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Suppression of phosphate release from coastal sediments using granulated coal ash

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ABSTRACT

In order to evaluate suppression of phosphate release from sea bottom sediments using granulated coal ash (GCA), a byproduct from coal thermal electric power stations, field experiments were carried out at oyster culture sites where the sediments are rich in organic matter. Two types of GCA with the diameter of <40 mm were applied on separate bottom sediment plots of 50 m × 75 m with ca. 20 cm depth each. The first GCA type is composed of pulverized coal combustion ash (ExpA) and another consists of pressurized fluidized bed combustion ash (ExpB). Sediment pH was increased up to ca. 8.5 in both experimental plots due to hydrolysis of CaO. Phosphate concentration in the interstitial water of the sediment was significantly decreased in ExpA due to adsorption onto the GCA. A numerical model to quantify chemical and biological processes in the sediment was applied to estimate how phosphorus cycle was changed due to the application of GCA. The model output showed that release rate of phosphate from the sediment at ExpB was suppressed to the level of about 1 order lower (400 μmol m^{-3} d^{-1}) than that in the control site (4800 μmol m^{-3} d^{-1}), whereas decomposition of detrital phosphorus in the experimental site was estimated to have been enhanced 4.8 times compared to the control site. Considering both laboratory and simulated field experiments carried out previously, it was proven in the present study that GCA can effectively adsorb phosphate from interstitial water in organically enriched sediments and suppress the release of phosphate from the sediments to the upper water column in the field experiments.

1. Introduction

According to the Hiroshima City Fisheries Promotion Center (2009), 17,497 *t* of oyster meat were produced from Hiroshima Prefecture in 2009. Deterioration of sediment quality in Hiroshima Bay is a serious problem due to deposition of organic waste from the intensively cultured oysters. Decomposition of organic matter in the surface sediments leads to formation of oxygen deficient water in the bottom water. Benthic phosphorus release, which exceeds terrigenous total phosphorus (TP) loads, may be the major factor for eutrophication of Hiroshima Bay. Since the release of phosphate is enhanced under anaerobic conditions in warm thermally-stratified seasons, it should be a target for control and suppression (Lee and Hoshika, 2000; Kittiwanich et al., 2007).

Coal ash is a byproduct from coal thermal electric power stations, with 11 Mt produced in Japan in 2005 (Japan Coal Energy Center, 2009). Granulated coal ash (GCA) used in this study is produced by mixing fly ash with cement as a binder. The GCA has usually been used as materials for road construction and coarse aggregates for concrete. In addition to these, along with the strategy to contribute towards promoting recycling consciousness and waste reduction within the society, utilization of GCA in the other fields are being explored.

Our previous studies have proven that GCA can effectively remediate sea bottom sediments through these different experimental mechanisms (Asaoka et al., 2009a,b): (1) neutralization of acidified sediments due to hydrolysis of CaO; (2) adsorption of nutrients and; (3) of hydrogen sulfide, and; (4) resulting decrease of oxygen consumption. In the present study, we aim at evaluating the efficiency of GCA on the suppression of phosphate release from organically enriched sediments in a field experiment by applying a numerical model to quantify chemical and biological processes in the sediment.

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2. Materials and methods

2.1. Granulated coal ash (GCA)

The GCA is a commercially sold product named ‘Hi-beads’ (Energia Eco Materia Co., Inc., Japan). Two types of GCA were used in this study. One is granulated by adding ca. 10–15% cement as a binder to pulverized coal combustion fly ash produced from Shin-Onoda coal power plant, and the other one is composed of pressurized fluidized bed combustion ash produced from Osaki coal power plant of Chugoku Electric Co., Japan.

Both GCAs are mainly composed of SiO_2, Al_2O_3, CaO and Fe_2O_3 with quartz and aluminosilicate crystal phase (Table 1). It was reported that environmentally regulated elements (heavy metals) are 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).

2.2. Field experiments

Field experiments were carried out off Kirikushi Port at the northern coast of Etajima Island located in the northern part of Hiroshima Bay, Japan where intensive oyster cultures are conducted (Fig. 1(a)). Two types of GCA were used to cover the muddy sediments below the oyster culture rafts on Oct. 30, 2008, and monitoring of water and sediment conditions were carried out. At the experimental sites, the plots with GCAs were ca. 20 cm depth and areal dimensions of 75 × 50 m (Fig. 1(b)). The GCAs from Shin-Onoda and Osaki were used in ExpA and ExpB, respectively. The two experimental sites stand 80 m apart to provide reasonably uniform environmental conditions there. Control site (Cont) without any treatments was also set beside them as a reference site.

Observations for physical, chemical and biological conditions were carried out in February, May, August and November 2009 in addition to the initial observation made in October 2008. Vertical profiles of temperature, salinity, pH and concentration of dissolved oxygen were measured using a Multiparameter Sonde (YSI-model 6600, YSI Inc.). Water samples were collected from the surface (0 m) and the middle of the water column using a Van-Dorn water sampler, and 1 m above the bottom without unduly disturbing the bottom sediments by divers. Water sample of 300 ml was filtered through a 0.45 μm pore size hydrophilic PVDF filter with a diameter of 47 mm (MILLEX: Millipore) to separate particulate matter and solution. Precipitate collected on the filter was used for determination of particulate phosphorus (PP). The solution was used for analyses of dissolved inorganic phosphorus (DIP) and dissolved total phosphorus (DTP). The analytical methods are described below. The data sets of phosphorus obtained from 1 m above the bottom were used as boundary data for calculations (see the description below).

Table 1
Chemical composition of the granulated coal ash (GCA) used in the present study. The GCAs produced from Shin-Onoda and Osaki were applied to ExpA and ExpB, respectively.

Chemical components	Contents (%)	
	GCA from Shin Onoda	GCA from Osaki
SiO_2	53.8	39.3
Al_2O_3	23.4	13.5
CaO	10.8	28.0
Fe_2O_3	5.0	4.08
K_2O	1.58	0.76
TiO_2	1.40	No data
MgO	1.19	1.84
SO_3	0.879	5.54

Interstitial water of the sediment was collected by centrifugation (3000 rpm, 20 min), and concentrations of DIP were determined by the standard method (American Public Health Association et al., 1989) using an auto analyzer (SWATT, BLTEC). Concentration of DTP and PP were also determined as DIP using the auto analyzer after wet degradation with hyperchloride-nitric acid following Japanese standard methods (Ministry of the Environment, 2001). Dissolved organic phosphorus (DOP) was calculated by subtracting the DIP value from the DTP value.

2.3. Numerical experiments

2.3.1. Model framework

A numerical model was constructed to evaluate the suppression efficiency of phosphate release by the GCA as shown in Fig. 2. Calculation was conducted for the surface sediment layer of 20 cm thickness, and the overlying water of 100 cm thickness and the consolidated sediment layer below the surface 20 cm sediment layer were used as boundary layers. Calculation was conducted for both the experimental sites and the control site. The model was developed using the software STELLA 9.0.3J (isee systems, inc.).

2.3.2. Ecological process

To express phosphorus cycles in and around the sediment–water interface, 6 compartments were set to represent various forms of phosphorus in the model: dissolved inorganic phosphorus (DIP), dissolved organic phosphorus (DOP), benthic microalgal phosphorus (BMA), particulate phosphorus (detritus and minerals) excluding BMA (DetP), filter-feeder benthos phosphorus (FIB), and detritus-feeder benthos phosphorus (DEB).

$$\begin{aligned}\frac{\mathrm{dDetP}}{\mathrm{d}t} = {} & \mathrm{SF}[w\mathrm{DetP}] - \mathrm{HF}\left[\mathrm{DetP}, s\mathrm{DetP}\right] + \mathrm{MF}[\mathrm{BMA}] + \mathrm{MF}[\mathrm{FIB}] \\ & + \mathrm{EF}[\mathrm{FIB}] + \mathrm{MF}[\mathrm{DEB}] + \mathrm{EF}[\mathrm{DEB}] - \mathrm{IF}[\mathrm{DEB}]_{\mathrm{DetP}} \\ & - \mathrm{KF}[\mathrm{DetP}]_{\mathrm{DOP}} - \mathrm{KF}[\mathrm{DetP}]_{\mathrm{DIP}} \end{aligned} \quad (1)$$

$$\begin{aligned}\frac{\mathrm{dDIP}}{\mathrm{d}t} = {} & -\mathrm{DF}[w\mathrm{DIP}] - \mathrm{DF}[s\mathrm{DIP}] + \mathrm{KF}[\mathrm{DetP}] + \mathrm{EF}[\mathrm{FIB}] + \mathrm{EF}[\mathrm{DEB}] \\ & - \mathrm{VF}[\mathrm{BMA}] - \mathrm{AF}[\mathrm{DIP}] \end{aligned} \quad (2)$$

$$\frac{\mathrm{dDOP}}{\mathrm{d}t} = -\mathrm{DF}[w\mathrm{DOP}] - \mathrm{DF}[s\mathrm{DOP}] - \mathrm{KF}[\mathrm{DOP}]_{\mathrm{DIP}} + \mathrm{KF}[\mathrm{DetP}]_{\mathrm{DOP}} \quad (3)$$

$$\frac{\mathrm{dBMA}}{\mathrm{d}t} = \mathrm{VF}[\mathrm{BMA}] - \mathrm{MF}[\mathrm{BMA}] - \mathrm{IF}[\mathrm{FIB}]_{\mathrm{BMA}} - \mathrm{IF}[\mathrm{DEB}]_{\mathrm{BMA}} \quad (4)$$

$$\frac{\mathrm{dFIB}}{\mathrm{d}t} = \mathrm{VF}[\mathrm{FIB}] - \mathrm{MF}[\mathrm{FIB}] - \mathrm{UF}[\mathrm{FIB}] - \mathrm{EF}[\mathrm{FIB}] \quad (5)$$

$$\frac{\mathrm{dDEB}}{\mathrm{d}t} = \mathrm{VF}[\mathrm{DEB}] - \mathrm{MF}[DEB] - \mathrm{UF}[\mathrm{DEB}] - \mathrm{EF}[\mathrm{DEB}] \quad (6)$$

where, wDetP and sDetP are DetP in the overlying water and the consolidated sediments, respectively, SF is sinking rate, DF is diffusivity, HF is deposition rate, KF is decomposition rate, MF is mortality rate, EF is excretion rate, UF is urination rate, IF is ingestion rate and AF is adsorption rate on GCA.