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Unraveling complex removal behavior of landfill leachate upon the treatments of Fenton oxidation and MIEX® via two-dimensional correlation size exclusion chromatography (2D-CoSEC)

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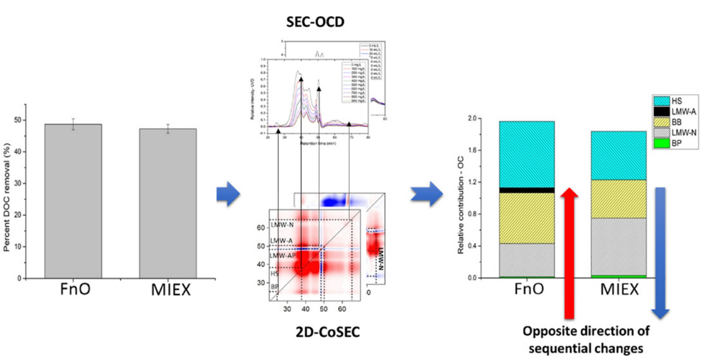
GRAPHICAL ABSTRACT



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ABSTRACT

The complex removal behavior of stabilized landfill leachate was explored for the treatments of Fenton oxidation (FnO) and magnetic ion exchange (MIEX®) resin using two-dimensional correlation size exclusion chromatography (2D-CoSEC) and fluorescence excitation emission matrix-parallel factor analysis (EEM-PARAFAC). The overall removal rates of the bulk parameters (~45% for dissolved organic carbon and ~78% for UV absorbance) were similar between the two treatment options, while distinct differences were found with respect to different molecular sizes and chemical composition. The resin treatment eliminated humic substances (HS) and low molecular weight acid (LMWA) fractions to a greater extent than other fractions (i.e., HS: 62% and LMWA: 99%), while low molecular weight neutral (LMWN) and biopolymers (BP) fractions were more effectively treated by the FnO with the removal rates of 56% and 92%, respectively. The 2D-CoSEC further revealed that the sequential or preferential changes of different size fractions with increasing the resin or H_2O_2 were opposite between the two treatment options in the order of HS → LMWA → LMWN → BP for MIEX®. Due to their complementary roles in treating leachate, the combined processes removed a wider ranges of different molecular sizes compared to the single operation.

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1. Introduction

Rapid urbanization and industrialization have led to the production of a tremendous amount of municipal solid wastes each year over the world (1.3 billion tons as of the year 2010) [1]. Most of these wastes are ultimately disposed in landfills, which can limit their hazard to public health. To date, landfills have been regarded as the most economical solution for solid waste disposal [2]. However, the continuous production of leachate acts as a long-term obstacle for the environmentally safe operation of landfills. As a high strength wastewater, landfill leachate typically contains a wide array of different refractory organic compounds, including humic substances (HS) which are difficult to be removed by conventional treatment processes [3]. The presence of these organic compounds in natural environment can be responsible for aesthetic concerns like color/odor and affects the subsequent treatment processes (e.g., membrane fouling and/or clogging of adsorption media) [4,5]. With the increase of landfill age, such non-biodegradable organic matters tend to be enriched in stabilized landfill leachate, heightening the demand of an effective physio-chemical treatment for the control [6,7].

A number of the physio-chemical treatment options have been practiced for landfill leachate, which encompass coagulation/flocculation (C/F), adsorption by activated carbon, chemical oxidation, ion exchange, and membrane filtration [8–11]. Among those, Fenton oxidation (FnO), an advanced oxidation process to generate reactive ·OH radicals via the interactive reactions of H_2O_2 and Fe(II), has been proved to be highly potent in removing refractory organic matter from landfill leachate [12–15]. However, FnO alone cannot sufficiently deal with all the constituents contained in landfill leachate as discussed in a recent review article [16]. Furthermore, Fenton sludge production is another demerit for the sustainable operation. For this reason, FnO has often been compared or coupled with other treatment options to enhance the organic removal or decrease the required FnO chemical doses [17–19]. C/F and granular activated carbon (GAC) are the most frequently practiced alternative options to the FnO in treating landfill leachate [20–22]. GAC adsorption has been widely used to remove low molecular weight (LMW) organic fractions [23]. Relatively mature landfill leachate likely contains a large amount of HMW refractory organic fraction [24], which are not readily removed by the GAC pretreatment, consuming a high dose of GAC. Meanwhile, the usage of a large amount of coagulants and the subsequent sludge production are viewed as the major demerit of combining FnO with coagulation [25,26]. In general, FnO consist of two steps (i.e., the oxidation followed by coagulation with the Fenton sludge containing iron) [27,28].

In this regard, MIEX®, a strong base anion exchange resin, can be a good replacement for C/F and GAC adsorption owing to many advantages including its removal tendency towards a wide range of different leachate constituents, high surface area, effective recovery via gravity settling and/or magnetic separation, fast removal kinetics, and high regeneration efficiency [29–31]. The MIEX® resin possess a high regeneration efficiency and 2 ~ 5 times smaller particle size than other ion resins, resulting in a fast kinetics [32]. In addition to drinking water treatment applications, the resin has also been successfully applied for wastewater and landfill leachate [29,33,34]. For example, Boyer et al. [35] and Singh et al. [36] have demonstrated the superior treatment capability of MIEX® over C/F for a stabilized landfill leachate.

Considering the heterogeneous presence of different constituents in landfill leachate, the bulk parameter, mostly represented by dissolved organic carbon (DOC), may provide only limited information to obtain insight into the optimization of the treatment processes and the involved mechanisms. Landfill leachate can be conventionally fractionated according to its hydrophobic nature into humic/fulvic acid and hydrophilic fraction [37]. Recently, size exclusion chromatography (SEC) and excitation emission matrix combined with the parallel factor analysis (EEM-PARAFAC) have emerged as advanced tools to explore the heterogeneous character of dissolved organic matter (DOM) [38–40]. For example, EEM-PARAFAC has been successfully applied to track the individual behaviors of different fluorophores in the FnO process of treating landfill leachate. FnO can preferentially oxidize the refractory humic-like fluorescence components [14,41,42]. The removal behavior of different size fractions upon FnO processes were also studied for landfill leachate. FnO typically targets at HMW fractions in leachate, converting them into readily biodegradable fractions [14,43]. To date, however, the applicability of the two DOM analyses has yet to be utilized for MIEX® resin treatment of landfill leachate. Moreover, the combined processes of MIEX® and FnO have not been explored for the treatment of wastewater including landfill leachate.

Two-dimensional correlation spectroscopy (2D − COS), an advanced mathematical tool, has proven its strong potential to suggest sequential changes of different constituents in DOM upon certain reactions applied to given systems [44–46]. Although 2D − COS has been mostly coupled with the spectroscopic data of DOM, its linking to size exclusion chromatography (SEC), namely, two-dimensional correlation SEC (2D − CoSEC), can allow to the acquisition of detailed information of the DOM changes upon certain treatment processes with respect to molecular size [45,47]. For example, Phong and Hur. demonstrated a successful application of the 2D-CoSEC in examining the adsorptive behavior of DOM onto nanoparticles in terms of molecular sizes [47]. The applicability of 2D-CoSEC has yet to be tested for the treatment processes of landfill leachate.

In this study, stabilized landfill leachate was treated with the individual processes of FnO and MIEX®. The complex and heterogeneous removal behavior of leachate behavior was explored in terms of different molecular size and dissimilar fluorophores, and was compared for the two treatment options as well as their combinations. The specific objectives of this study were to (1) compare the preferential removal tendency of the two treatment options with respect to different constituents of leachate via SEC and EEM-PARAFAC, and (2) investigate the potential of 2D-CoSEC to unravel the complex removal behavior of the processes and the underlying mechanisms.

2. Materials and methods

2.1. Sampling and the basic water quality parameters of leachate

Raw landfill leachate (40 L) was collected in September 2017 from a stabilization tank of an on-site landfill leachate treatment facility (Sudokwon landfill site), located near the city of Incheon, South Korea. The landfill was operated for 8 years and closed from 2000 to the present. The sample was immediately transported to the laboratory, and stored in a refrigerator at 4 °C. Before treatment experiments, the sample was passed through the pre-ashed Whatman GF/F glass fiber filters (0.7 µm). The basic water quality parameters of the leachate are listed in Table S1.

2.2. FnO and MIEX® experiments

FnO experiments were performed in 500 ml beakers with a working volume of 200 ml. The raw leachate sample (200 ml) was placed on a magnetic stirrer (400 rpm) and the pH was adjusted to desirable values (3–9) by adding 2 N HCl or NaOH solution. A calculated amount of iron sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$, Sigma Aldrich) was dosed to the sample, followed by the single addition of all required H_2O_2 (Hydrogen peroxide, 33% assay, Daejung) to initiate the Fenton reaction. The amount of iron sulfate added in the sample was calculated according to the desired mass ratios of H_2O_2 (mg/L) to Fe (mg/L). After 1 h of mixing, the pH was adjusted to ~9.0 to cease the Fenton reaction [14,48]. The sample was moved to a jar test apparatus (20 rpm for 20 min). Residual H_2O_2 was measured by spectroscopic method using metavanadate [49] as this method had no effect of Cl^- and Fe ions on the measurement. The standard curve for measuring the residual H_2O_2 by the vanadate method is shown in Fig. S2. The residual H_2O_2, which