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Leaching techniques to remove metals and potentially hazardous nutrients from trout farm sludge

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ABSTRACT

A fish farm sludge high in P (2–6% w/w as dry matter), Fe (5–7%), C (40–50%) and N (0.8–4%) was subjected to a series of acid leaching treatments using HCl, organic acids, and biologically mediated acid production. Additions of biodegradable organic acid solubilized heavy metals better than HCl, while additions of 1.5% w/v glucose followed by 7 day incubation stabilized the sludge releasing 92% P, 100% Fe. The use of homo-lactic *Lactobacillus plantarum* starter cultures were more effective than hetero-lactic *Lactobacillus buchneri*, solubilizing 81.9% P, 92.2% Fe, 93.0% Zn and 96.4% Ca in the sludge. The anaerobic sludge-glucose fermentation using *L. plantarum* produced a leached sludge that has low heavy metal and nutrient content while affording the recovery of nutrients. The potential of these methods for practical application are briefly discussed.

1. Introduction

The effluents and the sludge from recirculation aquaculture systems RAS are nutrient rich materials high in carbon, nitrogen and phosphorous and accumulated heavy metals. These materials can contribute to eutrophication when released to inland and coastal waters (Barak and Rijn, 2000; Cripps and Bergheim 2000; McIntosh and Fitzsimmons 2003; Lovitt et al., 2008). Furthermore an accumulation of P and Fe in recirculation water can interfere with pumping and bio-filters resulting in poor hygiene and disease (Barak and Rijn, 2000; Bayat and Sari, 2010; Lekang et al., 2000; Hunter et al., 2001; Pathak et al., 2009; Patterson et al., 2003). Therefore adequate treatment of these wastes prior to discharge or reuse of wastewater is highly desirable.

In RAS most nutrients (60–70% w/v) are in the form of solid particles of residual feeds and fish feces rather than liquid, and so can be easily reduced by removal of solid particles with coarse filters, drum filters, bio-filters and by gravitational sedimentation many of the potential harmful substances can

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be removed as, or absorbed onto, solids or destroyed in biological oxidation (Thomas et al., 2000; Van Rijn et al., 2006) prior to discharge (Hussenot et al., 1998; Klas et al., 2006). In addition, other processes typically involve insoluble salt precipitation, use of a microbial mass and plant biomass in constructed wetlands (Doyle and Parsons 2002; de-Bashan and Bashan 2004). The separated sludge from recirculating stream of rearing water also contains high concentrations of heavy metals and is a natural sink for such materials.

There are many methods of sludge disposal, however with growing environmental concerns, incineration can be a cause of the air pollution (Babel and del Mundo Dacera, 2006), land application and landfill can be restricted due to land contamination by heavy metals and the nutrient load in the sludge. In many cases heavy metals have to be removed prior to land application and landfill of the sludge other wise it can be considered a toxic waste (Sreekrishnan and Tyagi 1996). The dewatered sludge can be potentially recycled to composts, feeds and high value products if contaminating heavy metals, dioxins, poly-chlorinated biphenyls (PCBs) and human pathogens in the sludge are appropriately reduced by chemical or biological treatments.

The removal of P and heavy metals in the sludge can be carried out using acid-treatment, chelating agents, biological treatment, electroreclamation and supercritical fluid extraction (SFE). In metal leaching, the pH is very important and can be achieved by the use of mineral or organic acids or it can be mediated via microbial processes that form organic acids (Chen and Lin 2001). It has been shown that microbial mediated leaching, or bioleaching, can be superior to other treatments (Sreekrishnan and Tyagi 1996; Babel and del Mundo Dacera, 2006). Also, organic acids and biological treatments are better because of their biodegradability and the relative mildness of the processes involved. Bioleaching is also more beneficial than chemical leaching alone due to an improved bioavailability of materials and its detoxification during the microbe-mediated processing of sludge (Chen et al., 2005). The efficiency of bioleaching to reduce metal content and toxicity of the sludge was verified using terrestrial and liquid-phase bioassay, rendering the sewage sludge useable for land application (Renoux et al., 2001).

One drawback to biological treatment is that it is more difficult to carry out than chemical treatments as it is typically dependent on environmental control of pH and temperature and the provision of nutrient and energy sources. Energy sources like glucose, molasses and non toxic metal salts with high redox potential can accelerate leaching of the heavy metals. Acidophilic obligate chemolithoautotropics like *Leptospirillum ferooxidans* (Bosecker 1997), *Acidothiobacillus ferrooxidans* and *Acidothiobacillus thiooxidans* at $<$pH 4.0 with ferrous sulfate or the addition of sulfur can help release heavy metals from the sludge (Sreekrishnan and Tyagi 1996).

In the same way anaerobic heterotrophic bacteria such as the lactic acid bacteria can produce biodegradable organic acids using carbon sources in the sludge or supplemented with carbohydrate to make an acid stabilized silage material (Weinberg and Muck 1996). The organic acids produced during growth can reduce the pH and should elevate extraction and chelation of P, Fe and other metals in the acid conditions. *Lactobacillus plantarum* contains non-specific acid phosphatases, that are capable of degradating of organic material to release bound phosphates and poly-phosphates (Zamudio et al., 2001). Thus for example, they may be also be able modify sludge making it more easy to dewater.

The leached materials are then recovered as insoluble materials if treated appropriately. For example, phosphate can be captured in a form of hydrated Amorphous Calcium phosphate (ACP, $Ca_3(PO_4)_2$ X(H_2O)) or hydroxyapatite ($Ca_{10}(PO_4)_6$ $(OH)_2$), vivianite ($Fe_3(PO_4)_2$ $8H_2O$), struvite (NH4MgPO4 6H2O) in presence of appropriate concentrations of Calcium, Ferrous and Magnesium ions in neutral or alkaline condition. The resultant capture phosphate salts have considerable value for industrial applications (Doyle and Parsons, 2002; de-Bashan and Bashan, 2004; Suzuki et al., 2005). Struvite is a route for phosphate recovery used for land application as it is precipitated with nitrogen in wastewater. ACP and vivianite can be subjected to incineration. Hydroxyapatite has potential as this a major component of bone. Solubilized metals can also be recovered from the leachates by chemical precipitation at neutral pH as hydroxide, carbonates or sulphides.

In this investigation, sludges have been treated with organic and mineral acids together with inoculation of hetero-lactic bacterium, *Lactobacillus buchneri* or homo-lactic bacterium, *L. plantarum*. These processes were then compared for their ability to leach of metals and nutrients from sludge derived from a RAS trout farm. In this case we have investigated the leaching P, Zn, Ca and Fe from the sludge. In addition we have also studied the changes in sludge characteristics caused by the leaching process.

2. Materials and methods

2.1. The sludge and waste effluents from Danish trout farms

The aquaculture sludge used for stabilization and bioleaching studies were obtained from a RAS Trout Farm in Hoghoj, Denmark. The sludge from the farm was stored in an open sedimentation tank and is derived from two streams, one from a drum filter (75 micron mesh) backwash and was a continuous flow, the other stream sludge was from biofilter washing and intermittent stream from daily washing of the filter. Samples were collected and stored at 4 °C for up to 24 h prior to analysis. Wastewater and the sludge were collected and centrifuged (10,000 g at 4 °C) for concentration prior to treatments.

2.2. Measurement of total suspended solids and volatile solids

Wastewater from trout farm was filtered with GF/C (Whatman) filter which was soaked in deionized water for one day before filtration and then dried in an oven at 105 °C. GF/C filters with filtrates were dried at 105 °C dry oven for 4 h. Total suspended solids (TSS) (g L^{-1}) is a weight difference between the blank GF/C filter and the weighed according to the standard water and wastewater assay method (Standard methods for the examination of water and waste water 20th (ed.), 1998).