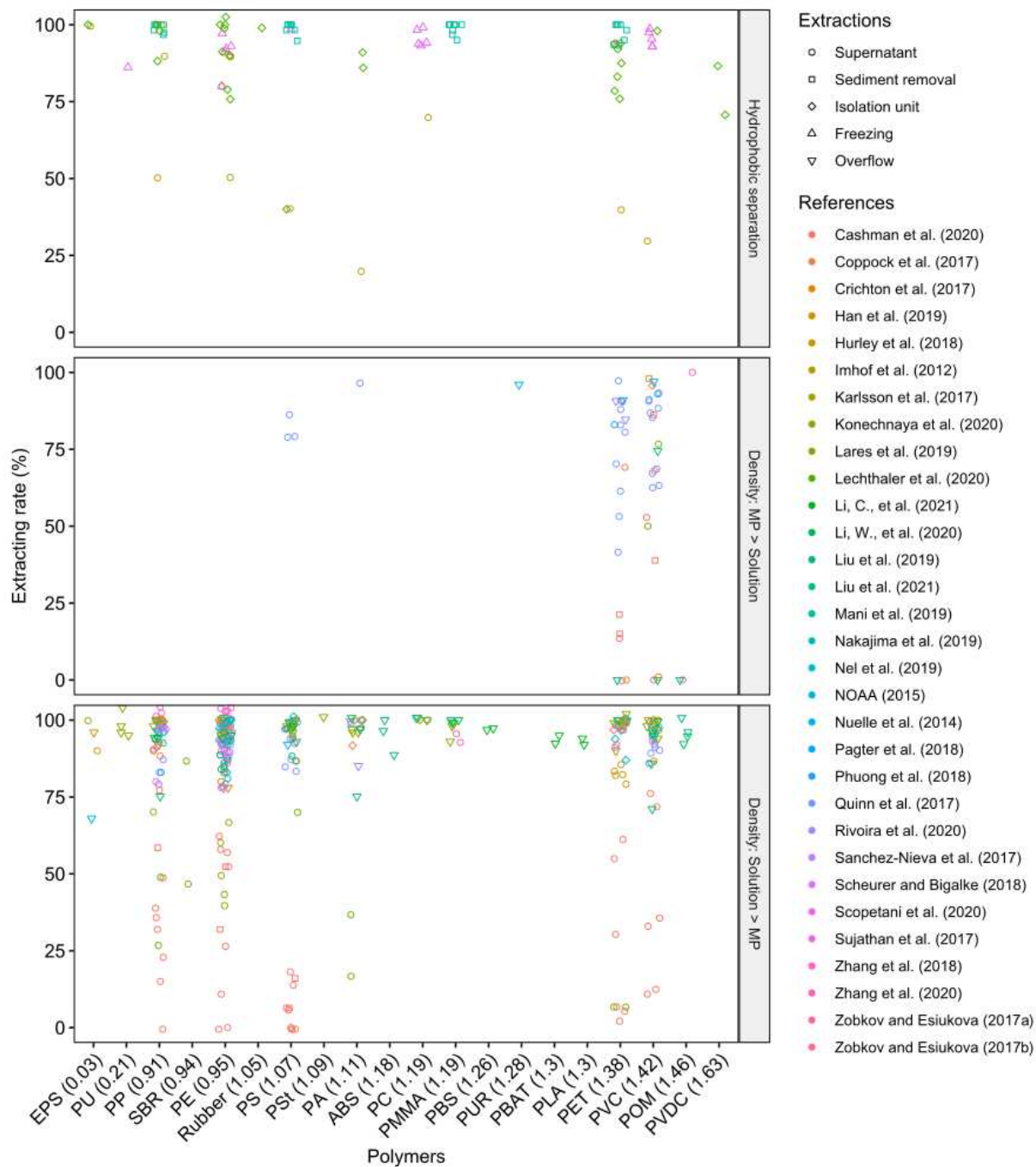
**Fig. 1.** Extraction rates (%) of low (orange) and high (red) density polymers for each extracting solutions. Small empty circles show results of each tests (i.e. 3 agitation methods  $\times$  2 sediment type  $\times$  1 or 3 replicates). Large solid circles and bars represent means and standard 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 ( $\times 3$ ).**

<b>Solution</b>	<b>Manual</b>	<b>Centrifugation</b>	<b>Air</b>	<b>Total</b>
Oil	2	2	2	6
Water	2	2	2	6
Water + Oil	$2 \times 3$	$2 \times 3$	$2 \times 3$	18
NaCl	2	2	2	6
NaCl + Oil	$2 \times 3$	$2 \times 3$	$2 \times 3$	18
NaI	$2 \times 3$	$2 \times 3$	$2 \times 3$	18
<b>Total</b>	<b>24</b>	<b>24</b>	<b>24</b>	<b>72</b>

**Materials**



Sediments

+



Microplastics

+

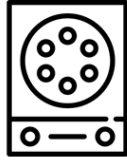


Extracting solutions

**Separation methods**



Air insufflation



Centrifugation



Hand-stirring

**Extraction rate (%)**

