Journal of Environmental Management 228 (2018) 189–196

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Removal of inert COD and trace metals from stabilized landfill leachate by granular activated carbon (GAC) adsorption

Ehsan Mohammad-pajooh[a,*], Ariel E. Turcios[b], Graham Cuff[a], Dirk Weichgrebe[a], Karl-Heinz Rosenwinkel[a], M.D. Vedenyapina[c], L.R. Sharifullina[d]

[a] *Institute for Sanitary Engineering and Waste Management, Leibniz University Hannover, Appelstr. 9a, D-30167 Hannover, Germany*
[b] *Institute of Botany, Leibniz University Hannover, Herrenhäuserstr. 2, D-30419 Hannover, Germany*
[c] *Zelinsky Institute of Organic Chemistry, Russian Academy of Sciences, Leninskii Pr. 47, Moscow, 119991 Russia*
[d] *Mendeleev University of Chemical Technology, Moscow, 125047 Russia*

ARTICLE INFO

Keywords:
Leachate treatment
Adsorption
Activated carbon
Trace metals
COD reduction
Dissolved air floatation

ABSTRACT

Landfills in Germany are currently approaching stabilization phase; as a result removal of inert organics and potentially toxic elements in the leachate is becoming a primary concern. Dissolved air floatation (DAF) at the secondary stage reduces only 27% of the residual chemical oxygen demand (COD) in the investigated treatment systems; downstream granular activated carbon (GAC) units are required to further reduce COD concentration by 40–56% to meet indirect discharge or direct discharge limits respectively. Therefore, in this study performance in terms of COD and trace metals adsorption of different types of granular activated carbon were compared over different contact times and dosages. GAC 1 with Brunauer-Emmett-Teller (BET) surface area of 719.5 ± 2.1 m^2/g and average pore diameter (D) of 4.81 nm was identified to be inappropriate for treatment of leachate from this landfill. GAC 2 (with BET of 1513.7 ± 6.4 m^2/g and D of 3.50 nm) was feasible for COD reduction from DAF-pretreated leachate, while GAC 3 (with BET of 644.5 ± 2.6 m^2/g and D of 5.65 nm) can be coupled either with biological step alone, or as a tertiary step after the DAF unit. Moreover, as COD is the primary remaining contaminant of interest after secondary and tertiary treatment, spectrometer probes provide a close estimation of COD concentration for use in online monitoring. Beside COD removal, GAC 3 also confirmed the effectiveness of trace metals adsorption even at trace level, as it removed 66, 64, 48, 47, 43, and 25% of copper, cobalt, chromium, manganese, nickel, and zinc, respectively.

1. Introduction

Municipal landfill leachate is often heavily contaminated in terms of organics (i.e. BOD_5, COD, and AOX), nitrogen (mainly in the form of NH_4-N and NO_3-N) and potentially toxic elements and thus requires treatment to avoid pollution of groundwater and surface waters. Characteristics of leachate (i.e. composition, volumes, and fluctuations) vary greatly due to numerous variables such as climatic conditions (i.e. changes in temperature and precipitation) (Chu et al., 1994), type of deposited waste, landfill design (Baig et al., 1999; Fitzke et al., 2013) and degree of stabilization of deposited waste (e.g. in Germany since 2005, waste must be pre-treated and relatively stabilized prior to landfilling).

Previous investigations have examined the transition in landfill leachate quality (as a result of waste pre-treatment), types of current leachate treatment processes and their energy demand in different states of Germany (Mohammad-pajooh et al., 2017). Fig. 1 compares the average concentration of ammonium (NH_4-N) and biodegradable organic matter (measured as 5-day biological oxygen demand BOD_5), as well as biodegradability ratio (BOD_5/COD) of raw leachate among three current landfills in Germany (in 2013), all of which pre-treat the waste before deposition. In these landfills, NH_4-N and BOD_5 concentrations range between 250–1150 mg/L and 50–250 mg/L respectively, while BOD_5/COD and BOD_5/N ratios are below 0.12 and 0.3 respectively. Such features imply that in the current landfills: (1) organic pollution in leachate derives primarily from non-biodegradable organic matter, (2) C/N ratio of the leachate is not favorable for aerobic treatment, and (3) the role of the biological unit in current leachate

* Corresponding author.
E-mail addresses: pajooh@isah.uni-hannover.de (E. Mohammad-pajooh), a.turcios@botanik.uni-hannover.de (A.E. Turcios), cuff@isah.uni-hannover.de (G. Cuff), weichgrebe@isah.uni-hannover.de (D. Weichgrebe), rosenwinkel@isah.uni-hannover.de (K.-H. Rosenwinkel), mvedenyapina@yandex.ru (M.D. Vedenyapina), alishari@yandex.ru (L.R. Sharifullina).

https://doi.org/10.1016/j.jenvman.2018.09.020
Received 17 May 2018; Received in revised form 2 September 2018; Accepted 4 September 2018
0301-4797/

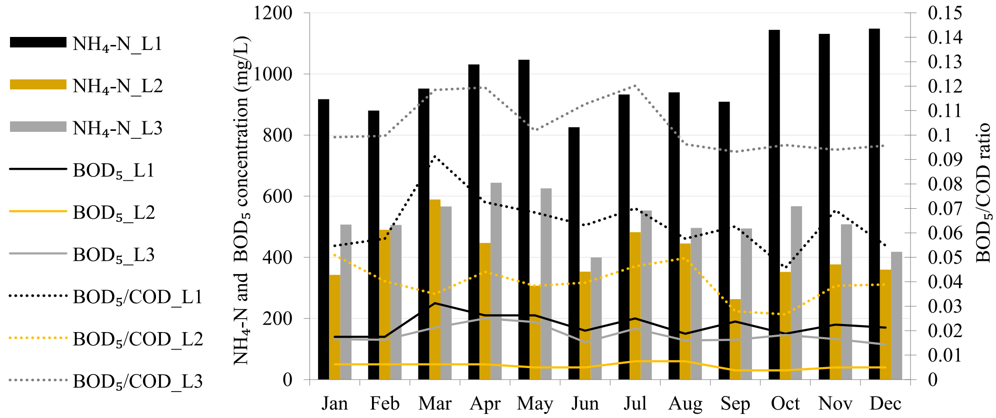

Fig. 1. Average BOD_5 and NH_4-N concentrations, BOD_5/COD ratio and their temporal variation in 3 landfills (L1, L2, and L3).

treatment plants is mainly nitrogen removal.

As many current landfills are undergoing a stable methanogenic phase and/or approaching a final aerobic phase then the aerobic treatment of leachate is not cost effective and therefore a better approach is to retrofit existing biological units with the deammonification process to reduce the annual operational costs and emissions (Mohammad-pajooh et al., 2017). Deammonification was first discovered in the early 1990s in a rotating biological reactor (RBC) configuration and subsequently well established through moving bed bio reactor (MBBR) configurations (Hippen et al., 1997; Rosenwinkel and Cornelius, 2005). Compared to conventional nitrification (conversion of nearly 100% of NH_4-N to NO_3-N with the aid of aeration and autotrophic bacteria) and denitrification processes (conversion of NO_3-N to N_2 in under anoxic conditions using soluble degradable organics and heterotrophic bacteria), the deammonification process is more economical, as it reduces aeration demand (only about 50% of NH_4-N is oxidized), sludge production, and the need for external carbon sources (Christensson et al., 2013; Jenkins and Wanner, 2014). However, to meet discharge standards the effluent from deammonification processes must be further treated to remove remaining NO_3-N and COD. Both the low amounts of NO_3-N which exist in raw leachate and the residual NO_3-N which is formed during the deammonification process can be removed in post denitrification by addition of an external carbon source (e.g. methanol), while COD and the remaining impurities (e.g. trace metals) can be separated in a secondary treatment step using a combination of various chemical-physical treatment processes (such as coagulation/flocculation or membrane filtration). Generally, the biological unit is able to remove between 30 and 50% of the COD; depending on the employed technology in the secondary stage, COD removal could reach up to 85% as in the case of coagulation/flocculation or ultra-filtration (Mohammad-pajooh et al., 2017; Radeck, 2015). At this stage, remaining COD consists of mainly humic substances (e.g. humic and fulvic acids) and is considered to be non-biodegradable (Gupta et al., 2014). Therefore, in case of indirect discharge and depending on volume and total loads, residual COD may reduce water quality in terms of turbidity and color and therefore lower the efficiency of UV-disinfection at wastewater treatment plants (WWTPs), see Appendix.

Effective UV-disinfection in WWTPs is achieved near wavelengths around 254 nm and is accomplished via penetration of ultraviolet energy through the outer cell membrane of the organism, passing through the cell body, and finally disrupting the DNA and preventing its reproduction (Alkan et al., 2007). Studies have shown that high molecular weight aromatic organic compounds from municipal landfill leachate such as humic acids (i.e representing dark-brown to black color) and fulvic acid (i.e representing light yellow to golden brown color) absorb UV light and therefore interfere with the disinfection process (Alkan et al., 2007; Pathak et al., 2017; Reinhart and Bolyard, 2015; Van Zomeren, 2008). Therefore, COD which remains in solution after secondary treatment and contains non-biodegradable compounds must be polished along with the remaining impurities (e.g. AOX, trace metals) by activated carbon in a tertiary/final step.

Activated carbon is utilized in landfill leachate treatment schemes both in powder -(PAC)- and granular (GAC) forms with average particle sizes between 15 and 25 μm and 0.2–5 mm, respectively (Çeçen and Aktas, 2011). However, GAC is applied more often, since unlike PAC it does not require coagulation/flocculation to be precipitated/separated (Stegmann et al., 2005). The number of activated carbon vessels in a given landfill leachate treatment plant depends on: (1) the amount of generated leachate, (2) the residual COD concentration from the chemical-physical treatment processes at the secondary stage, and (3) the desired effluent quality/target of treatment (i.e. direct or indirect discharge). Generally, between 2 and 6 GAC vessels are operated in the final phase of the treatment process; to ensure reliable operation they are often designed in parallel (e.g. 2 lines with 3 GAC units in each line) and operated in series. Loads (in terms of pollutant concentrations) are higher and the service life is shorter in the initial GAC units. Each vessel has a volume of 20–30 m^3, and depending on flowrate from secondary treatment, the pre-treated leachate will have a minimum contact time of 8–10 h with the GAC unit(s). For economic reasons and to avoid unnecessary loss of capacity of the GAC units, some plants have single or multiple low cost filtration units (e.g. bag filter, sand filter, gravel filter) to trap solids and further reduce the pollution loading to the GAC vessels. Moreover, additional storage units may be considered for re-use (e.g. backwash of membrane units) or for temporary storage of pre-treated leachate in cases where GAC effluent does not satisfy discharge requirements.

Among 54 landfill leachate treatment plants in Germany, 39 use activated carbon (37 GAC and 2 PAC) at a final treatment stage and in few cases for concentrate treatment (e.g. from membrane technologies such as nano-filtration). For these installations GAC treatment represented the third highest cost after aeration and external carbon supply. Therefore, identifying a GAC with higher treatment efficiency will reduce annual leachate treatment costs and also outflows of residual COD and trace metals, thereby mitigating negative impacts on wastewater treatment plants, water bodies and the surrounding environment. However, there is a scarcity of literature available with respect to GAC performance treating leachate from pre-treated waste, particularly under controlled conditions. Therefore the adsorptive and performance efficiency of GAC removing COD and other impurities such as trace metals has been determined for multiple GAC types, in order to identify optimal GAC characteristics and better inform future leachate treatment strategies.