Blast furnace slag can effectively remediate coastal marine sediments affected by organic enrichment

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Abstract

There is an urgent need to control nutrient release fluxes from organically-enriched sediments into overlying waters to alleviate the effects of eutrophication. This study aims to characterize blast furnace slag (BFS) and evaluate its remediation performance on organically-enriched sediments in terms of suppressing nutrient fluxes and reducing acid volatile sulfide. BFS was mainly composed of inorganic substances such as CaO, SiO_2, Al_2O_3 and MgO in amorphous crystal phase. Container experiments showed that the phosphate concentration in the overlying water, its releasing flux from sediment and AVS of the sediment decreased by 17-23%, 39% and 16% compared to the control without BFS, respectively. The loss on ignition was significantly decreased by 3.6-11% compared to the control. Thus, the application of BFS to organically-enriched sediment has a suppressive role on organic matter, AVS concentration and phosphate releasing flux from sediments and therefore, is a good candidate as an effective environmental remediation agent.

Key words

acid volatile sulfide, blast furnace slag, eutrophication, marine sediment, phosphate , remediation

1. Introduction

Eutrophication is one of the more serious environmental problems in enclosed water bodies which can lead to massive growth of algae. Subsequent decomposition of dead algal cells causes excessive reduction of dissolved oxygen (DO). Oxygen depletion in the overlying waters is often fatal to benthic organisms.

In Japan, the special law promulgated to reduce terrigenous phosphorus and nitrogen discharges has shown its positive effects in the Seto Inland Sea which is located in the southwestern part of Japan. The Seto Inland Sea is the largest semi-enclosed marine area in Japan with a size of 23,000 km^2 and an average depth of 38 m (Yamamoto, 2003). Total phosphorus (TP) and total nitrogen (TN) loadings into the Seto Inland Sea except in Osaka Bay were reduced by up to 50% of their peaks just after the enactment of the law (Setouchi Net, 2008). As a result, the number of red tides decreased by 1/3 from that recorded during the peak periods. However, the concentrations of TP and TN in the area have constantly remained high for the last 30 years at 0.023-0.026 mg L^{-1} and 0.25-0.28 mg L^{-1}, respectively (Setouchi Net, 2008), despite the legislation in place.

Significant amounts of nutrient released from the sediments have been identified as one of the plausible causes as to why the concentrations of TP and TN have been constantly high despite the reduction of terrigenous nutrient discharges since 1980 for TP and 1996 for TN (Setouchi Net, 2008). The flux of phosphorus from the sediments was estimated to be 4 t d^{-1} during stratified seasons, which was twice higher than that of terrigenous phosphorus loads (Yamamoto et al., 1998). Therefore, it is important for purposes of controlling eutrophication to reduce nutrient release flux from the sediments as well as to cut down on terrigenous nutrient loads.

Furthermore, hydrogen sulfide is formed in organically-enriched sediments through the reduction of sulfate ions by sulfate-reducing bacteria under anoxic conditions (Rickard et al., 2005). The presence of acid volatile sulfide (AVS) indicates the potential for hydrogen sulfide generation. Hydrogen sulfide is highly toxic to living organisms and uses up oxygen when it is oxidized. In the Japanese environmental criterion for sulfide concentration in sediments, 0.2 mg g^{-1}-sediment is the allowable level for maintaining conditions for sustainable aquaculture (Japan Fisheries Resource Conservation Association, 2006). Reported data on AVS concentration from different parts of the Seto Inland Sea revealed that about half of the entire area does not meet the criterion even though the area is the major center of aquaculture activities in Japan (Yamamoto et al., 1997). Thus, to reduce AVS concentration in the sediments is one of the most important considerations to sustain aquaculture while restoring and maintaining healthy aquatic ecosystems.

Iron and steel slag are by-products of iron and steel manufacturing. They are roughly classified into two types: blast furnace slag (BFS) produced from the conversion process of iron ore into pig iron, and steel making slag produced from the purification process of pig iron into steel. In 2005, a total of 38 Mt was generated in Japan with 65% as BFS (Nippon Slag Association, 2006). Commonly, the slag has been used for roadbed construction material, as coarse aggregate for concrete and as raw material for cement (Nippon Slag Association, 2006). In this age of recycling and growing environmental consciousness, it is practical to explore and test new applications for industrial by-products such as BFS.

According to previous reports, BFS shows an adsorption potential for phosphorus from aqueous solutions (Johansson et al., 2000; Oguz, 2004;

Oguz, 2005). However, there is not much information available on the application of BFS for the remediation of organically-enriched sediments except for some fragmentary data (Yamada et al. 1987; Numata et al., 1999; Minato et al., 2006; Minato et al., 2008; Hizon-Fradejas et al. 2009). The purposes of this study are (1) to characterize BFS, and (2) to validate the remediation efficiency of BFS for suppressing nutrient flux and reducing acid volatile sulfide (AVS) of organically-enriched sediments.

2. Materials and methods

2.1. Characterization of BFS

The BFS used in this study was provided by JFE Steel Corporation, Okayama, Japan. Before analyses, the material was dried in an oven at 60^{o}C for 24 h, and ground down using an agate mortar. Chemical composition of the material was determined by X-ray fluorescence (XRF) spectroscopy (ZSX-101e, Rigaku). Glass beads of BFS were made using a mixture of BFS (2.00 g) and $Li_2B_4O_7$:$LiBO_2$=1:4 (4.00 g) and $LiNO_3$ (0.60 g) to be used for the XRF analysis (Fukukawa et al., 2001). Organic carbon and nitrogen were also determined with a CHN analyzer (MT-5, Yanaco) after removing carbonates following the vapor method (Yamamuro et al., 1995). 100 μL of ultrapure water was pipetted into a sample boat containing 25 mg of BFS. The BFS was thoroughly soaked in ultrapure water to completely remove carbonates with 12 mol L^{-1} HCl vapor in a dessicator for 24 h. The BFS was precipitated in 6 mol L^{-1} HCl when the reaction was completely finished. To drive off HCl and water, the samples were then dried at 50°C for 12 h.

The powder X-ray diffraction patterns (XRD) were recorded by a XRD instrument (RINT2500V, Rigaku) using Cu Kα radiation with a scanning speed of 1.7° $\min^{-1}$ at 40 kV, 150 mA.

The specific surface area was determined by the Brunauer-Emmett-Teller method (nitrogen gas adsorption) using a Micromeritics adsorption equipment (ASAP 2000, Micromeritics).

2.2. Dissolution tests of environmentally regulated substances

Environmentally regulated substances dissolved from the BFS were examined following the determination method for soil pollution criteria established by the Ministry of Environment, Japan. The procedure was partially modified as follows: 200 g of the BFS was added to 2 L of filtered surface seawater (pH 7.7), which was collected from the waters off Ehime Prefecture, and filtered through a glass fiber filter (GF/C, Whatman), and stirred for 6 h using a magnetic stirrer at a speed of 100 rpm, 20°C. Ultra pure water was used as solvent for boron (B) and fluorine (F) instead of filtered seawater, because both elements are present in significant amounts in seawater. After stirring, the seawater was filtered through a 0.45 μm membrane filter (HA, Millipore). The filtrates were desalinated and concentrated except As, B, Cr^{6+}, CN, F, Hg and Se using a solid phase extraction column packed with chelating resin (InertSep ME-1, GL Science). Heavy metals in the concentrated filtrates were determined by ICP-AES (Optima 3000, Perkin Elmer). As and Se in the filtrates were determined by hydride generation AAS (APHA, 1989) using an atomic adsorption spectrometer (220FS, Varian), while B and Cr^{6+} in the filtrates were determined by ICP-AES (Vista MPX, Varian) followed by JIS K 0102 Japanese standard method (Namiki, 1982). Concentrations of CN in the filtrates were determined by 4-pyridinecarboxylic acid-pyrazolone adsorption spectrophotometry followed by JIS K 0102 Japanese standard method

(Namiki, 1982). Concentrations of F in the filtrates were determined by Lanthan-Alizarin complexone adsorption spectrophotometry (APHA, 1989). Concentrations of Hg in the filtrates were determined by cold vapor atomic adsorption spectrometry (APHA, 1989) using a Mercury analyzer (RA-3, Nic).

2.3. Container experiments

2.3.1 Experimental settings

The container experiment was conducted from June 6 to July 9, 2007. The schematic diagrams of container experimental set-up designed for simulating enclosed water bodies are shown in Fig. 1. The BFS and sediments (described below) were mixed in round-shaped black polyethylene containers (ϕ: 550 mm, h: 420 mm), and sand-filtered seawater was supplied and allowed to overflow at an exchange rate of 0.7 d^{-1}, which is the average exchange rate of seawater between the upper and the lower layers of the water column in the northern part of Hiroshima Bay (Ogawa, 2002). These containers were placed in a water bath (1000 L FRP container) to prevent rapid change in the water temperature during the experimental period.

The sediment was collected using a suction machine from the Ohko Inlet located at the northern end of Hiroshima Bay with an area of 0.047 km^2 and an average depth of 4 m. In Ohko Inlet, municipal discharge from a wastewater treatment plant is considerable and therefore, blue tides have been frequently observed. Debris and pebbles were manually removed from the sediments before use. Thereafter, 50 L of sediment and 13 L of BFS were mixed in the experimental container, while 50 L of sediment without BFS was used as a control. These same settings were prepared in triplicates.

2.3.2 Sediment analyses

Sediment was sampled into a small plastic container (ϕ:77 mm, h: 47 mm), and pH and oxidation and reduction potential (ORP) were measured immediately by electrodes (PRN-40, Fujiwara, RM-12P, TOA DK). Thereafter, the container was sealed tightly and transported to the laboratory while kept in a cool and dark condition. In the laboratory, AVS was measured with a Hedrotech-S kit: 2 mL of 9 mol L^{-1} H_2SO_4 was pipetted to the sediment sample (ca. 0.5 g) placed in a tube to generate H_2S, and which was determined with a detection tube (Arakawa, 1980). Loss on ignition (LOI) was also determined by combusting the sediment at 700^{o}C for 5 h after determining its water content. Organic carbon and nitrogen were analyzed with a CHNS analyzer (CHNS/0 2400II, Perkin Elmer) after removing carbonates by acidification with HCl (Yamamuro et al., 1995) and dried at 60^{o}C for 12 h. Concentration of total phosphorus in the sediment was determined using the ascorbic acid reducing molybdenum blue method after wet degradation with hyperchloride-nitric acid following Japanese standard methods (The Ministry of Environment, 2001).

2.3.3 Pore water analyses

The sediment was centrifuged at 3,500 rpm for 10 min to collect pore water from the sediment. The supernatant was filtered through a 0.45 μm membrane filter (MILLEX-HV, Millipore) and concentrations of PO_4-P, NH_4-N, NO_2-N and NO_3-N in the filtrate were determined by ascorbic acid reduction molybdate blue adsorption spectrophotometry, indophenol blue method, N-(1-naphthyl)-ethylenediamine adsorption spectrophotometry and Cd-Cu reduction N- (1-naphthyl)- ethylenediamine adsorption spectrophotometry, respectively, using an auto-analyzer (SWATT, BLTEC).

2.3.4 Overlying seawater analyses

Temperature, salinity and pH of the overlying seawater were measured with a multiple electrode (U-10, Horiba). The overlying water was also sampled from 5 cm above the sediment surface by siphoning to determine dissolved oxygen (DO) concentration and nutrient concentration. Dissolved oxygen (DO) was determined by Winkler method (APHA, 1989) immediately after the sampling. The sample aliquot for nutrient analyses was filtered through a 0.45 μm membrane filter (MILLEX-HV, Millipore) and transported to the laboratory while kept under cool and dark condition. Concentrations of PO_4-P, NH_4-N, NO_2-N, NO_3-N were determined by the same method as described above using an auto analyzer (SWATT, BLTEC).

3. Results and discussion

3.1 Characterization of BFS

The chemical composition of the BFS used in this study is shown in Table 1. The BFS is mainly composed of inorganic compounds such as CaO, SiO_2, Al_2O_3 and MgO. The contents of organic carbon and nitrogen were below the detection limits (<0.02 %). Sediments accumulated on the bottom of enclosed water bodies contain dense organic matter due to significant terrigenous loads and eutrophication. Thus mixing the BFS, which is mainly composed of inorganic substances, with organically-enriched sediment is effective in decreasing sediment organic matter content mainly by dilution.

Concentrations of trace elements in the BFS were lower than those of averages of marine sediments in Japan. For example, the average concentration of Cu, Zn and V in Japanese marine sediments are 19.7, 89.25, 66.9 mg kg^{-1}, respectively (Fukue et al., 1999). The analytical results proved that applying the BFS to organically-enriched sediments is environmentally safe in terms of heavy metal content even if BFS is a by-product from the steel industry.

The powder X-ray diffraction pattern of the BFS indicated two small peaks at 29.4°and 52.0° (2θ) which were attributed to Gehlenite ($Ca_2Al_2SiO_7$) (JCPDS, 1990). However, since diffraction intensities of these peaks were very weak, the BFS used in this study was mainly composed of amorphous phase. The specific surface area of the BFS used in this study was 0.2 m^2 g^{-1}.

3.2 Dissolution of environmentally regulated substances from BFS

Environmentally regulated substances dissolved from the BFS used in this study are shown in Table 2. As, CN, Co, Cr^{6+}, Se were not dissolved from the BFS. Some elements were detected but their concentrations were 0.006-0.2 times lower than those set by the Japanese environmental criteria for soil pollution control. Thus, BFS components occur in minute amounts and comply with the set maxima for soil pollution environmental criteria in Japan.

3.3 Container experiments

3.3.1 Changes in the sediment quality

The sediment pH in the BFS container was higher than that of the control container after Day 10 with a statistical difference ($p<0.05$) on Day 18 (Fig. 2). The increase of pH in the BFS container is due to the dissolution of $CaCO_3$ from the BFS (Vdović, 2006; Yu et al., 2008). The chemical reactions involving the dissolution of $CaCO_3$ that generates alkalinity and consumes protons are shown in Eqs. 1, 2 (Mayes et al., in press).

$$CaCO_3 + 2H^+ \rightarrow Ca^{2+} + H_2O + CO_2 \quad (1)$$

$$CaCO_3 + H_2CO_3 \rightarrow Ca^{2+} + 2HCO_3^- \quad (2)$$

Change in sediment ORP was insignificant between the BFS containers. Initial ORP value was -280 mV and then declining rapidly down to -375 to -395 mV after the experiments started. The possible cause of the high initial ORP values can be attributed to the experimental mixing of the sediments at the start when oxygen was inadvertently introduced into the sediments causing a high initial ORP value.

The LOI in the BFS container was significantly decreased by 3.6-11% compared to that of the control container ($p<0.05$) on Day 3, which can be interpreted as a decrease of organic matter content in the BFS container (Fig. 3). This observed decrease was also accompanied by decreases in organic carbon, organic nitrogen and total phosphorus at 79 to 67 mg g^{-1}, 4.9 to 4.3 mg g^{-1}, and 1.1 to 1.0 mg g^{-1}, respectively ($p<0.01$-0.05).

AVS in the BFS container decreased by 16% compared to that of the control ($p<0.01$-0.05; Fig. 4). Since the BFS used in this study contains 3.5 g kg^{-1} Fe_2O_3 (Table 1), hydrogen sulfide in the sediment was adsorbed onto the BFS resulting in the formation of iron sulfide (Rickard et al., 2005).

3.3.2 Change in pore water quality

PO_4-P concentration in the pore water did not decrease much in the BFS, which was changing in the range of 51.8 to 65.4 μM. NH_4-N concentration in the pore water of the BFS container was slightly higher than that of the control container on Day 18 (Fig. 5). Total concentration of NO_3-N + NO_2-N in the pore water of the BFS container tended to increase after Day 3 with levels maintained at 12-24% higher than those in the control container (Fig. 6) suggesting that nitrification was maintained in the BFS container. Generally, during decomposition processes, sediment pH decreases due to bacterial respiration as well as from nitrification and the resulting generation of organic acids. Optimal pH for nitrification is reported in the range of 7.5-8.0 (Park et al., 2003; Sarioglu, 2005). Nitrification is pH-sensitive and the rate declines significantly in conditions below pH 6.8 (Yu et al., 2008). The buffer reaction due to dissolution of $CaCO_3$ from the BFS as mentioned above (Eqs. 1, 2) may serve to maintain pH values suitable for nitrification in the BFS container.

3.3.2 Changes in overlying water quality

Temperature, salinity and pH of the overlying water did not show differences between the BFS containers and the control during the experimental periods, ranging 20.8-25.5°C, 2.91-3.07%, 8.3-9.4, respectively.

Difference in DO concentration in the overlying water between the BFS containers and the control was also insignificant and decreased after Day 3 (Fig. 7). Sediment oxygen demand (SOD) in Hiroshima Bay was reported to be mainly affected by biological consumptions and metabolic activities during summer and autumn and by chemical consumption of reducing substances during winter and spring (Seiki et al., 1993). Since the container experiment was carried out in summer, the DO in the containers is considered to be mainly consumed for biological processes such as nitrification. This may be evidenced by the observed increase in concentration of NO_3-N + NO_2-N coupled with decreasing DO in the pore water (Figs. 6, 7).

The concentration of PO_4-P in the overlying water increased in both BFS and control containers after Day 3 (Fig. 8). Sediment ORP was significantly negative (-375 to -395 mV) after Day 3, indicating that the sediment was in a highly reduced condition. Under these conditions, Fe^{3+} is reduced to soluble ferrous iron (Fe^{2+}) and this led to the liberation of P (Mortimer, 1971; Miao et al., 2006). This is why phosphate that was adsorbed into the sediment was released into the overlying water.

However, the concentration of PO_4-P in the overlying water of the BFS containers decreased by 17-23% compared to the control on Day 18 and showed a statistical difference ($p<0.01$) on Day 24 (Fig. 8). This is probably due to the formation of phosphate salts with aluminum, calcium and iron (Oguz, 2005).

We estimated the phosphate releasing flux from the sediment into the overlying water using the average phosphate-P concentration in the seawater (0.9 μmol L^{-1}), the volume of the overlying water (63.4 L) and water exchange rate (0.7 d^{-1}). The total amount of phosphate-P loaded into the experimental container is calculated to be 40 μmol d^{-1}. The amount of phosphate-P in the effluent water from the container is calculated by multiplying the phosphate concentration in the overlying water by the volume of overlying water (63.4 L). The phosphate releasing flux from the sediment is calculated by the difference of phosphate concentrations between the supplied seawater and the effluent, divided by the surface area of the sediment (0.26 m^2). On the basis of the calculation described above, the average phosphate releasing flux from the sediment in the control container is 280 μmol m^{-2} d^{-1}, which is within the range consistent with that of Hiroshima Bay in a stratified period, -130 to 460 μmol m^{-2} d^{-1} (Yamamoto,

2008) whereas that of the BFS container is calculated to be 170 μmol m^{-2} d^{-1}. These indicate that the phosphate releasing flux from the sediment of the BFS container was suppressed 39% compared to that of the control.

The NH_4-N concentrations were not statistically different changing only in the range of 0.6 to 0.9 μM. The concentrations of NO_3-N + NO_2-N showed a sharp decrease during the first three days but there was no statistical difference between the BFS containers and the control (Fig. 9). The sharp decrease in the concentration during the first three days is well correlated to the change in ORP from -280 mV to a highly reduced condition (-375 to -395 mV).

4. Conclusions

The purposes of this study are (1) to characterize BFS, and (2) to validate the remediation efficiency of organically enriched sediments using BFS in terms of suppressing nutrient fluxes and reducing AVS.

The BFS is mainly composed of inorganic elements such as CaO, SiO_2, Al_2O_3 and MgO with an amorphous crystal phase with almost no phosphorus, organic carbon and nitrogen. Elution of environmentally regulated substances from the BFS was obviously below the set maxima for soil pollution environmental criteria in Japan.

The results of container experiments proved that BFS is able to reduce the phosphate concentration in the overlying water by 17-23%. BFS effectively reduced phosphate flux from the sediments by as much as 39% with the AVS content in the sediment cut by 16% compared to the control. The LOI also decreased with the addition of BFS. The sediment pH was maintained within the range favorable for nitrification and other metabolic activities due to the $CaCO_3$ dissolved from the BFS. As such, BFS represents a potentially useful, effective, and environmentally friendly remediation agent.

As described earlier in the introduction, 38 Mt of BFS is generated in Japan every year. Therefore, it is not difficult to provide enough supplies of BFS for the remediation of coastal sediments. As a follow-up study, we plan to apply BFS to organically enriched sediment in situ and monitoring remediation processes of the sediment therein. Integrating the simulation model with field data and parameters collected in this study shall form the basis for proposing optimum strategies such as application dosage. Long-term environmental effects can also be estimated based on the remediation efficiency of the BFS.

This simple and inexpensive technology is a promising strategy for the remediation of organically-enriched sediments commonly encountered in many developing countries and where expensive technologies are generally impractical. The effective and safe application of the BFS can address two environmental solutions at the same time, namely the remediation of organically-enriched sediments and the elimination of costly disposal problems for an otherwise hazardous industrial by-product that might lead to often catastrophic consequences for the environment and human health. Therefore, BFS represents a promising solution towards the restoration of cleaner environments by way of remediation of organically-enriched sediments in enclosed coastal seas. For developing countries, in particular, this represents a revolutionary protocol that requires no significant financial and technological investment while promising a very practical solution to one of their most pressing environmental challenges.

Acknowledgements

The authors would like to acknowledge the Sasagawa Scientific Research Grant (grant 19-703M) from the Japanese Science Society and Association for Sustainable Material Cycles and Management of Hiroshima that supported this work. We also thank JFE Steel Corporation for kindly providing the blast furnace slag for our study. The manuscript was reviewed for clarity by Dr. Lawrence M. Liao of the Graduate School of Biosphere Science, Hiroshima University.

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Table 1 Chemical composition of the blast furnace slag (BFS)

Main compounds (g kg^{-1}-dw)		Trace compounds (mg kg^{-1}-dw)					
CaO	440	Ba	536	V	37.3	Hf	3.6
SiO_2	324	Sr	422	Nb	27.2	Zn	3.5
Al_2O_3	145	Zr	245	Sc	31.6	W	2.2
MgO	43.8	C	200	Th	22.6	Ga	0.5
TiO_2	4.9	P_2O_5	140	Rb	11.9	Pb	0.4
Fe_2O_3	3.5	Ce	98.8	Cr	9.7		
K_2O	3.0	Y	78.6	U	8.8		
MnO	2.7	La	53.1	Yb	5.8		
Na_2O	1.9	Nd	42.5	Cu	3.6		

Table 2 Environmentally regulated substances eluted from the blast furnace slag (BFS)

	Av (μg L^{-1})	SD(n=3)	RSD (%)	Criteria(<μg L^{-1})
As	<5	-	-	10
B	13	12	92	1000
Cd	0.06	0.0098	16	10
CN	<100	-	-	ND
Co	<0.02	-	-	-
Cu	0.4	0.0298	7.5	-
Cr^{6+}	<20	-	-	50
F	170	58	34	800
Hg	0.001	0.00099	99	0.5
Ni	5.2	3.1	60	-
Pb	0.2	0.015	7.5	10
Se	<2	-	-	10
Zn	2.1	0.3	14	-

Av: Average, SD: Standard deviation, RSD: Relative standard deviation

Figure Captions

Fig.1 Schematic diagram of container experiment set-up.

Fig. 2 Change in sediment pH over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 3 Change in LOI over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 4 Change in AVS over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 5 Change in NH_4-N concentration in pore water over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 6 Change in NO_3-N+NO_2-N concentration in pore water over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 7 Change in DO concentration in overlying water over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 8 Change in PO_4-P concentration in overlying water over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

Fig. 9 Change in NO_3-N+NO_2-N concentration in overlying water over time in the container experiment.
Control: 50 L of sediment only, BFS: mixture of 13 L of BFS and 50 L of sediment.
Bars represent standard deviation of triplicates.

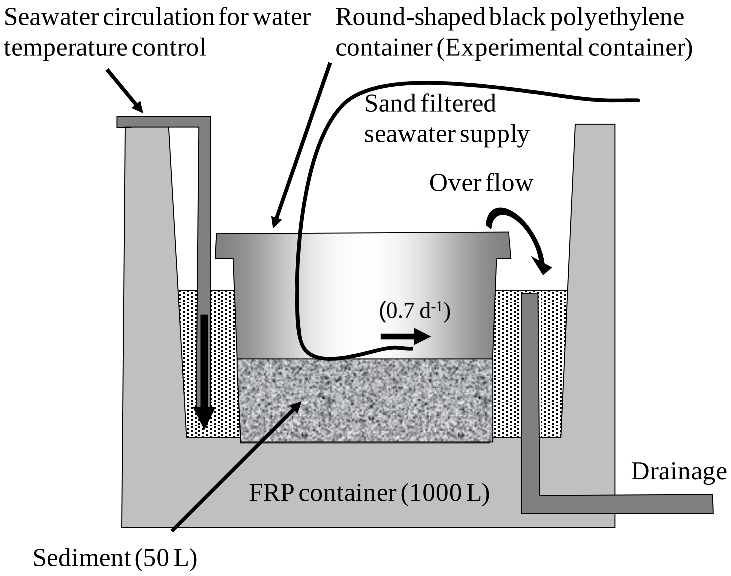
Seawater circulation for water temperature control
Round-shaped black polyethylene container (Experimental container)
Sand filtered seawater supply
Over flow
(0.7 d-1)
Drainage
FRP container (1000 L)
Sediment (50 L)

Fig.1

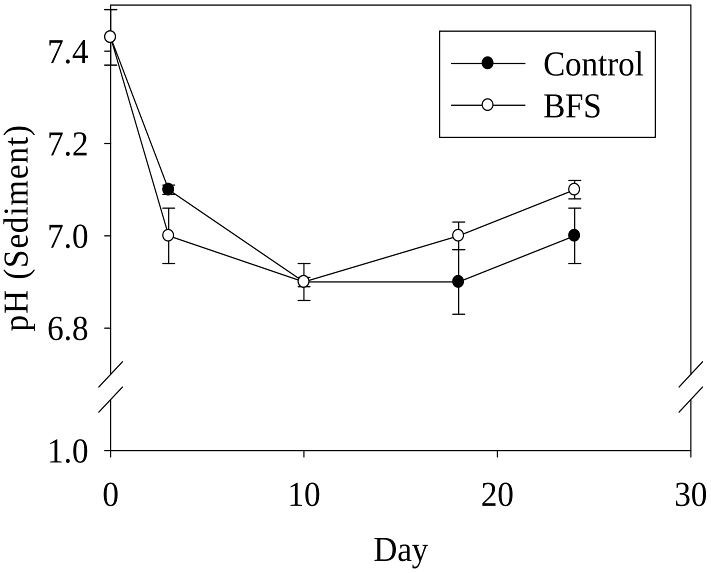

Fig. 2

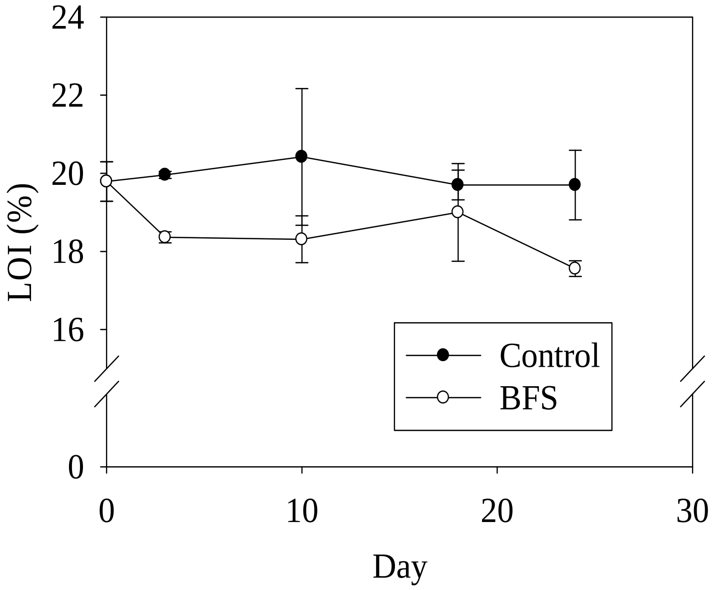

Fig. 3

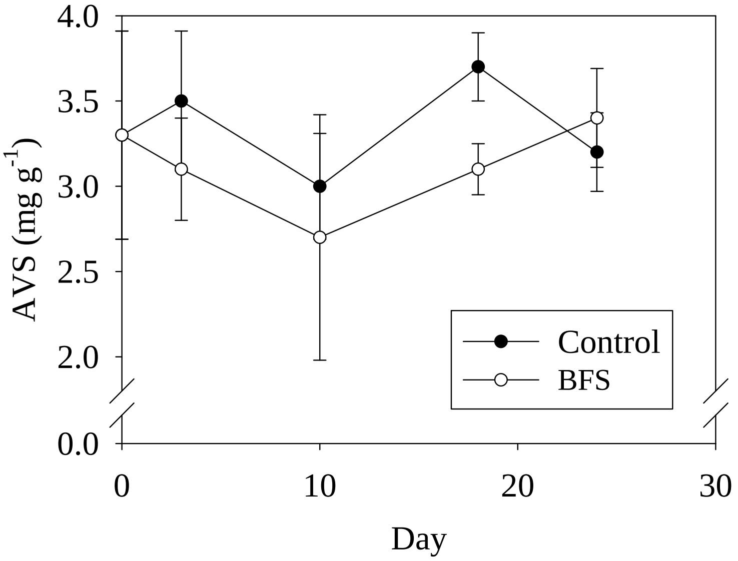

Fig. 4

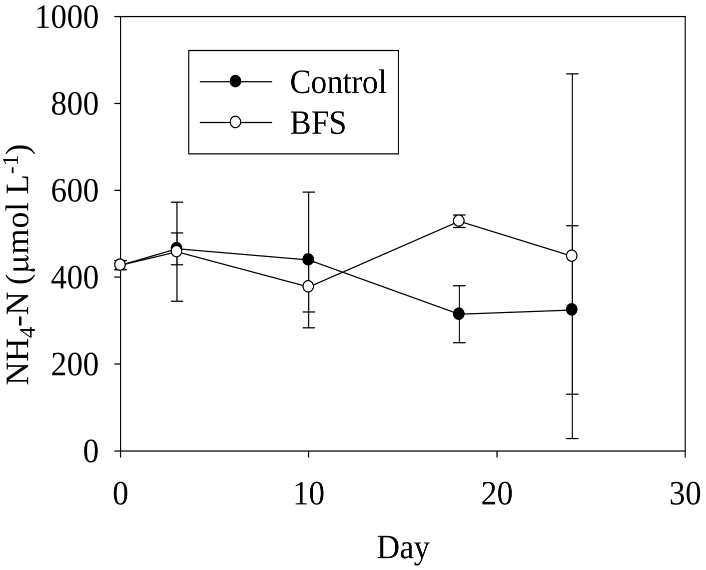

Fig. 5

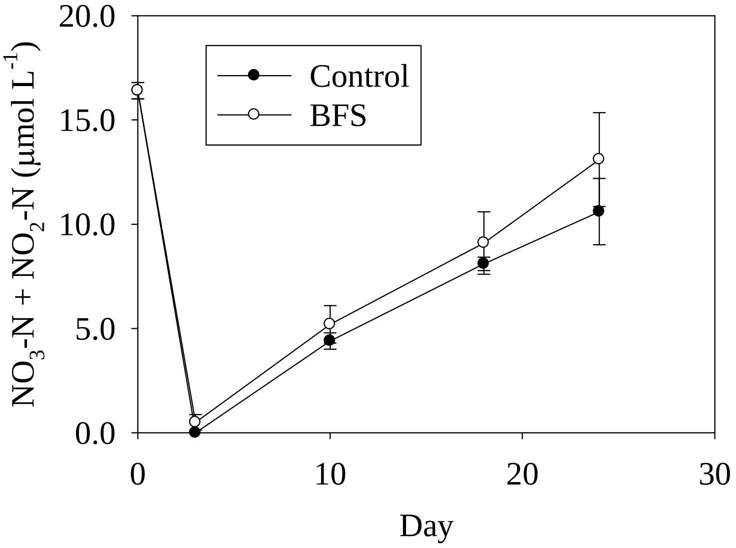

Fig. 6

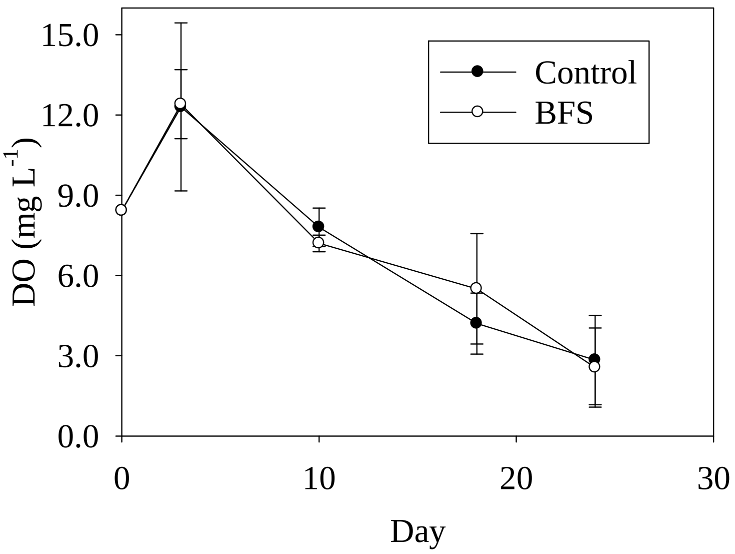

Fig. 7

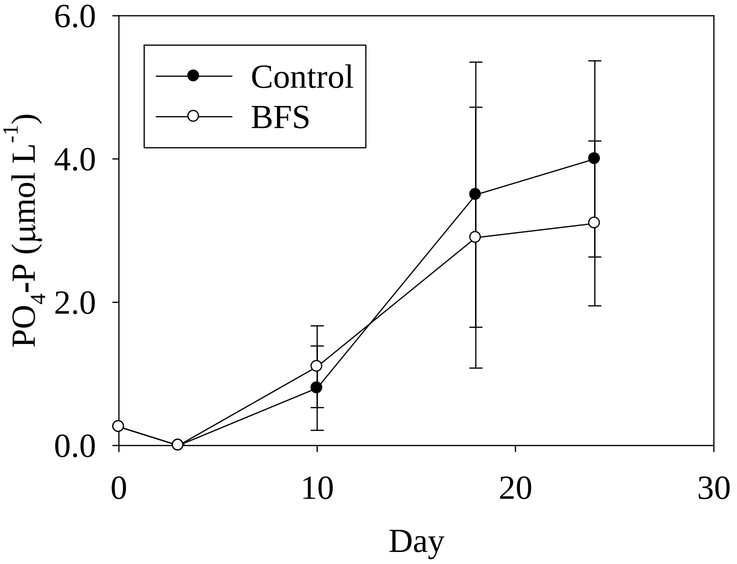

Fig. 8

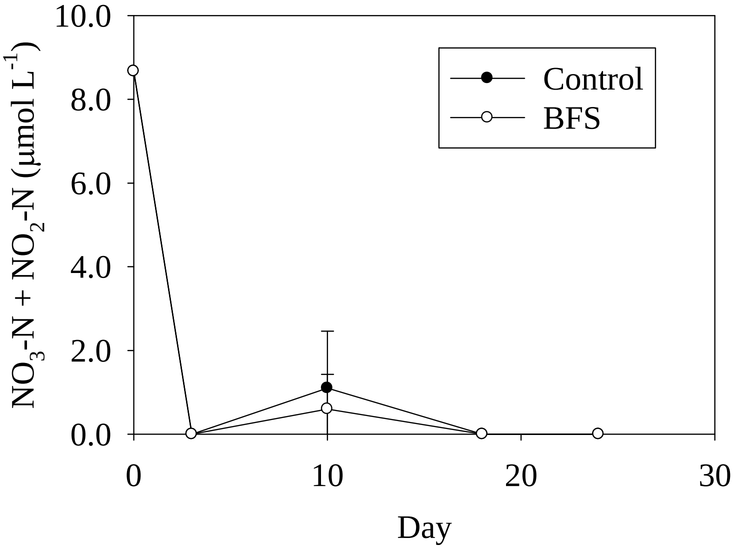

Fig. 9