Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2015.1060175

57 (2016) 13608–13614
June

Taylor & Francis
Taylor & Francis Group

Removal of dissolved organic nitrogen by using biological activated carbon and MIEX treatment

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Received 1 March 2015; Accepted 30 May 2015

ABSTRACT

The performance of biological activated carbon (BAC) and magnetic ion exchange resin (MIEX) has been evaluated for the removal of dissolved organic nitrogen (DON) in secondary wastewater effluent (SWWE). Their effectiveness was analyzed both individually and in combination. BAC removed DON up to 42%, while MIEX appeared to be relatively better and removed up to 60%. Their effectiveness was further investigated using their combinations. Result showed that MIEX followed by BAC treatment provided no further removal of DON but complete removal of DON was observed on reversing the combination order (BAC/MIEX). Further analysis showed that although BAC offers less DON removal, it has an ability to produce amenable DON fraction. When BAC is followed by MIEX treatment, there is an availability of additional MIEX-amenable DON to be removed by subsequent MIEX treatment. This particular mechanism remains unexploited upon reversing the combination (MIEX/BAC). As a result, BAC/MIEX combination order appeared to be more effective than MIEX/BAC combination order for the removal of DON from SWWE.

Keywords: Dissolved organic nitrogen; Biological activated carbon; Magnetic ion exchange resin; Secondary effluent

1. Introduction

Application of nitrification–denitrification process in municipal wastewater treatment leads to the formation of dissolved organic nitrogen (DON) in secondary wastewater effluent (SWWE). DON in SWWE is a complex and composed of various nitrogenous compounds such as urea, amino acids, low molecular weight amines, and chelating agents. However, these identified compounds in DON sum up only around 10% of bulk DON, while rest is yet to be characterized [1].

Presence of DON in SWWE affects the subsequent treatment process in many ways. It acts as a precursor for the formation of carcinogenic disinfection byproducts (DBPs) such as haloacetonitriles, halonitromethane, and N-nitroso-dimethylamine (NDMA) during chlorination [2]. Among them, NDMA is highly hydrophilic in nature and even RO membrane is not fully effective to reduce its concentration down to a desired level in treated water [3]. DON also acts as membrane foulant and decreases the efficiency of membrane filtration process. Many proteinaceous compounds have been identified as one of the major foulants by many studies [4–6]. It is also one of the major causes of water quality degradation by eutrophication of receiving water bodies [7]. Thus, removal of DON is an essential step in advanced wastewater treatment process intended to use for direct or indirect recycling application.

Many physicochemical treatment processes such as coagulation, activated carbon, and advanced oxidation were evaluated earlier for the removal of dissolved organic matter (DOM). However, previous studies paid less attention for the removal of DON as it comprises relatively small fraction of DOM and there is limited literature available that deals with the removal of DON alone [8]. The efficiency of coagulation using alum salt has been investigated in SWWE and shown DON removal up to 60%. However, the applied alum dose was significantly higher (250 mg/L) [9]. In another study with the wastewater effluent-containing melanoidin, the maximum removal of DON reached only up to 42% by 30 mg/L alum [10]. Lee and Westerhoff studied alum coagulation in combination with cationic polymer (polydiallyldimethyl-ammonium chloride) which shown to improve the DON removal efficiency by 15–20% [2].

Activated carbon is one of the widely used adsorbents for the successful removal of DOM in water and wastewater treatment process [11]. However, its performance against DON appeared to be less effective for its hydrophilic character that has lower tendency of adsorption onto activated carbon [1]. Similarly, advanced oxidation treatment process such as UV/H_2O_2 also appeared less promising and removed only up to 25% DON from the wastewater-containing melanoidins [12].

These studies clearly show the limitation of different conventional treatment method for the removal of DON and hence, still a great challenge for water industry. However, previous studies have also shown the effectiveness of biological treatment for the removal of DON in activated sludge treatment process [13]. Similarly, microbial activity in activated carbon bed has been reported for the effective removal of NDMA; a DBP produced due to DON precursor [14,15]. Magnetic ion exchange resin (MIEX) has been increasingly used to remove the anionic fraction of DOM [16–18] that could also support the removal of DON fraction. In this context, the performance of biological activated carbon (BAC) and MIEX for the removal of DON from the SWWE has been investigated.

BAC is an extended use of activated carbon even after the exhaustion of physical adsorption sites. Beds and pores of activated carbon start to serve as a habitat for bacteria after the entrapment of organic matter. Such organic matters approaching to the bed act as a source of nutrients for microbial community. This helps to establish a natural biofilm and regenerates the activated carbon while carbon bed is in operation [19,20]. As a result, the life of activated carbon extends significantly and makes it more economical [21]. MIEX, on the other hand, is a resin-based treatment. It is a micro-sized strong base resin with ammonium functional group, consisting of a macroporous, polyacrylic structure [17]. It effectively removes the DOM that supports its anion exchange properties [22,23].

While the use of BAC and MIEX both individually and in combination have been reported for the effective removal of DOM in previous studies [24–26], their performance against DON is yet to be understood. Thus, this study aims to evaluate the individual performance of BAC and MIEX for their ability to remove DON from SWWE. In addition, it evaluates whether BAC and the MIEX treatment in combination support each other for the enhanced DON removal that would be greater than the sum of two individual treatment processes.

2. Material and methods

SWWE (dissolved organic carbon (DOC) and DON concentrations, respectively, 7.7 and 1.33 mg/L) was used in this study. The individual and combined performance of BAC and MIEX were investigated first and followed by different orders of their combination as presented in Fig. 1.

2.1. Method 1: BAC treatment

Granular activated carbon (GAC) having 2–3 mm in size obtained from Rowe Scientific, Australia was packed into a column (diameter of 3.9 cm) to a depth of 15 cm (~180 cm^3 of GAC). Six identical reactor columns were used in the study. Each column was provided with three different ports to be used as the feed port, backwash, and effluent discharge. The micro-organisms in the SWWE naturally seeded the GAC filter bed. The performance of activated carbon filter was continuously monitored by measuring DOM concentration in order to examine whether the BAC filter had attained a steady state of operation. The BAC filter column was operated as a continuous flow reactor. The flow on the reactor was regulated using a peristaltic pump with a variable speed. The speed of the pump and corresponding flow rate were adjusted to achieve an empty bed contact time (EBCT) of 40 and 120 min (B120). In order to obtain EBCT of 240 min (B240), the B120 treated effluent was recirculated through the same BAC filter. Additional information on reactor setup could be found elsewhere [11,27].

2.2. Method 2: MIEX treatment

The regenerated MIEX resin (obtained from Orica Watercare, Australia) was used in this experiment. The resin was composed of 150–180 μm size beads

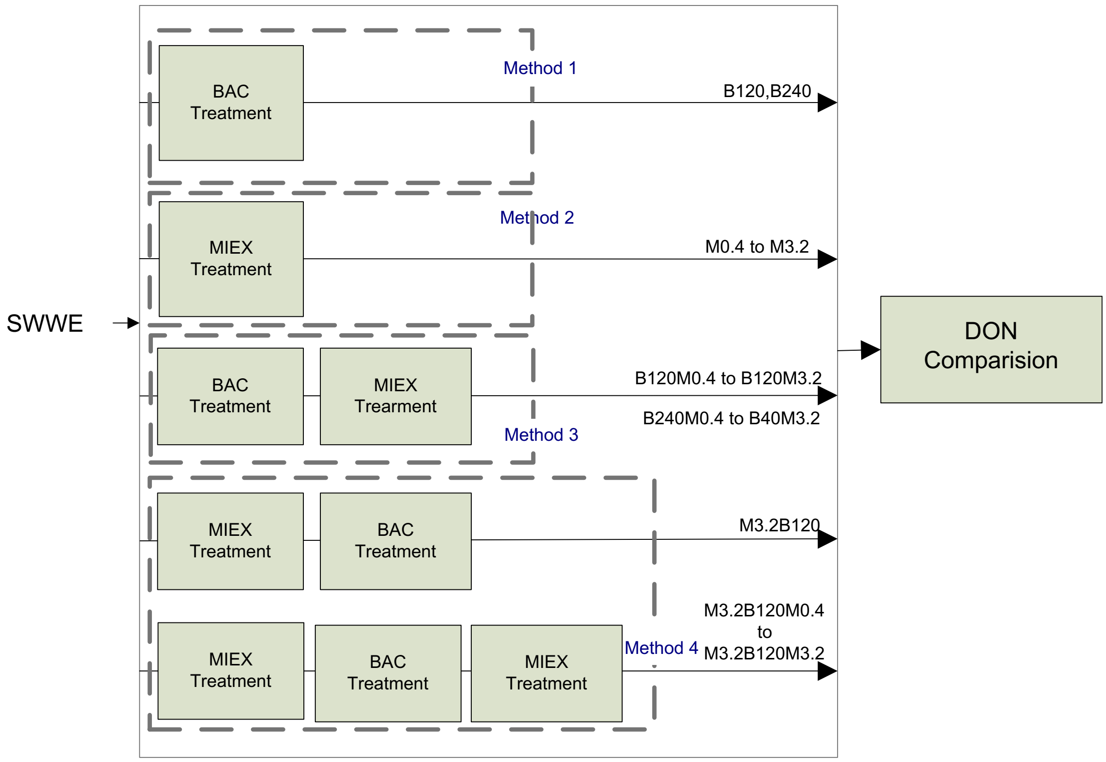

Fig. 1. Experimental flow chart.

and with initial concentration of 90% v/v (based on settled resin volume). Prior to each use, MIEX resin was regenerated using solution of sodium chloride (10% w/v). MIEX resin (100 ml) was mixed with sodium chloride aqueous solution (600 ml) and mixed at 150 rpm for 30 min followed by 5 min settling time. The sodium chloride solution was then decanted. Afterwards, RO-treated water (600 ml) was added to MIEX resin and stirred for another 10 min. The RO water was then decanted and the regenerated settled resin was used for the treatment of SWWE.

Five different resin concentrations (0.4, 0.8, 1.2, 1.6, and 3.2% (M0.4, M0.8, M1.2, M1.6, and M3.2, respectively)) were applied under the same mixing protocol and settling time used in this study.

2.3. Method 3: combined BAC/MIEX treatment order

This order of combination comprised of BAC treatment followed by a subsequent MIEX treatment. SWWE was first treated with BAC at an EBCT of 120 and 240 min. Afterwards, they were individually subjected to various MIEX doses (0.4–3.2% v/v) (B120M0.4-B120M3.2 and B240M0.4-B240M3.2).

2.4. Method 4: combined MIEX/BAC and MIEX/BAC/MIEX treatment order

This order of combination comprised of MIEX treatment followed by BAC treatment. In order to understand whether BAC removes the DON fraction that cannot be removed by MIEX, the resin dose was progressively increased until the residual DON remained constant. It was then filtered through 0.45-μm filter paper to avoid the entering of residual MIEX resin into BAC bed. The pH (7.3) and phosphorus concentration (1.7 mg/L) were also measured and ensured they will not affect the microbial activities in BAC bed. Further to avoid contamination from previous run, the BAC bed was first flushed with 2 L of RO water followed by MIEX-treated effluent itself. Afterwards, the MIEX-treated water (only with MIEX non-amenable fraction) was subjected to BAC treatment at an EBCT of 120 min (M3.2B120). The first 2 L of sample was discarded prior to sampling for DON analysis. It was further treated with MIEX (M3.2B120M0.4–M3.2B120M3.2) again to determine whether these two treatments support each other for the enhanced DON removal from the SWWE.

3. Analytical measurements

The DON concentration was calculated by obtaining the difference between total dissolved nitrogen (TDN) and dissolved inorganic nitrogen (DIN) using the following equation

$$DON = TDN - NO_3^- - NO_2^- - NH_3/NH_4 \quad (1)$$

TDN was measured using Shimadzu TOC-V_{CSH} analyser (high temperature combustion at 720˚C; non-dispersive infrared detection) with a TNM-L unit (Chemiluminescence detection) (Shimadzu Corporation, Japan). The measurement of NH_3, NO_2^-, and NO_x (NO_2^- + NO_3^-) were carried out using an Aquakem 200 (Thermo Scientific, Finland), high precision wet chemistry analyzer. NH_3 and NO_x were measured spectrophotometrically according to the method illustrated in DoE [28]. NO_2^- was measured by the sulfanilamide method (4500-NO_2^-B) [29]. The concentration of NO_3^- was then calculated from the difference between NO_x and NO_2^-. All the samples were first filtered through 0.45-μm pore size filter media (GE water and process technologies, Cat. No. A04SP04700, Acetate plus supported) prior to measurement of both TDN and DIN. Four replicate measurements were carried out for each sample and average values are presented with standard deviation.

4. Results and discussion

4.1. Source water

The SWWE from Beenyup wastewater treatment plant (BWTP), Western Australia was used in this study. It uses a secondary treatment (activated sludge) process for the removal of nitrogen and DOC from the influent wastewater. The plant has no provision for chemical or biological methods for the removal of phosphorus. The general characteristic of SWWE from this plant is presented in Table 1.

4.2. Individual performance of BAC and MIEX

Fig. 2 clearly shows that both treatments have the ability to remove DON from SWWE. When the BAC treatment was undertaken at an EBCT of 40 min, DON concentration reduced from 1.33 to 1.14 mg/L that accounts only around 15% removal. The DON concentration was further decreased with increased EBCT (120 min) in the BAC bed and obtained around 0.8 mg/L (overall 42% removal). However, additional EBCT (240 min) afterwards did not show further improvement. This indicates that the microbial activity at 120 min EBCT is sufficient for the removal of biodegradable DON fraction. Compared to BAC treatment, MIEX showed its ability to remove more DON. As can be seen from Fig. 2(b), MIEX dose of 1.2% v/v reduced DON concentration from 1.33 to 0.54 mg/L (60%). Although, MIEX dose was increased up to 3.2% v/v, further reduction in DON remained very marginal. Compared to the current practice (Wanneroo Ground Water Treatment Plant, Perth, Western Australia) this is indeed a small amount for the treatment of SWWE. This plant normally uses 2.0% v/v MIEX for the removal of DOC 8–10 mg/L for the production of drinking water [24]. This clearly shows that the MIEX dose under practice could remove up to 60% DON while applying for the removal of DOC.

4.3. Combined performance of BAC/MIEX treatment order

Although both BAC and MIEX were able to remove DON from SWWE, both of them left significant fraction of residual DON that could still be detrimental for a post-treatment process such as disinfection and membrane filtration process. Previous studies showed that microbial activities in BAC treatment has an ability to change the characteristics of secondary effluent which makes subsequent physical treatment process more efficient in terms of DOC removal [11,18]. Thus, in order to understand whether the same mechanism enhanced the performance of

Table 1
General characteristics of SWWE

Parameters	pH	mg/L						
		DOC[a]	TN[b]	NH_4^+-N	NO_2^--N	NO_3^--N	DON[c]	PO_4^{3-}-P
Average	7.32 ± 0.2	7.70 ± 0.35	12.662 ± 0.341	0.069 ± 0.011	0.002 ± 0.00	11.115 ± 0.216	1.333 ± 0.341	9.391 ± 0.731

[a]Dissolved organic carbon.
[b]Total nitrogen.
[c]Dissolved organic nitrogen.

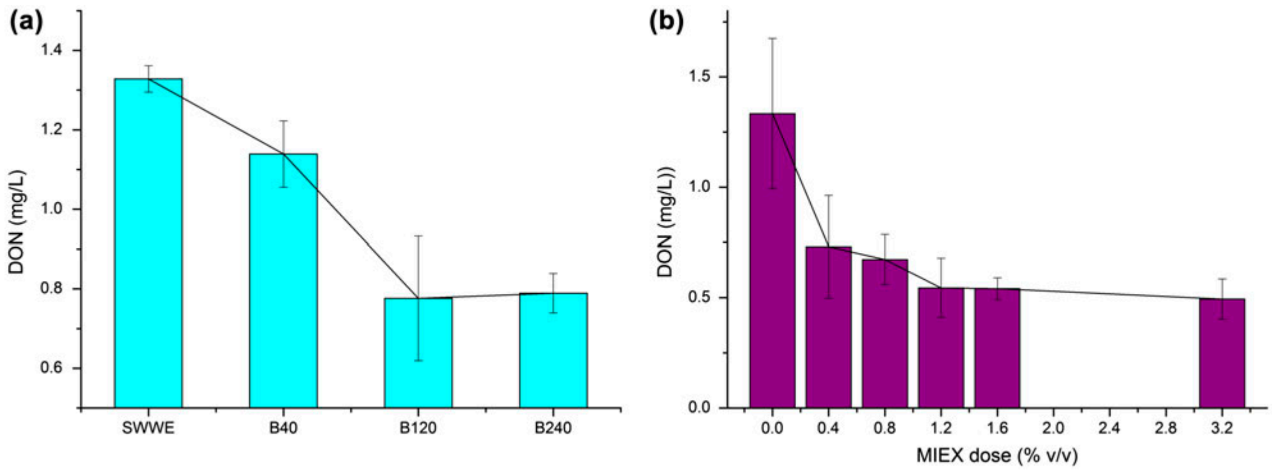

Fig. 2. Individual performance of (a) BAC and (b) MIEX for the removal of DON.

DON removal, SWWE was first treated with BAC (at two different EBCT of 120 and 240 min) followed by the MIEX treatment.

As presented in Fig. 3, prior to BAC treatment, there was no further DON removal in SWWE and remained constant around 0.5 mg/L with increased MIEX dose (SWWE + MIEX). However, once it was treated with BAC at an EBCT of 120 min, the residual DON that remained constant with the MIEX dose of 1.2% v/v, further started to decrease continuously and no DON was detected when MIEX dose was increased to 3.2% v/v. This phenomenon was further investigated with extended BAC treatment (EBCT 240 min). Result showed improvement in DON removal indicated the possible role of microbial activities in BAC bed for the enhancement of subsequent MIEX

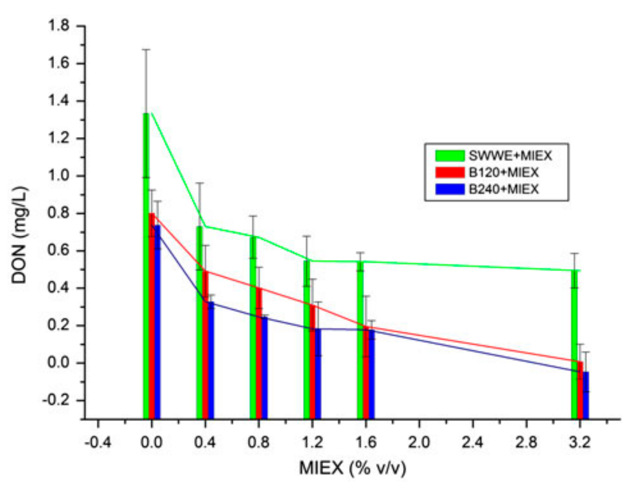

Fig. 3. Combined performance of BAC/MIEX treatment order.

treatment process. However, BAC and MIEX individually removed 0.5 and 0.8 mg/L, respectively, out of 1.33 mg/L. While the sum of the removal of these two treatments is equal to the removal obtained in BAC/MIEX treatment, it is not yet clear that whether BAC has really enhanced the subsequent treatment process or not.

4.4. Combined performance of MIEX/BAC and MIEX/BAC/MIEX treatment order

To further understand whether the DON removal during BAC/MIEX treatment is only a summation of two individual treatments or BAC influences subsequent MIEX treatment process, the treatment order was reversed to MIEX/BAC. During this, the MIEX dose was increased until residual DON concentration in SWWE remained constant. In order to ensure there is no MIEX amenable fraction left, SWWE was treated with MIEX dose of 3.2% v/v, although no further removal was obtained beyond the MIEX dose of 1.2% v/v. Contrary to BAC/MIEX order, no further removal of DON was obtained when MIEX-treated water (3.2% v/v) was subjected to BAC treatment (Fig. 4; MIEX/BAC red shading part). This clearly indicates that the DON fraction that BAC can remove has already been removed by MIEX. Thus, when BAC treatment was carried out on MIEX-treated water, probably there was no biodegradable DON fraction remained to be removed. This clearly showed that the BAC/MIEX combination order is more effective than MIEX/BAC order; however, the influence of BAC treatment over MIEX still remained unexplained.

MIEX/BAC treated water was further subjected to MIEX treatment. It was hypothesized that if the BAC

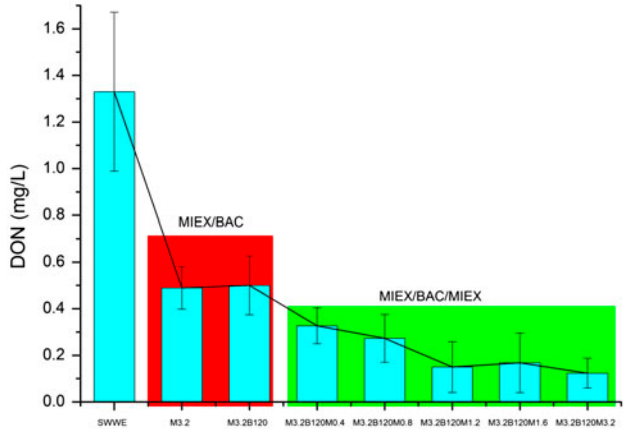

Fig. 4. Combined BAC and MIEX performance in MIEX/BAC and MIEX/BAC/MIEX order.

treatment has no effect, the effluent after MIEX/BAC (fed with MIEX non-amenable DON fraction only) would still be non-amenable to MIEX. However, the non-amenable residual DON (produced after M3.2 treatment), when subjected to BAC treatment produced DON fraction since it was removed by MIEX again. As can be seen from Fig. 4, when a range of MIEX dose was applied, it further reduced by 76% (0.5–0.12 mg/L). This indicates that although BAC could not remove DON fraction remained after MIEX treatment, it produced the amenable fraction out of non-amenable one. It is quite clear that BAC/MIEX treatment is significantly better than MIEX/BAC order. In addition, this observation shows the benefit of additional MIEX-amenable fraction resulting from BAC treatment in BAC/MIEX treatment order that could help to make treatment process more efficient.

4.5. Practical aspect of the study

Recycled wastewater is increasingly emerged as an alternative source of water supply to meet the increasing demand resulting from population growth, industrial as well as agricultural need. The water from BWTP has already been started to reuse via ground water replenishment. The SWWE obtained from treatment plant further goes to ultrafiltration and reverse osmosis followed by UV disinfection before being injected into the aquifer [30]. Similarly, Western Corridor Recycling Plant (Queensland, Australia), one of the largest in southern hemisphere, also recycles wastewater for power station and industrial use. It uses microfiltration and reverse osmosis followed by advanced oxidation process using UV/H_2O_2 [31]. Both plants are using membrane (microfiltration or ultrafiltration) as a pretreatment step prior to reverse osmosis. However, the abundance of effluent organic matter in SWWE fouls the membrane and demands frequent replacement. Thus, their prior removal is important for the extended life of the membrane.

One of our previous studies identified that BAC and MIEX could be used for the synergistic organic removal from SWWE. Their proper combination not only maximized the DOC removal but also decreased the MIEX dose (by more than 85%) for the given DOC removal [24]. In addition to DOC, same combination is found to be highly effective for the removal of DON from the SWWE in this study. While many nitrogenous DBPs (nitrosamines including NDMA, halonitromethane, haloacetonitriles) and carbonaceous DBS (trihalomethanes, haloacetic acids) produced during the disinfection process is a great deal of concern for water treatment industry, efficient removal of these precursors prior to disinfection could be highly effective treatment strategy. In this context, the pretreatment combination of BAC and MIEX could be an alternative solution either for the replacement of membrane pretreatment step or to reduce the DBP precursors and membrane foulants by removing both carbonaceous and nitrogenous organic compounds prior to membrane filtration.

5. Conclusions

The individual as well as the combined performance of BAC and MIEX treatment was investigated for the removal of DON. The result suggests that both BAC and MIEX have the ability to decrease DON in SWWE. However, no additional removal was obtained when MIEX was followed by BAC treatment possibly due to prior removal of biodegradable DON. At the same time, BAC/MIEX order of combination offered additional decrease in DON clearly indicating a supporting role of BAC treatment for subsequent MIEX treatment by producing more MIEX-amenable DON. Thus, effective removal obtained by BAC/MIEX order of combination is not only a result of sum of two individual performances but also a production of additional amenable fraction resulting during BAC treatment.

Acknowledgements

The author would like to thank National Centre of Excellence in Desalination for the financial support. Author is also grateful to Gary Ash and Eduardo Cosa from Water Corporation, respectively, for their help in providing MIEX resin and access to the treatment plant for sample collection.

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