Treatment of contaminated marine sediments by sonolysis

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Abstract

The sediments of the port areas are often affected by the presence of several contaminants, which are generated from anthropic activities. These compounds can be toxic for the human health, thus, also to ensure a correct management and avoid the disposal in landfills, the sediments need to be treated. A novel approach proposal by international literature for sediments remediation is the use of advanced oxidation processes, which include ultrasonic technologies. Ultrasounds could be applied mainly to mineralize organic compounds and to desorb inorganic compounds through chemical and mechanical effects, respectively. The present paper is focused on the effectiveness of sonication to abate organic contaminants. The tests were carried out by ultrasound bath at different times of the treatment and sonication frequencies. The treatment efficiency was evaluated in terms of removal percentage. High percentages of degradation were achieved during the treatments in all the tests. The results demonstrated that ultrasound technology is a possible alternative technology for the treatment of contaminated sediments.

Keywords: ultrasounds, sediments, organic contaminants, AOPs.

1. Introduction

The port areas are often subject to intense maritime traffic. These sea-based activities with other anthropogenic activities focus in these areas, could release hazardous substances in the sea (Tornero and Hanke 2016; Tournadre 2014). Among these substances there are a lot of contaminants as heavy metals, polycyclic aromatic hydrocarbons and pesticides (El Nemr and El Sadaawy 2016; Liu *et al*., 2015; Cannarsa *et al*., 2014; Marini and Frapiccini 2013), which are toxic or potentially toxic for aquatic organisms, ecosystem and the human health (Zhang *et al*., 2017; El Nemr *et al*., 2016). These substances can deposit at the bottom of the sea and can accumulate in the sediments (Neira *et al*., 2017). However, varying the physicochemical parameters the contaminants can return to in suspension (Liu *et al*., 2017). The sediments which are in the port areas can be, therefore, potentially contaminated. Each year, because of the dredging operations usually carried out to ensure the navigability or to implement the reclamation activities, the sediments are brought ashore in large quantities. In order to avoid the landfill disposal it is necessary a treatment of this matrix to reduce the contaminants content and to allow a possible reuse. In the scientific literature there are many treatments used to reduce the contaminants from the solid matrices like sediments. Among the most innovative treatments there are those who exploit the advanced oxidation processes (AOPs), which include also the ultrasound processes. Several studies concerning the application of the ultrasound waves demonstrate their efficiency for the treatment of various matrices. In these studies the ultrasounds are widely used to remove emerging contaminants from the wastewater, as unique treatment (Naddeo *et al*., 2013) or in combination with other treatments (Prado *et al*., 2017; Naddeo *et al*., 2015; Secondes *et al*., 2014; Cesaro *et al*., 2013). Ultrasounds are also used as pre-treatment of organic waste for energy production (Cesaro *et al*., 2014) and to remove different compound from the soil or sediments (Chen *et al*., 2016; Pee *et al*., 2015). Ultrasonic treatments efficacy is based on the cavitation phenomenon, which concerns the formation, growth and collapse of vapor bubbles, causing locally high variation of pressure and temperature (Thompson and Doraiswamy 1999; Mason *et al*., 1997). Chemical and mechanical effects are the consequence of this phenomenon. In particular mechanical effects led to fragmentation of the sediments (Son *et al*., 2012), whereas chemical effects involve the degradation of the organic compounds through the action of the hydroxyl radicals (Suslick, 1989). In the present study it was evaluated the ability of ultrasound technology to reduce the content of organic contaminants from the sediments.

2. Materials and methods

2.1. Materials

Fine sand was selected as model of sediment and the particle size distribution was analyzed according to ASTM D422-63. Sediments were spiked with a solution of polycyclic aromatic hydrocarbons (PAHs) (Benzo(α)pyrene (B(α)P), Benzo(α)anthracene (B(α)A)) in order to achieved the double value of the concentration for soil and sediments established from the Italian limit law (D.Lgs. 152/06). All contaminants used were purchased

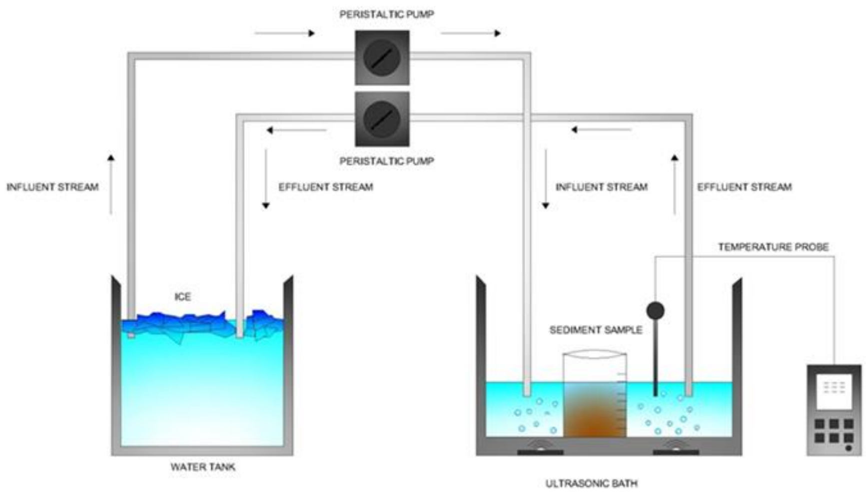

Figure 1. Experimental set-up

from Sigma Aldrich. Nominal solution was prepared for the organic contaminants using dichloromethane (VWR CHEMICALS) according to Russo *et al*. (2012). To determinate the contaminant concentration achieved it was performed the extraction in accordance with the methods US EPA 3550b. The determination was carried out by the gas chromatograph coupled with a mass selective detector (GC-MS Thermofinningan DSQ Trace) (US EPA 8270). The concentration achieved for B(α)A and B(α)P was of 17,05 (±1.95) mg/kg_{ss} and 17.50 (±2.35) mg/kg_{ss}, respectively. After the treatment also the water of the sample was analyzed for checking the possible presence of contaminants. For water it was used the liquid-liquid extraction method US EPA 3510 and the determination was carried out by GC-MS. The tests and analytical determinations were repeated to evaluate the reproducibility of the results. The outcomes shown are in terms of average values.

2.2 Experimental set-up and procedure

Ultrasound treatment was carried out using an ultrasonic bath (Elma TI-H 10) at 200W. The tests were implemented at times from 5 to 60 minutes for two frequencies, a lower frequency of 35 kHz and a higher frequency of 130 kHz. To define the work conditions reference was made to previous studies (Pee *et al*., 2015; Collings *et al*., 2006; Mason *et al*., 2004). To enable to apply ultrasound waves, the sample was prepared with 20 gr of contaminated sand and 40 ml of deionized water (w/w 1:2) (Shrestha *et al*., 2009) in a beaker of 250 ml. The sample was placed into ultrasonic bath at an ultrasonic density of 110 W/L (Fig.1). To avoid overheating of the sample, it was realized a cold water recirculation with two peristaltic pumps (323 S/D Watson-Marlow, UK).

3. Results and discussion

Ultrasound treatment was used to remove contaminants from the sediments, the results were represented in terms of removal percentage. The removal of PAHs has reached peaks of 93% with a slight difference of removal for B(α)A (Fig.2a) and B(α)P (Fig.2b). Higher removal efficiency was observed for all experiments, however, the variation of the treatment time was not significant in accordance with the previous study of Shrestha *et al*. (2009). Also the variation of the frequency of sonication has not shown important changes. Already after 5 minute of treatment and at a frequency of 35 kHz, it was reached a range of removal percentage between 83% and 89%, respectively for B(α)A and B(α)P. This outcome highlights that is possible to obtain good results in a brief time and with a low frequency. To evaluate the possible transition of contaminants from the sediments to the water of the sample, the analyses were carried out also for this matrix. In each sample it was founded a concentration of PAHs not significant. This outcome has confirmed the mineralization of organic contaminants and then the prevalent chemical effect caused by the ultrasounds on this compounds.

4. Conclusions

In this paper ultrasounds were used to treat contaminated sediments in order to reduce the content of organic contaminants. Ultrasounds were applied mainly to mineralize organic compounds through chemical effects. The tests were carried out in batch at two frequencies of sonication and five times of treatment. The results showed that the variation of these work parameters were little significant for the reduction of the contaminants. Indeed, the treatment achieved a high percentage of removal of 89%, already at 5 minute of treatment and at the lower frequency of sonication, 35 kHz. In particular the highest degradation percentage was of 93% for B(α)P. The analyses carried out on the water of samples, have highlighted that during the treatment occurred the degradation of the organic contaminants. Ultrasound treatment was able to obtain high percentage of organic contaminants removal without chemical solvent and in reduced times. The outcomes achieved in this work showed that the ultrasound treatment is potentially

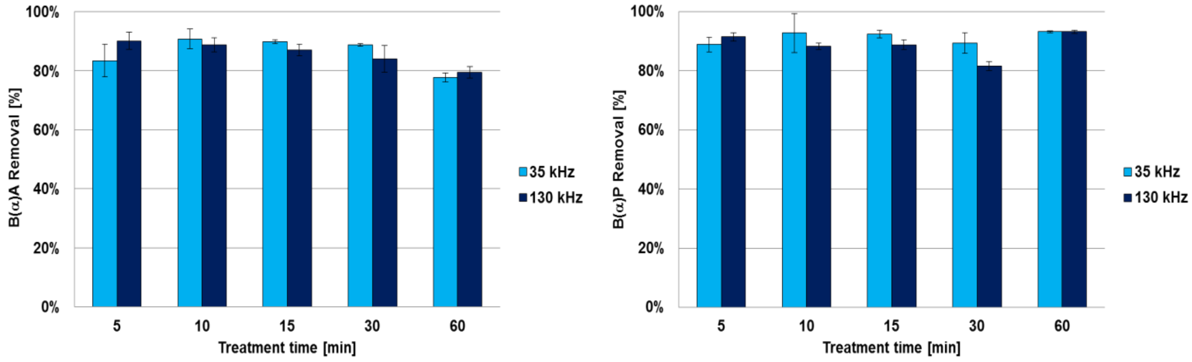

Figure 2. Removal percentage of Benzo(α)anthracene (a) and Benzo(α)pyrene (b).

effective in order to reduce organic compounds in the sediments.

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