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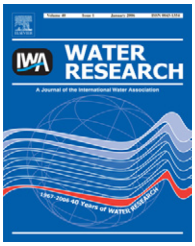

Treatment of leachate from MSWI bottom ash landfilling with anaerobic sulphate-reducing process

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ABSTRACT

Removal of sulphate and toxic elements from the leachate of a field landfill lysimeter ($112\,m^3$), containing municipal solid waste incineration (MSWI) bottom ash, was studied. The leachate was treated in two parallel laboratory upflow anaerobic sludge blanket (UASB) reactors without and with ethanol as additional carbon source. With ethanol more than 65% of sulphate was removed, while without ethanol removal was negligible. The treatment removed Ba, Ca, Cu, Mn, Mo, Ni, Pb, Tl, Sb, Se, Sr, and Zn of the studied 35 trace and other elements. The sequential extraction of the reactor sludge at the end of runs confirmed that with a few exceptions (Ba, Ca, and Cu) the main mechanism by which the elements were removed was precipitation as sulphides.

1. Introduction

Municipal solid waste incineration (MSWI) is an increasingly used method of solid waste pre-treatment. However, incineration produces residues which need to be properly managed. Bottom ash typically forms 85–90% by weight of the incineration residues, which is 25–45% of the mass of the incinerated municipal solid waste (MSW) and about 10% of its volume (Hjelmar, 1996).

Bottom ash can be utilised for example in road bases and as concrete aggregate, but with a number of restrictions. Bottom ash contains corrosive compounds and as a heterogeneous material it may require homogenisation or screening into size classes (Wiles, 1996; Forteza et al., 2004). In addition, for the economic and logistical reasons site of utilisation must be within reasonable proximity to the incinerator. Bottom ash will be landfilled, whenever safe utilisation is not possible, at least in countries where solutions such as placement in salt mines are not available (Hjelmar, 1996).

Emissions from bottom ash landfilling should be assessed and landfills designed such that the environmental burden and period of active management of landfills are minimised. Leachate is the major emission from bottom ash landfilling both in quality and quantity. Bottom ash leachate differs markedly from MSW landfill leachates, which are rich in organic material and nitrogen compounds; instead, it contains trace elements and inorganic salts such as chlorides and sulphates of calcium, potassium, and sodium (Sabbas et al., 2003).

Treatment of ash leachates has been previously studied e.g. in constructed wetlands and by reverse osmosis. Constructed wetland removed trace elements from coal ash leachate effectively over a long period (Ye et al., 2001). Salts and dioxins have been successfully removed by reverse osmosis from leachate of incineration residues (Ushikoshi et al., 2002). However, the presence of sulphate and trace elements in the leachate makes biological sulphate-reduction a potential technology following on from its application to the removal of metals from mining waste waters and groundwater (Lens

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and Hulshoff Pol, 2000). Contrary to acidic mining waste waters, which have high concentrations (hundreds of milligrams per litre) of few elements (e.g. Cu, Zn) ash leachates are alkaline and they contain relatively low concentrations of many trace elements.

In the sulphate-reducing process, sulphate-reducing bacteria (SRB) use sulphate as an electron acceptor in a process where hydrogen or organic compounds are oxidised and sulphate simultaneously is reduced to sulphide. Sulphide reacts with divalent metal cations and with some metals or metalloids of the other oxidation states (e.g. Ag_2S, As_2S_3, Sb_2S_3) to form sulphide compounds with a low solubility, whereas sulphides of e.g. alkaline earth metals are less stable in the presence of water. When the ratio of the chemical oxygen demand (COD) and sulphate in waste water is greater than 0.67, which is calculated using a stoichiometric equation, all of the sulphate can be reduced to sulphide by SRB (Widdel, 1988). Bottom ash leachate contains some organic carbon, but the amount is small compared to the sulphate concentration, and therefore the addition of a source of organic carbon may be necessary in order to remove all the sulphate.

The objective of this study was to assess feasibility of a sulphate-reducing process in the treatment of bottom ash leachate. The chemical properties of leachate from a landfill lysimeter containing MSWI bottom ash were studied over a period of 280 days after landfilling. The ability of the sulphate-reducing upflow anaerobic sludge blanket (UASB) reactors to remove COD, sulphate, trace elements, and other elements as well as the concentration and binding mechanism of the elements in the sludges of the reactors were studied.

2. Materials and methods

2.1. Leachate

Bottom ash was obtained from an incinerator using fixed bed technology (31 years old, modernised 1995, Turku, Finland). The incinerated MSW from which glass, paper, and metal were source-separated was from Turku region. Bottom ash was transported immediately after quenching with trucks to Mustankorkea landfill in Jyväskylä (Finland) where it was placed in a field landfill lysimeter (height 3.9 m, width 2.4 m, length 12 m). The ash was compacted with a sheep foot roller and was exposed to ambient weather conditions. Leachate was collected from the drainage system into the well by gravity, from which samples were taken. Preliminary experiments (data not shown) suggested inhibition of sulphate reduction when undiluted leachate was used, hence leachate was diluted with tap water (1:1) before feeding to the reactors. Concentrations of the elements were all under the detection limits in the tap water. Ethanol (96%) was added (1 g/l) as an electron donator in the feed of reactor R2 from day 30 onwards. Influent COD without ethanol addition was 314 (SD 181, $n = 57$) mg/l and biological oxygen demand (BOD_7) 15 mg/l (1 sample).

2.2. Reactor experiments

Two parallel continuously fed UASB reactors (0.5 l, height 345 mm, inner diameter 47 mm, glass) were run at $20 \pm 2\,^{\circ}C$. Reactors were fed with diluted leachate, reference reactor R1 without added ethanol and R2 with the ethanol addition. The feed was prepared twice a week and was stored in 2 l glass bottles in the refrigerator (4 °C) under nitrogen. The feed was pumped to the reactors with constant flow rate (hydraulic retention time 1 day). Both reactors were inoculated with 300 ml of anaerobically digested sludge (total solids (TS) 17.8 g/l, volatile solids (VS) 9.9 g/l) from a local municipal waste water treatment plant (Jyväskylä, Finland), after which reactors were flushed with nitrogen and air-sealed. The tubing was made of PVC and the influent and effluent sampling sites were located immediately in front of and behind the reactors. Aluminium gas bags were used to collect biogas from the top of the reactors.

2.3. Analyses

COD, BOD_7, TS, and VS were determined according to Finnish standard methods (SFS 5504, 1998; SFS-EN 1899-2, 1998; SFS 3008, 1990). Sulphate was determined according to the Standard Methods (APHA, 1998). pH was measured immediately after sampling with a Metrohm 744 pH meter. Sulphide was determined according to the colorimetric method (Trüper and Schlegel , 1964), the absorbance was measured at 670 nm (Hitachi U-1500 UV/VIS).

Concentrations of Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, Hg, In, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Sb, Se, Si, Sr, Tl, U, V, and Zn were determined from both the influents and effluents. Samples were preserved by making them acidic (pH 2) with HNO_3 and stored at 4 °C before the measurements. Concentrations of the elements were determined with a Perkin–Elmer Optima 4300 DV ICP-OES using the default parameters of the instrument (nebuliser flow 0.8 l/min, plasma power 1300 W, auxiliary gas flow 15 l/min). The concentrations of As, Ca, Cr, Cu, Mo, P, Pb, S, and Zn in the inoculum were determined with an ultrasound assisted extraction method followed by ICP-OES at the beginning of the runs and the binding mechanisms of the same elements were evaluated with the sequential extraction method at the end of the runs. Five fractions were obtained from the sequential extraction: (1) elements bound by ion exchange pH 7, (2) elements bound to carbonates pH 5, (3) elements bound to Fe and Mn oxides pH 2, (4) elements bound to organic matter and sulphides pH 2, and (5) the residual phase. The method used is described in detail by Väisänen and Kiljunen (2005).

3. Results

The feasibility of the sulphate-reducing process to treat the bottom ash leachate was studied using two UASB reactors (R1 and R2). In reactor R1, which was fed with the diluted leachate COD removal varied and was negligible, while in reactor R2 60–70% of the COD was removed after ethanol addition (Fig. 1). The influent pHs ranged from 7.45 to 8.46, and the pH of the effluents followed those changes (Fig 1). At the beginning of the runs the amount of TS was 5.35 g/reactor and VS 2.97 g/reactor. At the end of the runs TS was 3.62 g/reactor in R1 and 10.0 g/reactor in R2. Sulphide concentration