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A comparative study on the efficiency of ozonation and coagulation–flocculation as pretreatment to activated carbon adsorption of biologically stabilized landfill leachate

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ABSTRACT

The present work investigates the potential of coagulation–flocculation and ozonation to pretreat biologically stabilized landfill leachate before granular activated carbon (GAC) adsorption. Both iron (III) chloride ($FeCl_3$) and polyaluminium chloride (PACl) are investigated as coagulants. Better organic matter removal is observed when leachate was treated with $FeCl_3$. At a dose of 1 mg $FeCl_3$/mg COD_o (COD_o: initial COD content), the COD and α_{254} removal was 66% and 88%, respectively. Dosing 1 mg PACl/mg COD_o resulted in 44% COD and 72% α_{254} removal. The settle-ability of sludge generated by PACl leveled off at 252 mL/g, while a better settle-ability of 154 mL/g was obtained for $FeCl_3$ after dosing 1 mg coagulant/mg COD_o. For ozonation, the percentage of COD and α_{254} removal increased as the initial COD concentration decreased. Respectively 44% COD and 77% α_{254} removal was observed at 112 mg COD/L compared to 5% COD and 26% α_{254} removal at 1846 mg COD/L. Subsequent activated carbon adsorption of ozonated, coagulated and untreated leachate resulted in 77%, 53% and 8% total COD removal after treatment of 6 bed volumes. Clearly showing the benefit of treating the leachate before GAC adsorption. Mathematical modeling of the experimental GAC adsorption data with Thomas and Yoon–Nelson models show that ozonation increases the adsorption capacity and breakthrough time of GAC by a factor of 2.5 compared to coagulation–flocculation.

1. Introduction

The generation of landfill leachate is an inevitable consequence of municipal solid waste landfilling. Landfill leachate is high strength waste water generated when waste moisture and rain water percolate through the landfill with the solid waste having a water content above the field capacity (Abdoli et al., 2012). This leachate is characterized by a high concentration of organic matter (chemical oxygen demand (COD) >15,000 mg/L), inorganic ions (chlorides 4500 mg/L) and heavy metals (Nickel 13 mg/L) (Gao et al., 2015; Kjeldsen et al., 2002). These characteristics vary depending on several factors such as waste composition, landfill age and climatic conditions. Furthermore, recent analytical advancements have led to the detection of a wide variety of organic micro-pollutants (Eggen et al., 2010). If not properly collected and treated, landfill leachate can contaminate surface water, ground water and soils. To prevent this, collection, treatment and safe disposal of leachate is mandatory.

Several methods are used to treat landfill leachate. Biological techniques such as sequential batch reactor (SBR), aerated lagoons, activated sludge, up-flow anaerobic sludge blanket (UASB) are preferred in the treatment of young leachate due to their economic competitiveness (Kheradmand et al., 2010; Zhao et al., 2012). However, conventional biological methods are inefficient in the treatment of stabilized leachate. This is due to the low biodegradability (BOD_5/COD <0.1), high ammonia nitrogen (>400 mg/L), and low phosphorus content of stabilized leachate (Abbas et al., 2009; Amokrane et al., 1997; Kim et al., 2007; Li et al., 2010a). Therefore, to achieve the required discharge limits, post treatment of biological effluent is required. Post treatment of biological effluent is preferred because all chemicals and energy used in treatment are

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aimed at removing the bio-recalcitrant fraction. These post treatment techniques are described in detail by (Gao et al., 2015). Briefly, physical–chemical methods such as membrane filtration, ozonation, fenton oxidation, activated carbon adsorption, coagula tion–flocculation, are extensively used in post treatment of landfill leachate.

Adsorption is commonly used in the post treatment of biologically treated landfill leachate (Foo and Hameed, 2009). Though other adsorbents such as zeolites are used, (Amokrane et al., 1997) the most common adsorbent is activated carbon. This is due to its large specific surface area, thermo-stability and fast adsorption kinetics. Furthermore, activated carbon is highly versatile in the removal of a wide range of organic and inorganic pollutants regardless of their concentrations (Turki et al., 2013). Adsorption of biologically treated leachate resulted in 90% COD and 30% NH_4^+-N removal (Gao et al., 2015). Renou et al., 2008 also reports 85% decrease of COD after adsorption of biologically treated leachate. Despite the advantages offered by activated carbon, the operation and regeneration costs are high, which results in increased treatment costs (Chys et al., 2015a) or even forms a barrier for its application (Foo and Hameed, 2010).

Because of its simplicity in operation and implementation, coa gulation–flocculation has been employed successfully in treatment of stabilized leachate (Wang et al., 2009). Coagulation–flocculation facilitates the removal of organic matter and other pollutants from landfill leachate by use of coagulants. However, sometimes moderate COD removal and sludge production are mentioned as drawbacks (Li et al., 2010b). Aluminium sulphate, polyaluminium chloride, ferrous sulphate, and ferric chloride are the most commonly used coagulants (Amokrane et al., 1997). Coagulation–floc culation of leachate was shown to remove 36% NH_4^+ (Vedrenne et al., 2012), up to 87% suspended solids (Li et al., 2010b) and 100% of leachate color (Tatsi et al., 2003). Survey of literature revealed that the efficiency of the coagulation–flocculation technique depends on the type of coagulant, coagulant concentration, pH, temperature, etc. (Rui et al., 2012). Color reduction, organic matter and heavy metals removal provide valuable information in judging the suitability and effectiveness of coagulation–floccula tion in treating landfill leachate. However, few works focus on certain aspects such as settle-ability of produced sludge which is of importance if certain units such as clarifiers are to be added into the treatment train.

Ozonation is one of the advanced oxidation processes(AOP) widely investigated in the treatment of landfill leachate (Gao et al., 2015). This is because molecular ozone is a strong oxidant ($E^O = 2.07$ V) (Kurniawan et al., 2006a) with high reactivity toward recalcitrant organic substances such as humic acids (Cortez et al., 2010). At alkaline pH, in combination with oxidants such as H_2O_2, or in the presence of large amounts of dissolved organic matter; ozone produces hydroxyl radicals (Tizaoui et al., 2007). Hydroxyl radicals have a higher oxidizing potential ($E^O = 2.80$), are non-selective in nature, and thus can rapidly oxidize organic compounds which oxidize slowly with ozone. Furthermore, ozonation facilitates the conversion of complex high molecular weight compounds into simple and more easily biodegradable compounds. This in-turn increases the biodegradability of the leachate (Bila et al., 2005; Chaturapruek et al., 2005). Recirculation of such ozonated effluent into a biological unit shows improved removal efficiency of biological systems and reduced ozone consumption of leachate during subsequent ozonation (Chaturapruek et al., 2005). Application of ozonation is however limited by the high operating costs. For instance, ozonation of 1 m^3 of leachate at pH 3.5 and 743 mg/L initial COD costs 64 € to remove 1 g of COD (Cortez et al., 2010). To achieve 10% COD removal from landfill leachate (initial COD 729 mg/L) 3.1 € was required (Chys et al., 2015a).

Combination of adsorption with either coagulation–flocculation or ozonation can reduce the drawbacks of a single process. For instance, coagulation–flocculation can be used to remove moderate COD hence reducing the COD load to the activated carbon. Subsequently, this reduces the amount of activated carbon consumed. The feasibility of such a combined set up was reported by Li et al. (2010b). In their study, the COD removal increased to 80% after combined treatment as opposed to 70% when only coagu lation–flocculation was used. Granular activated carbon adsorption of ozonated leachate resulted in total COD reduction from 3135 mg/L to 270 mg/L. Furthermore, a reduction in iron from 12.6 mg/L to 0.27 mg/L and a total elimination of nitrites was achieved (Cataldo and Angelini, 2013).

Several studies (Asakura and Matsuto, 2009; Foo et al., 2013a; Papastavrou et al., 2009) use batch adsorption to evaluate the performance of activated carbon. Though batch adsorption studies provide vital equilibrium data on removal of pollutants from landfill leachate, column adsorption studies provide a more realistic representation of the use of carbon adsorption in real practice. Furthermore, batch studies are not effective in studying the uneven flow patterns, non-equilibrium conditions, recycling and regenerating aspects associated with operation of an activated carbon column (Sivakumar and Palanisamy, 2009). Therefore, batch experiments are not appropriate to study the effect of pretreatment techniques on the operating time of activated carbon. For instance (Kurniawan et al., 2006b) uses column studies to demonstrate a 4 times improvement in breakthrough time of COD from GAC, when ozonated leachate is treated using ozone modified GAC. To compare the effect of different types of treatment on the operation time of an activated carbon bed, (Ramirez Zamora et al., 2000) utilized coagulation–flocculation and Fenton oxidation as pre-treatment steps before fixed bed activated carbon adsorption. Results showed that the operation time of the activated column fed with coagulation–flocculation effluent was extended by a factor of 1.3 compared to the column fed with Fenton oxidation effluent. Therefore, coagulation–flocculation of leachate results in less activated carbon columns used, and as such less operational costs. However, the costs incurred in disposal of the large amounts of sludge formed from coagulation–flocculation and Fenton oxidation can nullify the benefit of saved costs.

Based on the discussion above it can be concluded that there is a need for a technique which provides optimum removal efficiency while extending the operation time of the activated carbon step. It is in this regard that a comparative study on ozonation and coa gulation–flocculation is presented in which both techniques are combined with a granular activated carbon (GAC) column for efficient removal of organic matter. The pretreatment steps will be evaluated based on their removal efficiency and their effect on activated carbon column properties. Specifically, evaluation of the pretreatment steps and their combined use with GAC will be based on COD and α_{254} removal. Attention will also be paid to the settle-ability of the sludge produced during coagulation–floccu lation. The Thomas and Yoon–Nelson adsorption model will be used to predict the effect of ozonation and coagulation–floccula tion on adsorption capacity and breakthrough time of the activated carbon column.

2. Materials and method

2.1. Leachate sampling and characterization

Biologically treated leachate was collected from the IMOG municipal landfill leachate treatment facility in Moen (Belgium). The on-site treatment facility and sampling procedures are described by Chys et al. (2015a). The main characteristics of the

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