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Efficient microplastics extraction from sand. A cost effective methodology based on sodium iodide recycling

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ABSTRACT

Evaluating the microplastics pollution on the shores requires overcoming the technological and economical challenge of efficient plastic extraction from sand. The recovery of dense microplastics requires the use of NaI solutions, a costly process. The aim of this study is to decrease this cost by recycling the NaI solutions and to determine the impact of NaI storage. For studying the NaI recyclability, the solution density and the salt mass have been monitored during ten life cycles. Density, pH and salt mass have been measured for 40 days to assess the storage effect. The results show that NaI solutions are recyclable without any density alterations with a total loss of 35.9% after the 10 cycles of use. During storage, chemical reactions may appear but are reversible. Consequently, the use of recycling methods allows for a significant cost reduction. How far the plastic extraction by dense solutions is representative is discussed.

1. Introduction

The plastic production has increased exponentially during the second half of the 20th century (PlasticsEurope, 2013). In the same time, due to the lack of waste management plants, the environmental pollution by plastics is rising inexorably (Jambeck et al., 2015). A lot of studies have shown that plastic particles, and more particularly microplastics, could be ingested directly by a large range of organisms, living in aquatic environment, from plankton (Cole et al., 2013; Frias et al., 2014; Setälä et al., 2014) to mammals (Bravo Rebolledo et al., 2013; Denuncio et al., 2011; Secchi and Zarzur, 1999) passing by crustaceans (Murray and Cowie, 2011), molluscs (Van Cauwenberghe et al., 2015), fishes (Lusher et al., 2013) and birds (Ryan, 2008). Organisms which live in sediments, such as lugworms *Arenicola marina* (Green et al., 2016; Van Cauwenberghe et al., 2015) or sandhoppers *Talitrus saltator* (Ugolini et al., 2013), are particularly exposed to microplastics mixed with sand. Microplastics can also be ingested indirectly by feeding on contaminated preys (Farrell and Nelson, 2013). Hence, all the food chain could be involved (Andrady, 2011; Fendall and Sewell, 2009; Lima et al., 2014). Different effects of the plastic ingestion have been highlighted on feeding activity (Azzarello and Vleet, 1987; Sussarellu et al., 2016; Wright et al., 2013), mobility (Rehse et al., 2016) or reproduction (Sussarellu et al., 2016). From this ingestion of plastics arises a major threat for marine life and implies to survey the plastic pollution.

Microplastics pollutions have been observed in various natural environments. They are well documented on the water surface of rivers (Gasperi et al., 2014), lakes (Biginagwa et al., 2016; Eriksen et al., 2013; Free et al., 2014; Imhof et al., 2016), sea (Lusher et al., 2014; Pedrotti et al., 2016) and oceans (Desforges et al., 2014; Shim and Thompson, 2015). They are also present in benthic environments (Koutsodendris et al., 2008; Van Cauwenberghe et al., 2013b) and in shore environments (Hidalgo-Ruz and Thiel, 2013; Imhof et al., 2013; Ng and Obbard, 2006), where they are generally mixed with sand. To recover them, different extraction methods are required. Dense salt solutions are generally used like sodium chloride (density: 1–1.18 g/mL) (Frias et al., 2016; Thompson et al., 2004). However, this method has a low capacity to extract denser microplastics than 1.20 g/mL (Claessens et al., 2013; Imhof et al., 2012; Nuelle et al., 2014). As a consequence, denser solutions have been proposed such as zinc chloride (density: 1–1.81 g/mL) (Imhof et al., 2012; Liebezeit and Dubaish, 2012), sodium polytungstate (density: 1–3.2 g/mL) (Corcoran et al., 2009) or sodium iodide (NaI; density: 1–1.84 g/mL) (Dekiff et al., 2014; Van

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Cauwenberghe et al., 2013a). Nevertheless, these salts are very expensive. Consequently, in the aim to decrease the extraction cost, different pre-treatment methodologies based on water flow extraction have been proposed such as the AIO system (Nuelle et al., 2014) or the elutriation system (Claessens et al., 2013; Kedzierski et al., 2016). Eventually, the extractions need to be refined and sodium iodide is then generally used after the water flow extraction (Claessens et al., 2013; Nuelle et al., 2014). These two works have shown that NaI solution still be an indispensable step to extract microplastics from sand even if this process remains a little expensive. As an alternative, the recycling and the reusing of the NaI could be considered in the aim to decrease the cost of extraction. Furthermore, the use and reuse of this salt involve storing it. Unfortunately, NaI salt is very hygroscopic and unstable in solution (McDonnell and Russell, 1999). As chemical reactions may occur along storage, it is not assessed that stored NaI, as salt or solution, can be efficiently reused to extract plastics from sand.

In the present study, we aim to determine: *i*) if sodium iodide solutions can be recycled without changes in density, *ii*) the impact of the solution recycling on the cost of a microplastics extraction and *iii*) the optimal storage conditions, either as salt or solution.

2. Materials and methods

2.1. Density of different NaI solutions

In the aim to determine the evolution of the density as a function of the NaI weight to weight content, 8 different NaI solutions have been prepared ranging from 11.8 up to 59.4% (*w*/w) in NaI content (solubility limit of NaI salt in water at 25 °C: 64.8%). Sodium iodide salt (Sigma-Aldrich, ≥99.5) is weighed (d = 0.1 mg, Denver Instrument SI-114) and distilled water is added to obtain a solution with a total mass of 50 g. The solution is then manually stirred until salt complete dissolution. Each solution is prepared in triplicate. From these results, the amount of NaI salt necessary to prepare a 1.8 g/mL solution is determined.

The density of NaI solutions is measured by the weighing method. A volume (V) of 2 mL of each preparation is weighed (m_i) and for each preparation, three measurements are performed. Hence each density value is the mean (d_{mean}) of nine (n) mass measurements. The Eq. (1) allows for calculating the solution density:

$$d_{mean} = \frac{\sum_{i=1}^{n} m_i}{V.n} \tag{1}$$

2.2. Recycling of a NaI solution at 1.8 g/mL of density

2.2.1. Kinetics of evaporation of a pure NaI solution

The recycling process of NaI salt proposed in this work is based on water evaporation. In a first time, two NaI solutions at 1.80 g/mL and 1.19 g/mL of density, corresponding respectively to the densities of the solutions recovered without and with a rinsing step, have been prepared, in a 250 mL beaker (final volume of 28 mL and 128 mL, respectively). Each solution is prepared in triplicate (Table 1). These experimental conditions are identical at those used for recycling NaI salt. Then, these solutions are heated at 130 ± 5 °C, boiling temperature of a 1.80 g/mL density NaI solution. Every 10 min up to complete evaporation of water, the solution is weighed. This experiment has allowed us to determine the time required for evaporating the water under these two different experimental conditions.

Table 1
Composition of the different NaI solutions used for the evaluation of the evaporation time.

	NaI mass (g)	Water mass (g)	$W_{NaI}/W_{tot.}$ (%)
Experiment 1 $\boldsymbol{d_{mean}} = 1.80\boldsymbol{g/mL}$ $V_{solution} = 28\ mL$	30.05 ± 0.04	20.09 ± 0.08	59.93 ± 0.12
Experiment 2 $\boldsymbol{d_{mean}} = 1.19\boldsymbol{g/mL}$ $V_{solution} = 128\ mL$	30.02 ± 0.01	120.90 ± 0.86	19.89 ± 0.11

2.2.2. Recycling process

Three experiments have been performed in the aim to test if the sodium iodide could be reused up to ten plastic extractions (Fig. 1). The density of each initial NaI solution is 1.80 g/mL. For the first experiment, the use of a pure NaI solution is performed by dissolving 30 g of NaI with 20 mL of water (Table 2) and boiling it at 130 °C for 1 h. After weighing the sample, a new cycle is operated by adding water to obtain a 60% (*w*/w) NaI solution and so forth until the ten cycles are achieved.

In the second experiment, the impact of marine sediments on the recycling is tested. The sediment used has been sampled from the foreshore of the Kernevel beach (GPS coordinates: 47.7173, −3.3666; Lorient, France) and dried in an oven (Memmert) at 60 °C during three weeks. For the first cycle, a volume of sand of 15 mL is added to the NaI solutions (28 mL). Then, sand and solution are manually stirred during 10 min. This step promotes eventual NaCl and organic matter dissolution, impurities that may have a significant impact on the NaI solution density of the next cycle. The solution is then filtered through a filter funnel (diameter: 120 mm, porosity 4) then the solution is evaporated during 1 h, the mass is measured and the sand used is thrown away. A second cycle is then operated. Due to NaI salt loss during the recycling cycle, the volume of sand added is, from the second cycle, 0.5 times the NaI solution volume.

In the third experiment, the sediment is rinsed with 100 mL of distilled water during the filtration step. As a consequence, the total volume of the solution reached ranges between 100 and 130 mL and 3 h are necessary to obtain a total evaporation. After ten cycles, impurities related to the sand (NaCl, organic matter) may have contaminated the NaI salt. Accordingly, the purity of the NaI salt after ten cycles has been checked by using a Scanning Electron Microscope (SEM; JEOL 6460-LV) coupled with Energy-dispersive X-ray spectroscopy (EDS; Oxford Instrument X-ACT SATW 10mm^2).

2.3. Evaluation of the extraction cost

A 250 g sample of sediment has been sieved on a sieve set as in the protocol proposed by Kedzierski et al., 2016. The residues were then weighed to determine a realistic granulometric distribution of the sand. The mass of the different subsamples, based on the sieve size ranges, is then calculated for a theoretical 1 kg sample of sand. If the subsample mass is higher than 20 g, the subsample needs to be elutriated. This technic has shown its capability to remove between 91 and 100% of the sand and a high yield of plastic extraction (>90%) (Kedzierski et al., 2016). Here, the calculation is based on the hypothesis that 90% of the sand is removed. Then plastic extraction needs to be refined with sodium iodide solution (d = 1.80 g/mL). The price of the NaI salt is 215 euros for 500 g (Sigma Aldrich, 17-05-2016). A mass of 30 g of NaI with 20 g of water allows creating a 28 mL solution. The volume of NaI solution required for extraction is twice the sand volume and the calculation takes into account the salt loss during the recycling phase. From these considerations, the extraction cost per kilogram has been calculated for three different protocols:

- an extraction only based on sodium iodide;
- an elutriation and a refined with sodium iodide;
- an elutriation and a refined with sodium iodide with a recycling step.