Removal of organics and nitrogen in sewage treatment using anoxic-aerobic recirculated filter

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Abstract The anoxic-aerobic recirculated filter (AARF) process was investigated on removal efficiencies of organics and nitrogen with regard to loading rates, recirculation ratios of nitrified liquor and contribution of methane production and sulfate reduction in the treatment of the municipal sewage. The AARF process is composed of an anoxic filter for denitrification and an aerobic filter for nitrification and some of the nitrified liquor in the aerobic filter is recirculated to the anoxic filter. The AARF process successfully removed organics and nitrogen achieving high removal rates of 88% for COD and 64–74% for nitrogen. The recirculation ratio (Re) did not affect the COD removal efficiency but did affect the nitrogen removal, which was enhanced at a higher ratio (Re = 4). The methane production was not contributive to the COD removal but the COD consumed by the sulfate reduction was equivalent to 17% of total COD removed at Re = 2. We confirmed that the AARF process was applicable to the sewage treatment including nitrogen removal at a hydraulic retention time close to that of the conventional activated sludge process.

Keywords Anoxic-aerobic recirculated filter; denitrification; nitrification; sewage treatment; sulfate reduction

Introduction

Anaerobic filters are one of low-cost technologies used to treat various types of wastewater including municipal sewage in developing countries. As a eutrophication problem has become serious in those countries, it is required to remove such nutrients as nitrogen and phosphorus as well as organic matter in sewage treatment. In general, nitrogen can be biologically removed by a nitrification/denitrification process which is called the anoxic-aerobic process. The anoxic-aerobic recirculated filter (AARF) process is composed of an anoxic (in absence of oxygen) filter for denitrification and an aerobic filter for nitrification, and some of the nitrified liquor in the aerobic filter is recirculated to the anoxic filter. The aerobic filter also works for polishing the effluent from the anoxic filter in decomposition of organic matters.

Several studies on nitrogen removal from the municipal sewage using biological filters have been reported. Chudoba et al. (1998) reported that their process consisting of an anoxic filter and an aerobic complete-mixing tank achieved 10 mg/L of BOD and 12 mg/L of total nitrogen as effluent quality. Koopman et al. (1990) studied the denitrification performance of a moving bed upflow sand filter using secondary effluent. Canziani et al. (1999) revealed that nitrification and denitrification were strongly dependent on temperature and the hydrolysis of particles could be the rate limiting step of denitrification. The process using biological filters for both nitrification and denitrification, however, has not been reported except for a few studies such as Canziani et al. (1999). Thus, we feel that treatment characteristics and operational conditions of the AARF process have not been clarified yet enough to apply the process to the sewage treatment at once.

The main objective of this study is to investigate removal efficiencies of organic matter and nitrogen with regard to loading rates, recirculation ratios of nitrified liquor and
contribution of methane production and sulfate reduction when the AARF process is applied to treat the municipal sewage.

**Materials and methods**

**Apparatus and operation**

The AARF process consists of a cylindrical anoxic filter (AnF) and a cylindrical aerobic filter (AeF) as shown in Figure 1. Both filters were packed with plastic (PVC) lace media (Bio Module 1350, Shigensuiko-sha, Inc., Japan) with a 5% filling ratio and their space volumes were 1.6 L in AnF and 0.2 L in AeF. Supernatant from a primary sedimentation tank of a municipal sewage treatment plant in Tokyo was continuously introduced into the AnF operated at 37°C and then into the AeF at a room temperature of about 20°C. A hydraulic retention time (HRT) was set close to that of a conventional activated sludge process, i.e. 8 h in the AnF and 1 h in the AeF. The AeF was aerated to maintain DO at 2–3 mg/L for degradation of organics and nitrification, and some of nitrified liquor was recirculated to the AnF for denitrification with recirculation ratios (Re) of 2 and 4. Biogas produced from the AnF was collected by a gas holder. The AnF was inoculated by digested sludge from an anaerobic digester and the AeF was inoculated by activated sludge from an aeration tank of the sewage treatment plant. The process was operated for a month or more before reaching steady state conditions.

**Analytical methods**

Parameters assayed were dichromate COD, TOC, nitrogen (TN, NH$_4^+$, NO$_3^-$, NO$_2^-$), sulfate (SO$_4^{2-}$), DO, ORP, transparency, pH and biogas composition. TOC was measured by a TOC analyzer (TOC-500, Shimadzu). TN and NH$_4^+$ were determined by the ultraviolet spectrophotometric method using potassium persulfate and the phenate method, respectively. Anionic compounds (NO$_3^-$, NO$_2^-$, SO$_4^{2-}$) were analyzed by an ion chromatograph with a column packed with Shim-pack IC-A1 (HIC-6A, Shimadzu) and the biogas composition was determined using a TCD gas chromatograph with a stainless column packed with active carbon (GC-8AIT, Shimadzu). The remaining parameters were analyzed by following the Sewage Test Methods of Japan (1997).

**Process evaluation**

Evaluation of the AARF process was conducted as a whole process and also for each filter, i.e. for AnF on denitrification and sulfate reduction and for AeF on nitrification. A schematic diagram of mass flow in the AARF process was shown in Figure 2. Volumetric loading rate and removal rate can be computed by the following equations, where C: concentration of substance C, Q: flow rate, V: tank volume, Re: recirculation rate, and

![Figure 1](https://iwaponline.com/wst/article-pdf/46/9/309/426445/309.pdf)  
**Figure 1** Schematics of the anoxic-aerobic recirculated filter (AARF) process
subscripts $i$: influent, $n$: AnF effluent, $e$: AeF effluent. The slope of the linear regression line in between loading rate and removal rate gives us a reaction rate in terms of $\%$, e.g. a COD removal rate ($\%$) of the AARF process will be given by the slope of the regression line in between COD loading by Eq. (1) and COD removal by Eq. (4).

Volumetric loading rate (kg C/m$^3$.d)

<table>
<thead>
<tr>
<th>Process</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>AARF</td>
<td>$CiQ/(Vn + Ve) = Ci/HRT$ (1)</td>
</tr>
<tr>
<td>AnF</td>
<td>$(CiQ + CeReQ)/Vn = (Ci + CeRe)/HRTn$ (2)</td>
</tr>
<tr>
<td>AeF</td>
<td>$Cn(Re + 1)Q/Ve = Cn(Re + 1)/HRTe$ (3)</td>
</tr>
</tbody>
</table>

Volumetric removal rate (kg C/m$^3$.d)

<table>
<thead>
<tr>
<th>Process</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>AARF</td>
<td>$(Ci – Ce) Q/(Vn + Ve) = (Ci – Ce)/HRT$ (4)</td>
</tr>
<tr>
<td>AnF</td>
<td>${(Ci + CeRe) Q – Cn (Re + 1) Q}/Vn = {(Ci + CeRe) – Cn (Re + 1)}/HRTn$ (5)</td>
</tr>
<tr>
<td>AeF</td>
<td>${Cn (Re + 1) Q – Ce (Re + 1) Q}/Ve = {(Cn – Ce) (Re + 1)}/HRTe$ (6)</td>
</tr>
</tbody>
</table>

Results and discussion

Results of the sewage treatment by the AARF process were summarized in Table 1. All values are average by measuring 5 samples or more for each parameter at steady states. The effluent quality of the AARF process was desirable with low organic contents of 32–33 mg/L in T-COD, high transparency of 50 cm or more and neutral pH. Plotting volumetric COD removal rates versus COD loading rates using all measured values at steady states, a linear relation was obtained in between two rates as shown in Figure 3. The slope of the line gives us the COD removal rate based on Eqs. (1) and (4) as mentioned earlier. The COD loading rates were fluctuated in the range of 0.5 and 0.9 kg COD/m$^3$.day during operation, but the AARF process showed a stable performance and achieved a high COD removal rate of 88% at both $Re = 2$ and $Re = 4$. The recirculation ratio did not affect a COD removal efficiency within the range of this study.

Tanaka et al. (1991) reported that the anaerobic filter (AF) process decomposed COD in sewage with a removal rate of 45% in the range of 0.2–0.9 kg COD/m$^3$.day of volumetric COD loading rates at 8 h of HRT. As compared with the AF process, the AARF process achieved two times higher removal rates in COD.

Nitrogen contents in influent were 26–28 mg/L as TN including 10–16 mg/L of $NH_4^+$-N, and they were reduced to 10.0 mg/L at $Re = 2$ and 6.6 mg/L at $Re = 4$, respectively. TN removal rates were 64% at $Re = 2$ and 74% at $Re = 4$ for the influent sewage with a $C/N$
Nitrogen composition and removal were graphically depicted in Figure 4 based on the results in Table 1. The AeF effluent contained more NO$_3^-$-N at Re = 2 than at Re = 4 but Kjeldahl nitrogen (KN), consisting of NH$_4^+$-N and Org-N, was less at Re = 2 than at Re = 4, i.e., 2.3 mg/L at Re = 2 and 5.3 mg/L at Re = 4.

Figure 3  COD loading and removal rates in the AARF process (Re: recirculation ratio)

(TOC/TN) ratio of about 3. Nitrogen composition and removal were graphically depicted in Figure 4 based on the results in Table 1. The AeF effluent contained more NO$_3^-$-N at Re = 2 than at Re = 4 but Kjeldahl nitrogen (KN), consisting of NH$_4^+$-N and Org-N, was less at Re = 2 than at Re = 4, i.e., 2.3 mg/L at Re = 2 and 5.3 mg/L at Re = 4.

Figure 4  Nitrogen composition and removal in the AARF process
Relations between nitrogen loading rate and removal rate were plotted in terms of KN in Figure 5 and in terms of NO$_3^-$-N in Figure 6. Since the sum of NO$_3^-$-N and NH$_4^+$-N in the AeF effluent was larger than that in the AnF effluent flowing into the AeF as observed at Re = 2 in Table 1, we assumed that the nitrification reaction proceeded on both NH$_4^+$-N and biologically ammonified Org-N, i.e. on KN. Nitrification rates computed by the slope based on Eqs. (3) and (6) in Figure 5 were 84% at Re = 2 and 49% at Re = 4, while denitrification rates based on Eqs. (2) and (5) were 100% in both cases as shown in Figure 6.

These results indicate that higher recirculation of nitrified liquor performed better nitrogen removal under the conditions of this study. Although the nitrification rate was apparently higher at Re = 2 than at Re = 4, the recirculation of nitrified liquor at Re = 2 was not enough to completely denitrify NO$_3^-$-N as a whole process, resulting in discharge of the remaining NO$_3^-$-N from the process.

Biogas production from the AnF was also studied and the methane production was found negligible at both Re = 2 and Re = 4 (data were not shown). Methane producing bacteria (MPB) seem not to be contributive to the COD removal in the sewage treatment by the AARF process probably because of low organic contents in sewage and relatively high ORP in the AnF for MPB.

Sulfate reducing bacteria (SRB), however, contributed to the COD removal in the AARF process. Sulfate in influent was reduced to sulfide in the AnF and the sulfide was again oxidized to sulfate in the AeF as supported by the change of sulfate contents at Re-2 in Table 1. Considering sulfate supplied for the AnF by both influent and recirculated nitrified liquor, the COD consumed for the sulfate reduction by SRB was equivalent to 17% and 4% of total COD removed at Re = 2 and Re = 4, respectively, as shown in Table 2. We consider that the contribution of SRB to the COD removal is not negligible at low recirculation ratios such as 2 in this study. In general, higher recirculation ratio is considered disadvantageous to the sulfate reduction because of an increase of ORP by recirculation of the nitrified liquor. The increase of ORP in this study, however, was only 10 mV from –180 mV to –170 mV, and the change of pH was also little around neutral. One of reasons for the low activity of SRB at higher Re could be that SRB was defeated in the organic substrate competition between SRB and the denitrifying bacteria.

![Figure 5](https://iwaponline.com/wst/article-pdf/46/9/309/426445/309.pdf)  
**Figure 5** Kjeldahl nitrogen (KN) loading and removal rates in the AeF of the AARF process (Re: recirculation ratio)

![Figure 6](https://iwaponline.com/wst/article-pdf/46/9/309/426445/309.pdf)  
**Figure 6** NOx-N loading and removal rates in the AnF of the AARF process (Re: recirculation ratio)

| Table 2 COD removal rates by the sulfate reduction in the AnF of the AARF process |
|-------------------------------------------------|---------------------|
| Recirculation ratio | Re = 2 | Re = 4 |
| Contribution of sulfate reduction to COD removal (%) | 17 | 4 |
Conclusions
The anoxic-aerobic recirculated filter (AARF) process successfully removed organics and nitrogen from the municipal sewage. The AARF process stably achieved high removal rates of 88% for COD and 64–74% for nitrogen, resulting in 33 mg/L as COD and 6.6 mg/L as TN in effluent at a recirculation ratio of nitrified liquor (Re) of 4. The recirculation ratio did not affect the COD removal efficiency but did affect the nitrogen removal with enhancement of the nitrogen removal at higher Re within the range of this study. The methane production was not contributive to the COD removal but the contribution of sulfate reduction was not negligible at low Re. The COD consumed by the sulfate reduction was equivalent to 17% of total COD removed at Re = 2. We confirmed that the AARF process was applicable to the sewage treatment including nitrogen removal at a HRT close to that of the conventional activated sludge process.

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