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# Effect of changes in the physicochemical properties of sand-alternatives on bacterial community structure in coastal sediments

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#### ABSTRACT

Steel slag and its granulated agglomerates, and the granulated agglomerates of ash from thermal power stations are reserved for and partially used in the construction of sand-alternatives (SAs) in coastal restoration and creation projects. These SAs contain CaO and leach calcium and alkalis that may affect biological activity of microorganisms colonizing surrounding sediments. We evaluated the chemical behavior of four SAs, namely, decarburized slag (DCSlag), dephosphorized slag (DPSlag), granulated agglomerates of DCSlag (GSlag), and granulated agglomerates of coal fly ash (GAsh) in coastal sediment and the impact of these SAs on bacterial community structure in the sediment. The SAs eluted calcium with a decrease in the surface calcium content in the sediment. The elution from slag and slag-based SA (DCSlag, DPSlag, and GSlag) continued for 8 weeks, whereas the elution from GAsh continued for 24 weeks. The different elution periods are explained by the depth of eluted calcium found in the SAs;  $0.19\pm0.07$ ,  $0.04\pm0.02$ ,  $0.27\pm0.12$  and  $2.4\pm0.97$  mm in DCSlag, DPSlag, GSlag and GAsh, respectively. After 1 year, the DGGE (Denaturing Gradient Gel Electrophoresis) profiles were different among all the SAs except for GSlag and GAsh. The taxon richness of bacterial communities was estimated by the number of DNA bands found in the sediment mixed with the SAs, and it was lower than that in the natural sediment. Physicochemical properties of the SA's have been shown to affect the bacterial community structure. On the other hand, the communities recovered to almost the same level as that in the natural sediment after 1 year. The long-term effects of chemicals eluted from the SAs on bacterial community structure would be limited or negligible.

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#### 1. Introduction

Seagrass beds and tidal flats have important functions in marine ecosystems. However, a number of tidal flats and eelgrass beds have disappeared throughout the world because of marine pollution and human activities such as dredging, development, and reclamation of coastal zones. The decrease in seagrass beds and tidal flats is significant in enclosed coastal areas. For example, 20% of tidal flats and 70% of seagrass beds have disappeared since the 1960s from the Seto Inland Sea, the largest enclosed sea in Japan (Ministry of the Environment and Japan, 2015). Because of the decrease of tidal

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http://dx.doi.org/10.1016/j.ecoleng.2015.12.025 0925-8574/© 2015 Elsevier B.V. All rights reserved. flats and seagrass beds, their restoration was demanded in recently. Therefore, restoration projects and studies for the recovery of sea grass beds and tidal flats have been conducted all over the world (Ealey et al., 2007; Terawaki et al., 2005).

Transplanting seeds or adult shoots is a method of seagrass bed restoration. Sand mounding is effective in areas where seagrass beds existed in the past and were lost through a decline in water transparency due to eutrophication (Sugimoto et al., 2008). A large amount of sand is required to restore such seagrass beds. Restoration and creation of tidal flats also require a large amount of sand. Sea and river sands are often used as a base material in coastal restoration projects (Hosokawa, 1997; Lee et al., 1998). However, the excavation of sand from sea and river beds disturbs aquatic ecosystems. In Japan, massive burrow (pits about 20–30 m in depth) are widely distributed in the bottoms of several coastal regions







as a result of sand excavation projects. About 610 million m<sup>3</sup> and 100 million m<sup>3</sup> of sand have been excavated from the Seto Inland Sea and Tokyo Bay, respectively (Naito et al., 2006). Severe oxygen depletion, blue tides, and damage to ecosystems within and near the pits have occurred (Naito et al., 2006; Takeoka, 2002; Tanaka et al., 2008). It is necessary then, to develop sand-alternatives (hereinafter referred to as "SAs") for use in coastal restoration and creation projects.

Iron and steel slags from steelmaking processes and coal ash from thermal power stations are industrial byproducts, and approximately 40 million tons and 10 million tons, respectively, are produced per year in Japan (Tanaka et al., 2010; Tekko Sulagu Kyokai, 2010; The Japan Society of Industrial Machinery Manufactures, 2012). Because they contain no organic material and have high durability, they have been used as roadbed construction material, a coarse aggregate in concrete, and raw material for cement mixing (Martin, 1990; Tanaka et al., 2010). They are also reserved for and partially used as constructed materials of coastal restoration and creation (Nishijima et al., 2015a, 2015b).

Iron and steel slags are mainly classified into two categories, namely, blast furnace slag and steelmaking slag (or steel slag) which includes converter slag. The former is produced from the conversion process of iron ore to pig iron, and the latter is produced with the addition of CaO through the purification processes from pig iron to steel. The steel slags are further subdivided into desiliconized slag, dephosphorized slag, decarburized slag, and others, depending on the specific purification process of pig iron being deployed. There is an accompanying granulated agglomerate using cement and fly ash from coal ash that can add value to its secondary use. (Utagawa and Matsunaga, 2008). An SA is also produced from coal fly ash from thermal power stations by agglomerating with about 10% cement, meaning that the agglomerate contains CaO (Asaoka et al., 2008; Murayama et al., 2001).

These SAs with particle sizes of 0.5-4.0 mm are almost the same size as natural sand in sandy beach. In addition, some of them have several advantages in terms of environmental remediation (Asaoka et al., 2009; Hayashi et al., 2011; Yamamoto et al., 2003). For example, Yamamoto et al. (2003) reports claim that steel slag suppresses phosphate liberation from sediment and decreases acid-volatile sulfide in organically enriched sediment because of the iron content. It can also create reef-like habitats for fish and promote proliferation of some marine phytoplankton. On the other hand, these SAs consisting CaO have been known to increase pH in pore water (interstitial water) by elution of CaO and to induce the solidified by hydration reactions of CaO in some case. The rise in pH triggers various chemical reactions between the SAs and seawater and sediment and directly affects microorganisms and other creatures (Etana, 2005; Futatsuka et al., 2003, 2004; Miki et al., 2004a, 2004b). CaO adsorbs P by forming Ca phosphate precipitation and crystallization on slag (Barca et al., 2014). However, limited information about chemical behavior and the fate of the SAs with CaO in environment have been reported. We have evaluated the chemical behavior of 6 SAs including two kinds of steel slag and the granulated agglomerates from the steelmaking process, clinker ash and granulated agglomerates from thermal power stations, molten slag from municipal solid waste incineration plants in seawater. We then obtained basic information on maximum calcium elution potentials and elution depth of the 6 SAs (Okuda et al., 2014). The maximum calcium elution potential of decarburized slag was 204 mg g<sup>-1</sup> and much higher than other SAs. The elution of calcium from decarburized and dephosphorized slags was limited to depths of about 140 and 300 µm from the surface. However, the chemical behavior of the SAs in the sediment was different from that in the seawater because diffusion of chemicals eluted from the SAs was limited and the environmental conditions of the SAs are quite different from those in the sediment. Moreover, the

SAs may affect microorganisms in sediment and may change the microbial community structure in the sediment. Microorganisms play a pivotal role in regulating major geochemical, ecological, and environmental processes in marine ecosystems, particularly the trophodynamics and biogeochemical cycles (Nogales et al., 2011). In this study, we evaluated the chemical behavior of the SAs with CaO in coastal sediment and the impact of the SAs on bacterial community structure in the sediment.

#### 2. Materials and methods

#### 2.1. Sand-alternatives

Decarburized slag (DCSlag), Dephosphorized slag (DPSlag), granulated agglomerates of DCSlag (GSlag), and granulated agglomerates of coal fly ash (GAsh) were used as sand-alternatives. The DCSlag and the DPSlag were generated through decarburization and dephosphorization processes that take place when converting pig iron into steel, respectively. The GSlag, which is composed of stone-like granules, is produced by granulating granulated agglomerates of steel slag (about 60%) and blast furnace slag (about 20%) with calcium hydroxide and the fly ash from coal (Matsunaga et al., 2003). The two slags and their granulated agglomerates were supplied by a steelmaking plant in Japan. GAsh is a commercial product made by the granulation of coal fly ash from coal thermal power stations with cement as fixation agent (Asaoka et al., 2008). These sand-alternatives contain CaO originating from an additive used during steelmaking process (DCSlag, DPSlag and GSlag) and production processes of (GSlag), and cement (GAsh).

The chemical compositions of the four SAs are summarized in Table 1 (Okuda et al., 2014). Ca, Si and Fe were major elements in the four SAs.

#### 2.2. Aging experiment in coastal sediment

The chemical behavior and the impact of each SA on the bacterial community structure in coastal sediment were evaluated by aging the SAs in the coastal sediment for one year. The aging site was located on the coast of Iwakuni in the Seto Inland Sea of Japan, and the water depth ranged from D.L. -1.0 to -3.0 m. The aging experiment started on March 5, 2010. The averages of major parameters of the seawater at its surface were pH 8.2, chemical oxygen demand (COD) 2.4 mg  $l^{-1}$  and dissolved oxygen (DO) 8.9 mg  $l^{-1}$ . SAs with a diameter of 8–20 mm were used for the aging experiment. At least 10 granules of each SA (40–130g) were mixed with the same amount of sediment taken from the site, and were placed into polypropylene nets with a 3 mm size mesh. The 10 nets with the sediment mixture were prepared for each SA. Since the mesh size was larger than the particle size of the sediment but smaller than the particle size of any SA, the nets were carefully inserted in plastic bags and transported by a diver to the aging point. At the aging point, 10 cm of sediment was dug out, and while the bags are submerged in the dug part, the nets were carefully removed and buried under 10 cm of sediment avoiding any spills. After the aging

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Chemical composition of surface of SAs (Okuda et al., 2014).

Elements (% of atom)	DCSlag	DPSlag	GSlag	GAsh
Ca	$61\pm8$	$44\pm29$	$31\pm5$	$17\pm8$
Si	$17\pm13$	$9\pm16$	$40\pm 6$	$44\pm20$
Mg	N.D.	$7\pm10$	$7\pm3$	$3\pm 2$
Fe	$14\pm21$	$30\pm 30$	$6\pm5$	$4\pm 2$
Al	$4\pm5$	$7\pm11$	$3\pm5$	$25\pm12$
Na	N.D.	N.D.	N.D.	$0.5\pm1$
Others	4	4	14	6

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