### **ARTICLE IN PRESS**

#### Marine Pollution Bulletin xxx (2014) xxx-xxx

Contents lists available at ScienceDirect



## Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

## A pilot study on remediation of sediments enriched by oyster farming wastes using granulated coal ash

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#### ARTICLE INFO

Article history: Available online xxxx

Keywords: Hydrogen sulfide Benthic environment Remediation Coal ash

#### ABSTRACT

In order to evaluate the ability of granulated coal ash (GCA), a byproduct of coal thermal electric power stations, to remove hydrogen sulfide from organically enriched sediments, a pilot study was carried out at oyster farming sites, where sediments were enriched with oyster feces and dead oysters. Concentration of hydrogen sulfide in the interstitial water of the sediment decreased to nearly zero in both experimental sites, whereas it remained over 0.2 mg/l in the control site. Concentration of acid volatile sulfide (AVS) in the sediment also decreased significantly in both experimental sites, while remained over 0.4 mg/g in the control site. Increases were observed in both the number of benthic microalgae species and the individual number of benthic animals in the surface sediments. This may have been due to the decrease in hydrogen sulfide.

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#### 1. Introduction

Deterioration of sediment quality in Hiroshima Bay is a serious problem due to the deposit of organic waste from intensive oyster farming. In such organically enriched sediment, hydrogen sulfide is produced by sulfate reduction (Richard and Morse, 2005). Hydrogen sulfide is highly toxic and fatal to living organisms; it also consumes oxygen when it is oxidized. Acid-volatile sulfide (AVS) is a sedimentary sulfur pools that generate gaseous hydrogen sulfide with the addition of 1 N HCl or a related acidification procedure (Richard and Morse, 2005). According to the Standard for Fisheries Water Quality in Japan (Japan Fishery Resource Conservation Association, 2000), AVS in sediment should be less than 0.2 mg/g to maintain a healthy environment for all fish species. It is thus important to find ways to make the cultivation of oysters sustainable by restoring deteriorated sediments and preserving healthy ecosystems in the area. Several articles have tried to develop techniques for remediating deteriorated sediments using recycled materials such as iron slags (Yamada et al., 1987; Johansson and Gustafsson, 2000), clam shells (Kwon et al., 2004; Asaoka et al., 2009a,b; Yamamoto et al., 2012), marine organisms (Vezzulli et al., 2004; Huang et al., 2013; Yamamoto et al., 2008).

Coal ash is a byproduct of coal combustion processes in thermal electric power stations; 11.6 Mt of coal ash were produced in Japan

http://dx.doi.org/10.1016/j.marpolbul.2014.11.022 0025-326X/© 2014 Elsevier Ltd. All rights reserved. in 2007 (Japan Coal Energy Center). Granulated coal ash is produced by mixing fly ash with cement as a binder. Our group recently developed a new technology using granulated coal ash (GCA) to decrease the concentrations of hydrogen sulfide in sediments (Asaoka et al., 2009a,b, 2012).

In the present study, we evaluated the efficiency of GCA in reducing hydrogen sulfide in deteriorated coastal marine sediments below hanging oyster rafts.

#### 2. Materials and methods

#### 2.1. Granulated coal ash (GCA)

Two types of GCA were used in this study. For the first, 10–15% cement was mixed with pulverized fly ash produced at the Shin-Onoda coal power plant (Onoda GCA); the second was composed simply of pressurized fluidized bed combustion ash from the Osaki coal power plant (Osaki GCA).

Both GCAs, which were analyzed with a fluorescent X-ray analyzer ZSX100e (Rigaku Co. Ltd.), were mainly composed of SiO<sub>2</sub>,  $Al_2O_3$ , CaO and Fe<sub>2</sub>O<sub>3</sub> with quartz and aluminosilicate in the crystal phase (Fig. 1) (Yamamoto et al., 2013). It has been reported that environmentally regulated elements (heavy metals) were not dissolved from the GCAs used in this study and were below the standard levels set for the soil pollution environmental criteria in Japan (Asaoka et al., 2008).

Please cite this article in press as: Yamamoto, T., et al. A pilot study on remediation of sediments enriched by oyster farming wastes using granulated coal ash. Mar. Pollut. Bull. (2014), http://dx.doi.org/10.1016/j.marpolbul.2014.11.022

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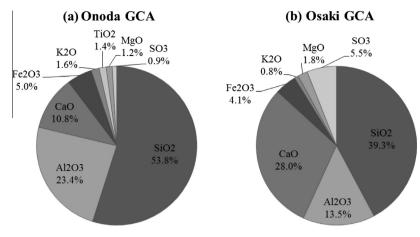


Fig. 1. Chemical composition of GCAs. (a) Onoda GCA and (b) Osaki GCA.

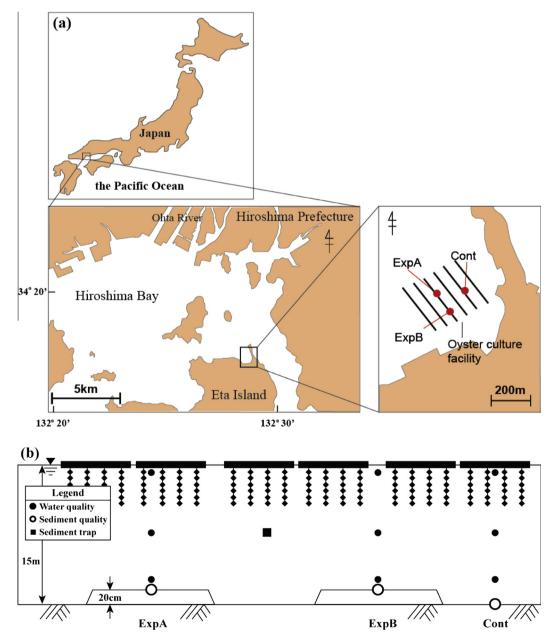


Fig. 2. Map showing the northern part of Hiroshima Bay and Kirikushi Port where the field experimental study was carried out.

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