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Removal of toxic metals during biological treatment of landfill leachates

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ABSTRACT

Progressive implementation of the European Water Framework Directive has resulted in substantial changes in limits for discharges of heavy metals both to watercourses, and to sewer. The objective of this paper is to provide original, real, full-scale data obtained for removal of metals during aerobic biological leachate treatment, and also to report on studies carried out to look at further trace metal removal. Polishing technologies examined and investigated include; the incorporation of ultrafiltration (UF) membranes into biological treatment systems, the use of ion exchange, and of activated carbon polishing processes. Ultrafiltration was able to provide a 60 percent reduction in COD values in treated leachates, compared with COD values found in settled/clarified effluents. Removal rates for COD varied from 30.5 to 79.8 percent. Additionally, ultrafiltration of treated leachates significantly reduced both chromium and nickel concentrations of effluents by 61.6% and 34.3% respectively (median values). Despite mean reductions of chromium (9.7%) and nickel (13.7%) noted during the ion exchange trials, these results would not justify use of this technology for metals removal at full-scale. Further preliminary studies used pulverized activated carbon (PAC) polishing of UF effluents to demonstrate that significant (up to 80 per cent) removal of COD, TOC and heavy metals could readily be achieved by doses of up to 10 g/l of suitable activated carbons. Additional evidence is provided that many trace metals are present not in ionic form, but as organic complexes; this is likely to make their removal to low levels more difficult and expensive.

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1. Introduction

As a result of both EU and national legislation, consented limits for discharge of treated landfill leachates into surface watercourses continue to become more restrictive. For example, denitrification processes, and phosphate removal, are increasingly having to be included in on-site biological treatment systems. Most recently, in the UK and Ireland, the attention of regulators has focused on concentrations of toxic (heavy) metals in treated leachates. Although there is much evidence that levels of these metals rarely exceed values routinely found in raw and treated domestic wastewaters (Vaverková and Adamcova, 2014; Kjeldsen et al., 2002; Kjeldsen and Christophersen, 2001; Christensen et al., 2000; Christensen et al., 2001; Christensen et al., 1994), nevertheless, consent limits are now routinely being set at levels in the order of 100 or 200 µg/l, for metals such as chromium.

Although there are extensive UK databases (e.g. Robinson, 2005, 2007; Robinson et al., 2004) for the presence and concentrations of toxic metals in leachates from domestic, commercial and industrial waste landfills, few detailed data exist to inform operators and regulators about rates of removal of individual metals that can be

anticipated during biological treatment of leachates, using processes such as Sequencing Batch Reactors (SBRs), which have been routinely and widely adopted.

This paper seeks to provide such data, using case studies from full-scale, on-site, biological leachate treatment plants in the United Kingdom, and also to provide data sets for a range of process configurations that have modified the SBR process; for example, by incorporation of ultrafiltration (UF) membranes for solids separation, to examine additional removal of metals that can be achieved.

Initially results were obtained following a study of twelve full-scale, on-site leachate treatment plants, comparing treated leachate from the biological treatment systems with effluent following passage through UF membranes. At each site, an extensive series of field trials incorporating a pilot-scale ultrafiltration (UF) plant was used to investigate the additional removal of COD and heavy metals which can be achieved, compared to the discharge of settled effluents normally adopted in the SBR process.

Other treatment processes which can be added for effluent polishing are also discussed, with data provided for the use of ion exchange resins, and activated carbon for several instances where metals may be present in organically complexed forms, rather than simply present as dissolved ions.

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Ion exchange resins are known to be very good at removing metals such as chromium, nickel and copper, where these are present in an ionic form, so poor removal would provide support to the belief that metals were not present as metal ions. These resin trials would look at the removal of both organic compounds (determined as COD and TOC), and selected trace metals, to determine whether removal rates were correlated, which would provide further evidence of the metals being present as organic complexes.

The paper focuses particularly on the metals chromium, copper and nickel, since these are generally of most concern to regulators. Chromium in particular has greatest potential for toxicity at relatively low concentrations (Christensen et al., 2000, 2001; Guertin, 2004), both to the natural environment, and also to biological treatment processes such as nitrification.

Removal of trace metals from treated landfill leachates by ion exchange is not a process that can be tested or optimised without suitable pilot-scale trials, in which actual liquids to be treated were passed through small-scale columns of selected resin at controlled rates. Although used successfully for removal of metals from a wide range of industrial effluents, removal efficiency depends critically on the chemical nature of the chromium, and the chemical matrix in which it is contained.

Based on a belief that the poor removal of trace metals measured during biological treatment almost certainly results from them being present as organic complexes, a series of experimental ion exchange trials were designed and carried out. Using the Puro-lite S930 Plus ion exchange resin, four different effluents from UK leachate treatment plants were treated, and metal removal observed.

Subsequent trials were carried out on one of these treated leachates, to examine adsorption using activated carbon, in an attempt to provide evidence of the correlation between removal of COD, TOC and particular trace metals, further examining the theory that some trace metals are present as organic complexes. The objective was to examine broadly the removal of organic compounds (measured as COD and TOC) as well as that of chromium, although removal of two other metals (copper and nickel) would also be determined.

The paper has been written in a very practical way, to provide information and advice that is relevant and realistic for landfill operators seeking reliable, robust, relatively simple and automated treatment of leachates at their sites. It is recognised that an increasing proportion of leachate treatment systems are now being operated at closed landfills, which need to take greater note of removal processes for toxic metals. Otherwise unpublished data from full scale leachate treatment systems are presented, and factors that might affect metal removal in these systems are explored.

2. Toxic metals in landfill leachates

2.1. Presence in raw leachates

Toxic or heavy metals, of which these studies will consider chromium, nickel, copper, zinc, cadmium, and lead, have not generally been a large problem in treatment or disposal of landfill leachates from either sites receiving primarily household wastes, or those where co-disposal of selected hazardous wastes (often including metal sludges, etc.) has been practiced according to scientific principles. A large review of leachate quality on behalf of the UK Government, (Robinson, 1996), concluded that, (with the exception of zinc in acetogenic leachates), concentrations of these toxic metals in leachates were no higher than those found in domestic wastewaters, and often significantly lower. Acetogenic leachates are produced during the earlier phases of decomposition of landfilled wastes, where hydrolytic, fermentative, and aceto-

genic bacteria produce an accumulation of carboxylic acids, and therefore a pH decrease (Kjeldsen et al., 2002).

Progressive implementation of the Water Framework Directive (Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy) has resulted in substantial changes in limits for discharges of heavy metals both to watercourses, and to sewer. For example, whereas limits such as <1.0 mg/l or <0.5 mg/l were previously typical for metals such as chromium and nickel, recent limits in the order of <200 µg/l or <300 µg/l are now routinely being applied.

On this basis therefore, whereas previously no specific treatment for removal of toxic metals was required for discharge compliance in the UK, during the last 12 or 18 months, this aspect of treatment has had to be considered more carefully. Leachate treatment plant designers have not had access to adequate data, that would enable treatment processes to be selected that will meet the newer standards reliably and consistently.

2.2. Sampling and analysis of trace metals in raw and treated leachates

The sampling and analysis of leachates, treated leachates, and river water samples for determination of a wide range of parameters, is not straightforward (Claret et al., 2011; Kjeldsen et al., 2002). UK Environment Agency specific guidance for sampling of landfill leachate, groundwater and surface water (for example, UK Environment Agency, 2014, page 114), contains several statements which are relevant. This guidance highlights the impact biological and chemical processes can have on changing the composition of samples, in addition to the importance of 'fixing' dissolved constituents within a sample, using preservatives. Additionally, the process of filtering samples and the critical importance of consistency in sampling practice are discussed.

The Environment Agency report also contains guidance that for determination of metals, an in-line filter (e.g. 0.45 µm) should be used immediately on sample collection, and prior to preservation with acid contained within prepared bottles. This is important, since heavy metals in leachate samples are significantly associated with particles, and the particle content will depend on sampling procedure and sample handling (Baun and Christensen, 2004; Kjeldsen et al., 2002). Such filtration is readily achievable on-site, immediately after samples are extracted, using simple disposable sterile syringes and in-line 0.45 µm filter cassettes, to prepare a small sub-sample, which can then immediately be placed into a prepared bottle containing nitric acid for metals preservation.

However, even this is not a foolproof method for accurate determination of many toxic metals in leachates and treated leachates. It has widely been demonstrated that at low concentrations (typically <1 mg/l) many toxic metals are not present in ionic form, but rather are present primarily as organic complexes, with organic compounds present in the leachates (e.g. Baun and Christensen, 2004; Christensen et al., 1999). During the recent studies it was observed that, upon acidification during sample collection and preservation, a scum regularly forms within the acidified sample bottle. This was understood to be a result of the reaction between the organic compounds present in the leachate or treated leachate, and the nitric acid preservative. It has also been noted that significant variations in reported results arise, which depend on whether the analyst routinely filters the acidified sample using a 0.45 µm filter membrane before analysis (as most laboratories do), or whether the entire sample is injected into the ICP-MS plasma flame. Significant variation from this source has been observed (discussed later), for metals such as chromium and nickel, where organic complexation is most important in speciation of the metals.

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