Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.966532

56 (2015) 2856–2866
December

Taylor & Francis
Taylor & Francis Group

Effect of low temperature on the performance of a gravity flow CANON-like pilot plant MBR treating surface water

Zhaozhi Wang[a], Fangshu Qu[a], Heng Liang[a,*], Guibai Li[a], Robert W. Field[b]

[a]*State Key Laboratory of Urban Water Resource and Environment (SKLUWRE), Harbin Institute of Technology, 73 Huanghe Road, Nangang District, Harbin, 150090, P.R. China, Tel. +86 451 86283001; Fax: +86 451 86283001; emails: zhaozhiwang_1985@hotmail.com (Z. Wang), qufangshu@163.com (F. Qu), hitliangheng@163.com (H. Liang), hitsteven@gmail.com (G. Li)*

[b]*Department of Engineering Science, University of Oxford, Oxford, UK, email: robert.field@eng.ox.ac.uk (R.W. Field)*

Received 30 March 2014; Accepted 9 September 2014

ABSTRACT

A gravity flow completely autotrophic nitrogen removal over nitrite (CANON)-like pilot plant Membrane bio-reactor was designed to treat surface water for indirect potable reuse and was operated for over seven months with a constant Trans-membrane pressure of 100 mbar after the start-up phase. The effect of low temperature on the performance of autotrophic nitrogen removal over nitrite was investigated over a seven-month period during which the feed water temperature changed from 29℃ to 1℃ and the viscosity of the permeate more than doubled. The process showed good biostability and sustainability across the different seasons. Initially when the temperature fell below 10℃ (which seems like a turning point for the activity of ANAMMOX bacteria) high fluctuations happened, but the activity of ANAMMOX bacteria recovered in 18 d. Increasing Hydrogen retention time during this period was the means by which one can successfully compensate for low temperature. The dissolved oxygen (DO) (7.12–10.9 mg/L) in the raw water supported the partial nitrification and anaerobic ammonium oxidation (ANAMMOX) process without the use of aeration. There was high ammonium (up to 5 mg/L) removal even at extremely low temperatures of 1℃.

Keywords: Low temperature; CANON; Surface water; ANAMMOX; Sustainable flux

1. Introduction

Developing countries may have the most to gain from Membrane bio-reactor (MBR) technology because it can address their pressing needs for improved sanitation. In particularly, their small footprint, flexible design, and automation make MBRs ideal for rapidly growing urban areas, where large-scale public-works projects are expensive and are often completed slowly. MBRs offer the potential for decentralized systems that make water management more sustainable, particularly in megacities in the developing countries [1]. Li and Chu [2] thought that a MBR with a short Hydrogen retention time (HRT) could be developed as an effective biological water treatment process to address the urgent need of many developing countries that are plagued by the serious contamination of surface water resources. His study showed that the MBR process can be both technically and economically feasible for use in drinking water treatment not only for nitrification and organic degradation but also for the

---

*Corresponding author.

possible replacement of the conventional treatment process.

ANAMMOX stands for anaerobic ammonium oxidation. It is a globally important microbial process of the nitrogen cycle and in water treatment the ANAMMOX process is a new, cost-effective, and low-energy consuming alternative to the conventional nitrogen removal processes. The combination of partial nitrification and ANAMMOX process can be achieved in a dual reactor system such as the Single reactor for high activity ammonia removal over Nitrite–ANAMMOX processor in a single reactor such as in the Oxygen-limited autotrophic nitrification–denitrification process or the Completely autotrophic nitrogen removal over nitrite (CANON) process [3]. This paper reports upon a CANON-like MBR treating surface water for indirect potable reuse. Of particular concern for deployment in Northern China and other regions with a harsh winter is the effect of temperature upon the efficiency of the bio-treatment as there is a little that most water utilities can do to change water temperature. Almost all previous studies on this process were operated close to room temperature (10–25℃) [2,4–6] with only one to our knowledge at an extremely low temperature condition (0–5℃) [7].

The temperature range for the operation of the ANAMMOX process in waste water treatment is wide, being reported as 15–40℃ [3,8–10]. This was the same temperature range when using a MBR as reactor for an ANAMMOX process [11]. In other areas, several works on ANAMMOX have been done with marine samples and reported measurable activities at low temperatures. Rysgaard et al. [12] working with sediments of the east and west coasts of Greenland, observed ANAMMOX activity between −2 and 30℃, the optimum temperature being 12℃.

Bacterial growth is sustained by Nature organic matter constituting assimilable organic carbon (AOC) or biodegradable organic matter. Ravindran et al. [13] demonstrated that hybrid MBR processes could efficiently control disinfection by-products (DBP) formation and bacterial regrowth in the distribution system. An important feature of the hybrid MBR technology is the effective control of membrane fouling and permeate flux decline by a combination of powder activated carbon (PAC) sorption and fluid management.

In actual operation, the water temperature can drop to almost 0℃ in North China. Although some bacteria still have activity, a concern is whether it is sufficient. So in the work reported below, a high dose of powdered activated carbon (PAC) was used as nutrition and biomass support, and biostability was assessed across different seasons. In particular, the aim of this study was to assess the effect of low temperature on the long-term performance of a gravity flow CANON-like pilot plant MBR under previously determined operating conditions. For the chosen location, the temperature decreased from 29 to 1℃ over the seven months of operation.

2. Materials and methods

2.1. Operation of MBR

The pilot plant was installed to treat reservoir water from the Yellow River at Dongying drinking water treatment facility, Shandong Province, China. The membrane fibers were made of Polyvinyl chloride and were provided by Suzhou Litree Ultrafiltration Membrane Technology Co. Ltd., China. The fibers had an average pore size of 0.01 μm, with inner and outer diameters of 0.85 mm and 1.45 mm, respectively. The membrane area was 72 m^2. More details are given in the Appendix. This study was conducted from 28 May 2010 to 30 December 2011.

The filtration was operated under gravity flow (*ca.* 1 m water head ≈ 100 mbar). Aeration was intermittent (11 h 50 mins off and 10 min on) carried out at a flow rate of 0.83 m^3/m^2 (membrane area) h. The back flush flux carried out during the aeration mode was two times of the operating flux. Filtration was therefore also intermittent (11 h 50 mins on and 10 min off). Details on the laboratory and pilot plant work that led to the choice of these conditions are given elsewhere [11,14]. The HRT varied between 34 min and 125 min, and therefore, the oxygen addition during the twice per day back flush/scour periods had minimal influence on the DO value which was primarily determined by the oxygen level in the influent.

As the concentration of ammonium in the raw water was around 0.35 mg/L, additional ammonium was supplied as NH_4Cl (26 g per day on nitrogen basis). The addition rate was not changed during this study. This initial gave a concentration of around 2 mg/L, but as the flux decreased the concentration in the feed water rose as the addition rate of NH_4Cl was unchanged. The concentration of ammonium, nitrite, and nitrate was recorded for the last six months of the study from 2 July 2010 to 30 December 2010.

2.2. Temperature corrected permeability loss

According to Field and Pearce [15], a temperature corrected change in permeability should be calculated based on J_{20}, which is given by:

$$J_{20} = J(T) * \mu(T)/\mu_{20} \quad (1)$$

where J(T) is the actual flux at temperature T, $\mu(T)$ is the viscosity of water at temperature T, $\mu(20)$ is the viscosity of water at 20℃.

Temperature corrected rate of permeability loss is based upon J_{20} fluxes. As the operation was observed to have periods of slow flux decline (labeled pseudo steady state, PSS) and periods of transition the following were defined.

Across the ith PSS, the mean temperature corrected rate of permeability loss [(L/m^2 h) per bar per day] was calculated as:

$$\text{Rate of loss of permeability across PSS}_i = \frac{J_{20}(\text{at start of PSS}_i) - J_{20}(\text{at end of PSS}_i)}{\text{TMP} * \text{Duration of PSS}_i} \tag{2a}$$

Across a transition from the ith to the $(i+1)$th PSS, the temperature corrected rate of loss of permeability was calculated as:

$$\text{Rate of loss of permeability across Transition}_i = \frac{J_{20}(\text{at end of PSS}_i) - J_{20}(\text{at start of PSS}_i + 1)}{\text{TMP} * \text{Duration of Transition}_I} \tag{2b}$$

2.3. NO$_X$-N concentration in the MBR

Given that the CANON process was the major mechanism in this system, one can [16] represent the chemical balances in the following manner:

NO$_X$-N concentration can be calculated by Eqs. (3)–(7):

The equilibrium of DO can be calculated:

$$\text{DO}_{\text{consume}} = \text{DO}_{\text{influent}} - \text{DO}_{\text{in MBR}} \tag{3}$$

where DO$_{\text{consume}}$ is the DO consumed during reaction, DO$_{\text{influent}}$ is the original DO in the influent, and DO$_{\text{in MBR}}$ is DO in the MBR. This is connected to the ammonium balance by Eq. (4).

$$1.94\left[\text{NH}_4^+\right]_{\text{canon removal}} + 4.57\left[\text{NH}_4^+\right]_{\text{nitrification removal}} = \text{DO}_{\text{consume}} \tag{4}$$

$[\text{NH}_4^+]_{\text{canon removal}}$ and $[\text{NH}_4^+]_{\text{nitrification removal}}$ are respectively the concentration of ammonium calculated as N removed by CANON and nitrification processes. The coefficient1.94 indicates that for every milligram ammonium per liter removed, 1.94 milligram DO per liter are consumed in the ANOMMOX process [3].

The removal of ammonium can be calculated from:

$$\left[\text{NH}_4^+\right]_{\text{total removal}} = \left[\text{NH}_4^+\right]_{\text{influent}} - \left[\text{NH}_4^+\right]_{\text{effluent}} \tag{5}$$

where $[\text{NH}_4^+]_{\text{total removal}}$, $[\text{NH}_4^+]_{\text{influent}}$ and $[\text{NH}_4^+]_{\text{effluent}}$ are respectively the concentrations related to removal, influent, and effluent.

The sum of the concentration of ammonium removed by CANON and nitrification processes is the total concentration of ammonium removed.

$$\left[\text{NH}_4^+\right]_{\text{canon removal}} + \left[\text{NH}_4^+\right]_{\text{nitrification removal}} = \left[\text{NH}_4^+\right]_{\text{total removal}} \tag{6}$$

$[\text{NH}_4^+]_{\text{canon removal}}$ and $[\text{NH}_4^+]_{\text{nitrification removal}}$ can be calculated through Eqs. (4)–(6).

The following balances are listed:

$$\left[\text{NH}_4^+\right]_{\text{nitrification removal}} = \left[\text{NO}_3^-\right]_{\text{nitrification}} \tag{7}$$

$[\text{NO}_3^-]_{\text{nitrification}}$ indicates the concentration of nitrate calculated as N produced by the nitrification process.

$$\left[\text{NO}_3^-\right]_{\text{canon}} = 0.13\left[\text{NH}_4^+\right]_{\text{canon removal}} \tag{8}$$

$[\text{NO}_3^-]_{\text{nitrification}}$ indicates the concentration of nitrate calculated as N produced in the CANON process. The coefficient 0.13 refers to the accepted ratio of $\text{NO}_3^-{}_{\text{nitrification}}/\text{NH}_4^+{}_{\text{total removal}}$ in a CANON system [16]. Also,

$$\left[\text{NO}_3^-\right]_{\text{produced}} = \left[\text{NO}_3^-\right]_{\text{nitrification}} + \left[\text{NO}_3^-\right]_{\text{canon}} \tag{9}$$

where $[\text{NO}_3^-]_{\text{produced}}$ indicates the concentration of nitrate calculated as N produced by CANON and nitrification process. This can be calculated through Eqs. (7)–(9). Additionally the sum of the produced and influent concentration of nitrate can be calculated by Eq. (10) and this gives a predicted value for the effluent.

$$\left[\text{NO}_3^-\right]_{\text{calculated}} = \left[\text{NO}_3^-\right]_{\text{produced}} + \left[\text{NO}_3^-\right]_{\text{influent}} \tag{10}$$

Now $[\text{NO}_3^-]_{\text{calculated}}$ was compared to the measured value, $[\text{NO}_3^-]_{\text{effluent}}$ to check the supposed reaction scheme.

2.4. Feed solution and analytical methods

Water quality analysis was conducted following standard methods [17]. Table A.1 gives a summary based upon means, standard deviations, and ranges, while Fig. 1 plots out six trends with time. NH_4^+-N, NO_2^--N, and NO_3^--N concentrations were determined by colorimetric methods using a spectrometer (UV_{754}, CANY, China); UV absorbance at 254 nm (UV_{254}) was also determined. Dissolved organic carbon (DOC) was measured by the TOC analyzer (TOC-VCPH, Shimadzu, Japan) after pre-filtrating through 0.45 μm membrane. pH and DO concentrations in the reactors were measured by a DO electrometer (pH/Oxi 340i, WTW, Germany) with a probe (Cellox® 325). Trihalomethanes formation potential (THMFP) and Haloacetic acids formation potential (HAAFP) were determined following US EPA methods 551.1 and 552.2, except that the incubation time after chlorine dosing was shortened from 7 to 3 days, recommended by Tian et al. [4]. After incubation, excess chlorine in the water sample was quenched with 10% NH_4Cl followed by MTBE extraction. The extracted sample was then analyzed for trihalomethanes (THMs) and haloacetic acids by a GC (Agilent 6890 N, USA), equipped with a capillary column (30.0 m × 0.32 mm × 0.25_m, HP-5, Agilent J&W, USA), and an electron capture detector. The THMFP presented was the sum of $CHCl_3$, $CHBrCl_2$, $CHBr_2Cl$, and $CHBr_3$; the HAAFP was the sum of $CH_2ClCOOH$, $CH_2BrCOOH$,$CHCl_2COOH$, CCl_3COOH, $CHBrClCOOH$, $CBrCl_2COOH$, $CHBr_2COOH$, $CBr_2ClCOOH$, and CBr_3COOH.

The measurement of assimilable organic carbon (AOC) was carried out using the modified method by Liu et al. [18], which was developed based on the procedures of Van der Kooij et al. [19]. The bioassay procedure employed two strains of microorganisms: *Pseudomonas fluorescens* (P17) and *Aquaspirillum* (NOX), P17 is capable of utilizing amino acids, carboxylic acids, alcohols, and carbohydrates; NOX is capable of utilizing carboxylic acids and oxalic acids. First, 104 colony forming units (CFU)/mL of P17 strain were inoculated into the water sample and incubated for 2 d at 22℃; the CFU were then counted. After that, the NOX strain (104 CFU/mL) was inoculated and incubated for 3 d at 22℃ before the CFU were counted. AOC concentration was calculated by comparing the cell formation unit number and yield coefficient, and the total AOC was the sum of both AOC_{P17} and AOC_{NOX}.

3. Results and discussion

3.1. Sustainability of MBR operation

From the preliminary work (briefly mentioned in the support information), the sustainable flux of raw water was < 10L/m^2 h. Furthermore, as discussed by Aimar and Bacchin [20], for less stable media (e.g. higher ionic strength or smaller particle), aggregation is observable over matters of weeks rather than days for suspensions of moderate stability and this leads to flux decline at fluxes below the nominal value of the critical flux. As described in Table A.2 and shown in Fig. 2, the performance of the MBR can be divided into five pseudo steady states (PSSs) corresponding to five near constant actual fluxes.

Excluding the transition periods, the highest rate of temperature corrected permeability loss (0.322 (L/m2 h)per bar per day) occurred in PSS Ⅰ which was the one period in which temperature increased significantly; the increase was 8℃. The permeability change in PSS II and III was small or even zero and the temperature change was small. This was similar in PSS IVa and Vb. When temperature decreased from 26.1 to 6.5 and 5.5 to 1℃ in PSS IVb and Va, respectively, the permeability losses were negative which indicates that the resistance decreased leading to the recovery of J_{20}.

The highest permeability loss happened in Transition II with temperature increasing. The other three transitions occurred when temperature was decreasing. The possible explanation for these sharp decreases in permeability will be similar to the reasons given [21,22] for the "trans-membrane pressure (TMP) jump". Zhang et al. [21] examined the factors affecting the membrane performance in submerged MBRs and described a three stage process. Stage 1 occurs in a period of a few hours and involves a small abrupt TMP rise due to "conditioning", presumably by pore blockage and closure. This stage is irrelevant for the time scale described in the present work (and the pore size of the membranes we used was small). Stage 2 is a prolonged period of slow TMP rise, which they ascribed to accumulation of extracellular polymeric substances and other products of bioactivity, either produced in biofilms on the membrane surface or deposited from the bulk liquor. Stage 3 is a sudden rise in TMP and which according to their work led quickly to inoperability of the modules. They operated under constant flux, and the present results were obtained under a constant TMP and so we were able to accommodate the increased fouling. Their Stage 2 corresponds to our PSS periods and our transitions to their Stage 3. The periods of a slow increase in resistance followed by a relatively sharp increase over a short period indicate a phenomenon related to the TMP jump [21]. This stage may have several causes as discussed elsewhere [21–23]. Each transition corresponded to a rapid increase of R_f. In case of metastability, a slow aggregation mechanism is to be expected, producing a membrane fouling limited by diffusion of particles to each other. As the flux decreases, this mechanism is meant to slow down, but this drift depends on particles stability, residence time, and shear rate in the modules [20].

Four explanations for increased membrane fouling at low temperatures were mentioned by Jiang et al. [24] but the results herein showed reduced fouling at lower temperature, and Table A.2 shows two periods of recovery in temperature corrected permeability(PSS IVb and PSS Va). The recovery in temperature corrected permeability at low temperature may be related to biofilm detachment caused not by nutrient starvation but by low temperature stress-induced sloughing [25].

3.2. Ammonium removal during seasonal change

Increasing the DO supply by aeration can increase the ability of treating ammonium, but more energy would be required. In this study, aeration was only applied for very short periods to scour the membrane. The DO needed for ammonium removal was all supplied by the DO in the raw water (7.12–10.9 mg/L). This was sufficient for ammonium not higher than 6 mg/L because it has been reported that entire nitrogen removal can be achieved in a single reactor with limited aeration (i.e. around 1.94 g O_2 per g-N) [3]. A single reactor process like the CANON requires efficient biomass retention and appropriate control of dissolved oxygen (DO) concentrations. It is assumed that AOB are active in the outer oxic region of biofilms or aggregates, while ANAMMOX bacteria are present in the inner anoxic region. The most important factor for controlling CANON is the maintenance of DO at concentrations between 0.2 and 1 mg/L, which ensures the ammonium oxidation and the ANAMMOX reaction occur simultaneously [8,26]. Our MBR system was designed like a CANON process. The DO in the reactor column and membrane bioreactor were nearly the same due to reflux and < 1 mg/L during the whole study.

All the PSSs and Transitions (Fig. 3(b) and Table 1) in CANON process were similar to that in flux decline (Fig. 2(c)) except PSS IV′ and Transitions a and b (the same duration as that of PSS IVa and IVb).

From Transition 1–3, the temperature fluctuated between 24 and 29℃, the $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value decreased from 0.64 to 0.13 (traditional CANON system, [16]) due to increase of ammonium in raw water from 2 to 4.4 mg/L, and the HRT increased from 34 to 75 min. During these periods, above 24℃, temperature seems not to have too much effect on CANON process.

When temperature decreased from 25.8 to 10.2℃, the $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value increased a little due to the increased DO in the raw water (PSS IV′).

The most drastic change of $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value occurred when the temperature fell below 10℃. It suddenly jumped to 0.81 which means nitrification was the dominant process (Transition a). The activity of ANAMMOX bacteria was only limited. In the next 18 d, the activity of ANAMMOX bacteria recovered with the $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value decreased to 0.25 (Transition b). As the ammonium and HRT increased from 4.4 to 7.34 mg/L and 75 to 125 min, respectively, the $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value decreased to 0.21 (Transition 4).

With the temperature still decreasing down to 1℃, no large influence was observed from temperature (PSS Va). At the extreme condition, the $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ value decreased to 0.16 (PSS Vb).

Li’s [2] study also showed high recovery of nitrification within just a few days of the temperature falling below 16℃. Andersson et al. [7] suggest that the major impact of low temperatures was a loss of the bacterial activity (expressed under the form of ammonium removal) but not a complete biomass loss.

Although the activity of nitrifying biomass declined at low temperature, the contacted time (HRT) had increased from 34 to 125 mins as a result of decline of flux over the whole period. By extracting typical HRT and ammonium removal concentrations for periods in which there was a near steady state lasting more than 10 d, it was shown that there was a good correlation between HRT and ammonium removal (Fig. 3(d)). Each point (HRT (mins), ammonium removal (mg/L)) is listed: point 1 (34, 1.99) lasted 19 d; point 2 (53.5, 3.13) lasted 11 d; point 3 (62.4, 3.77) lasted 12 d; point 4 (74.8, 4.36) lasted 46 d; and point 5 (124.7, 5.07) lasted 15 d. If point 5 is excluded (when temperature was below 10℃), a high correlation $R^2 = 0.994$ is found when temperature was above 10℃ (Fig. 3(d)):

$$[NH_4^+]_{\text{removal}} = 0.059\text{HRT} \tag{12}$$

$NO_3^-{}_{\text{calculated}}$ was almost the same with the nitrate concentration in the effluent (Fig. 3(a)) which confirms that the partial nitrification and ANAMMOX were the dominant process in the MBR system. The acid produced in CANON process was neutralized by hardness (around 300 mg/L calculated as $CaCO_3$) in the raw water so the pH in the influent and effluent did not change.

Table 1
Performance of CANON process under low temperature (↑) and (↓) mean the increase and decrease trend respectively. (F) means fluctuation

PSS	Duration	Ammonium in raw water (mg/L)	Temperature (°C)	DO in raw water (mg/L)	DO in MBR (mg/L)	$NO_3^-{}_{\text{nitrification}}$/ $NH_4^+{}_{\text{total removal}}$	$NH_4^+{}_{\text{canon removal}}$ (mg/L)	$NH_4^+{}_{\text{nitrification removal}}$ (mg/L)	HRT (mins)
PSS I	7/2–7/17	2	24.6–28.6 (↑)	7.92–7.33 (↓)	1.02–1.04 (F)	0.64–0.54 (↓)	0.82–1.05 (↑)	1.16–0.93 (↓)	34
Transition 1	7/17–7/24	2–3.15 (↑)	26.1–28.6 (F)	7.31–7.62 (F)	1.04–0.94 (↓)	0.54–0.16 (↓)	1.05–3.02 (↑)	0.93–0.11 (↓)	34–53 (↑)
PSS II	7/24–8/3	3.15	28.5	7.2–7.34 (F)	0.9–0.95 (F)	0.16	3	0.08–0.12 (F)	53
Transition 2	8/3–8/4	3.15–3.67 (↑)	28.5–29.1 (↑)	7.33–7.12 (↓)	0.93–0.34 (↓)	0.16–0.15 (↓)	3–3.33 (↑)	0.12–0.07 (↓)	53–62 (↑)
PSS III	8/4–8/24	3.67	29.1	7.13	0.35	0.15	3.33	0.07	62
Transition 3	8/24–8/26	3.67–4.4 (↑)	29.1–25.8 (↓)	7.13–7.68 (↑)	0.35–0.34 (↓)	0.15–0.13 (↓)	3.33–3.77 (↑)	0.07–0 (↓)	62–75 (↑)
PSS IV′	8/26–11/9	4.4	25.8–10.2	7.68–9.54 (↑)	0.29–0.35 (F)	0.13–0.19 (↑)	3.77–4.31 (F)	0–0.3 (↑)	75
Transition a	11/9–11/11	4.4	10.2–9 (↓)	9.54–9.55 (↑)	0.3–0.55 (↑)	0.19–0.81 (↑)	4.06–0.48 (↓)	0.3–1.76 (↑)	75
Transition b	11/11–11/28	4.4	9–6.5	9.55–10.06 (↑)	0.55–1.06 (↑)	0.81–0.25 (↓)	0.48–3.35 (↑)	1.76–0.55 (↓)	75
Transition 4	11/28–12/6	4.4–7.34 (↑)	6.5–5.5 (↓)	10.06–10.12 (↑)	1.06–0.3 (↓)	0.25–0.21 (↓)	3.35–4 (↑)	0.55–0.4 (↓)	75–125 (↑)
PSS Va	12/6–12/16	7.34	5.5–1 (↓)	10.12–10.9 (↑)	0.3–0.37 (F)	0.21–0.19 (↓)	4–4.69 (↑)	0.4–0.33 (↓)	125
PSS Vb	12/16–12/30	7.34	1	10.9	0.3–0.4 (F)	0.19–0.16 (↓)	4.69–4.93 (↑)	0.33–0.2 (↓)	125

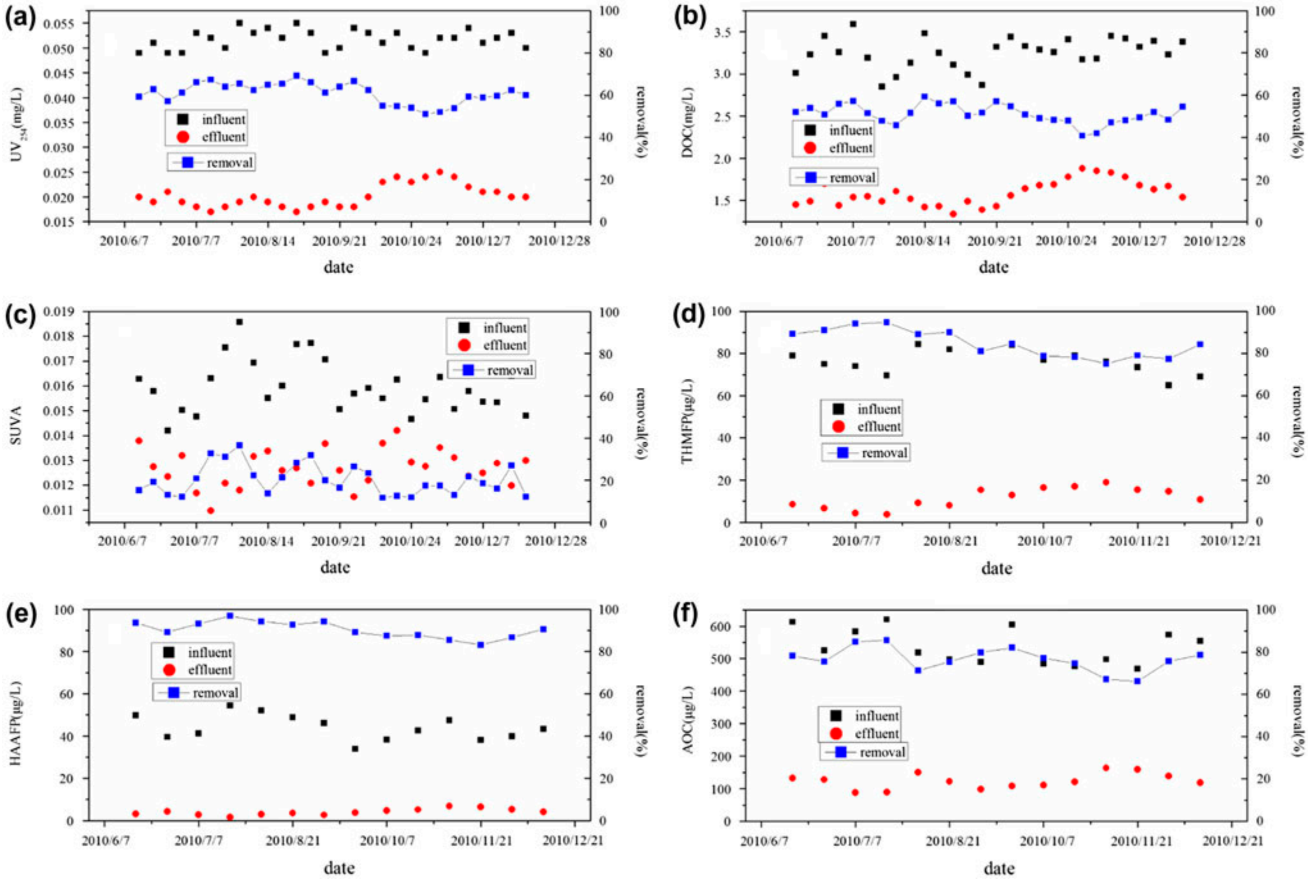


Fig. 1. Effect of low temperature on the biostability of the MBR: (a) UV_{254} removal efficiency; (b) DOC removal efficiency; (c) SUVA removal efficiency; (d) THMFP removal efficiency; (e) HAAFP removal efficiency; (f) AOC removal efficiency.

3.3. Biostability

The submerged MBR treating surface water was continuously operated for more than seven months. During its stationary operation (Table A.1 and Fig. 1), the MBR influent had average DOC concentrations of 3.22 ± 0.37 mg/L. In general, average 50% of the organics were removed by the MBR treatment, thus reducing the DOC to 1.61 ± 0.27 mg/L in the effluent. UV_{254} was reduced by average 60% from around 0.052 ± 0.003 to $0.021 \pm 0.004\ cm^{-1}$. A lower SUVA indicates that the organic in water might allow less chlorine substitution, resulting in less THMs and other DBPs formation. Thus, SUVA is a good surrogate for predicting the overall DBPs formation potential of the organic, regardless of its nature in a water source. The SUVA of the MBR effluent in our study was reduced by 24% on average from that of the raw water. This result indicates that the organic residues after the MBR treatment did not have higher THMFP, although they were largely non-biodegradable. This was coincidence with the high removal of THMFP and HAAFP,

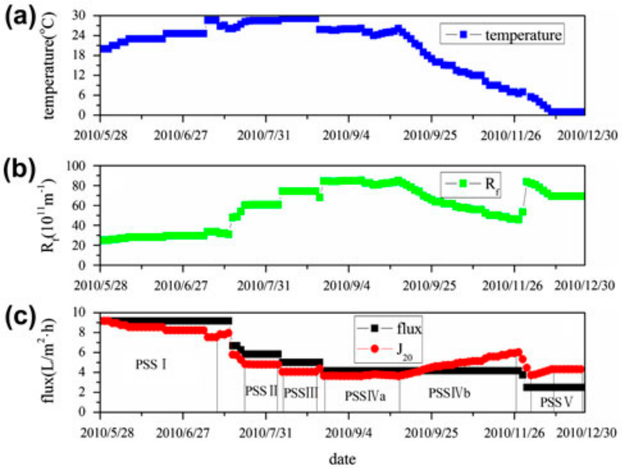


Fig. 2. Effect of low temperature on flux decline: (a) temperature variation during seasonal change; (b) resistance $R_f = R_{total} - R_m$ (according to Darcy's law) for each flux. R_{total} and R_m mean total resistance and original membrane resistance, respectively; (c) flux decline under gravity flow during seasonal change.

84.9 and 90% on average, respectively. With the biological treatment of MBR, the biostability of the finishing water was improved considerably as the AOC decreased from 531.1 ± 89.8 to 126.5 ± 37.2 μg/L.

Although this MBR system was designed for indirect potable reuse, biostability still needs to be proved. AOC is considered to be the main nutrient that controls microbial regrowth in water distribution systems. The operational goal for biologically stable water in the Netherlands has been set at 10 μg/L for the AOC, while in the United States, an AOC level of 50 μg/L has been recommended for coliform control [2].

The removal of all impurities was stable even at low temperature. Apossible explanation was the increasing of HRT from 34 to 125 min. This was coincidence with the removal of ammonium. The lowest level of removal for all impurities occurred from 7 November to 21 November (Transition a and b or PSS IVb) before the high fluctuation of NO_X started below 10°C. But with time passing, the removal increased to normal level again.

4. Conclusion

The membrane resistances increased as temperature increased, and vice versa. Low temperature can decrease resistance leading to the recovery of J_{20}.

Partial nitrification and ANAMMOX are a complex phenomenon, and to our knowledge, this was the first time that this process has been used to treat surface water under an extreme low temperature condition. Temperature of 10°C seems like a turning point for the activity of ANAMMOX bacteria. CANON process can be active even at extreme condition (1–10°C) treating surface water.

While temperature was found to impact on the performance of the CANON-like MBR, increasing HRT seems to be a means by which one can successfully compensate for low temperature. Seven months of operation under seasonal change verify that this CANON-like MBR has potential of indirect potable reuse even under low temperature of a few degrees Centigrade.

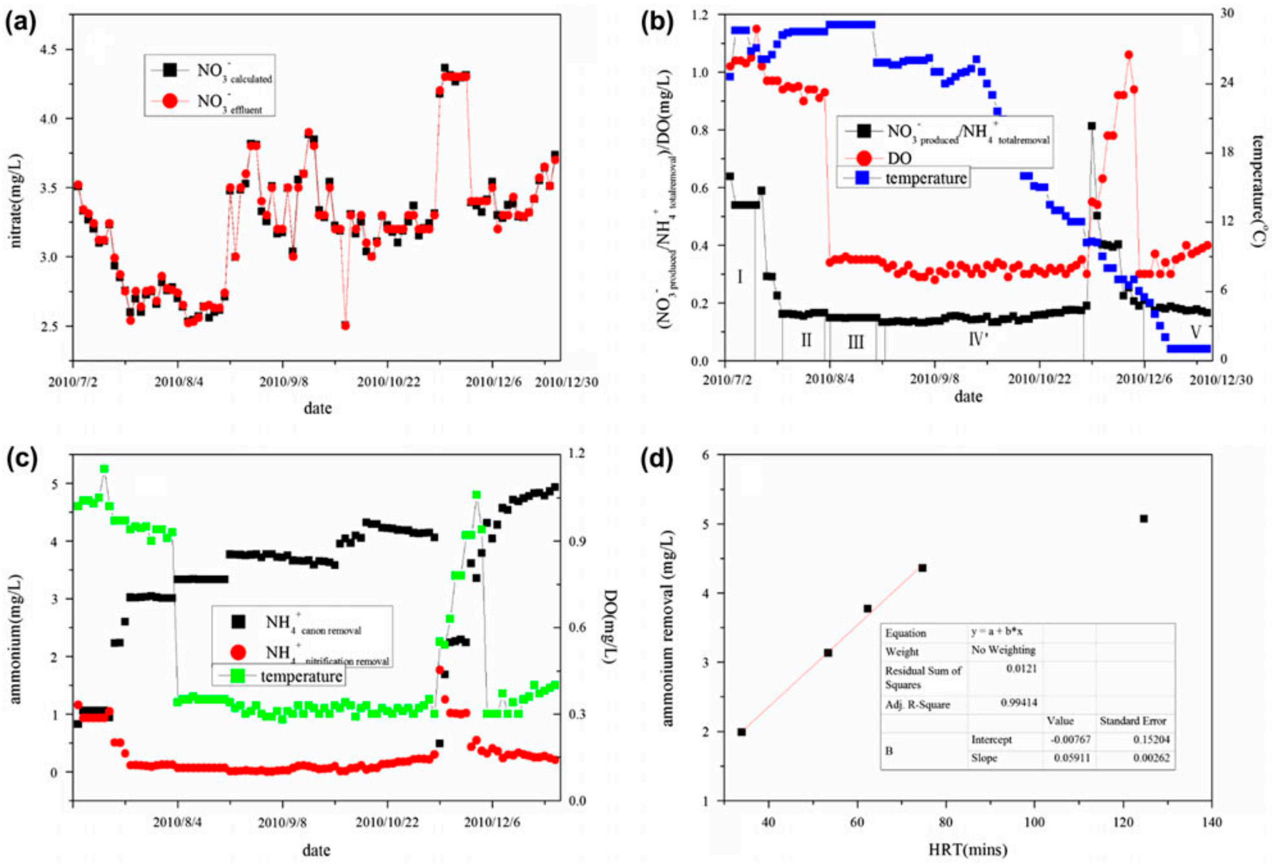


Fig. 3. Effect of low temperature on CANON-like process: (a) comparison of calculated and effluent concentration of nitrate; (b) $NO_3^-{}_{\text{nitrification}}/NH_4^+{}_{\text{total removal}}$ varied with DO and temperature; (c) ammonium removed by CANON and nitrification varied with DO; (d) the correlation between ammonium removal and HRT. The intercept was set to zero.

Acknowledgements

This research was jointly supported by Program for New Century Excellent Talents in University (NCET-13-0169), State Key Laboratory of Urban Water Resource and Environment (2014DX04), Science and Technology Planning Project of Chancheng District (2013A1044), and the Fundamental Research Funds for the Central Universities (AUGA5710050713).

List of acronyms

ANAMMOX	— anaerobic ammonia oxidation
AOB	— ammonia oxidizing bacteria
AOC	— assimilable organic carbon
AOC_{P17}	— AOC calculated as *Pseudomonas fluorescens* (P17)
AOC_{NOX}·	— AOC calculated as *Aquaspirillum* (NOX)
BOM	— biodegradable organic matter
CANON	— completely autotrophic nitrogen removal over nitrite
CFU	— colony forming units
DBP	— disinfection by-products
DO	— dissolved oxygen
DOC	— dissolved organic carbon
ECD	— electron capture detector
EPS	— extracellular polymeric substances
HAAs	— haloacetic acids
HAAFP	— haloacetic acids formation potential
HRT	— hydrogen retention time
MBR	— membrane bio-reactor
NH_3	— ammonium
$[\text{NH}_4^+]$	— ammonia calculated as nitrogen
$[\text{NO}_3^-]$	— nitrate calculated as nitrogen
$\text{NO}_3^-{}_{\text{produced}}/\text{NH}_4^+{}_{\text{total removal}}$	— ratio of produced NO_3^--N concentration and total NH_4^+-N concentration
NOB	— nitrite oxidized bacteria
NOM	— nature organic matter
NOX	— *Aquaspirillum*
OLAND	— oxygen-limited autotrophic nitrification–denitrification
PAC	— powder activated carbon
PSS_i	— the *i*th pseudo steady state. definition is given in Section 2.2
PVC	— polyvinyl chloride
SHARON	— single reactor for high activity ammonia removal over nitrite
SRT	— sludge retention time
SUVA	— UV_{254}/DOC*100
THMs	— trihalomethanes
THMFP	— trihalomethanes formation potential
TMP	— trans-membrane pressure
UV_{254}	— UV absorbance at 254 nm

References

[1] G. Daigger, E. Rittmann, S. Adham, G. Andreottola, Are membrane bioreactors ready for widespread application? Environ. Sci. Technol. 39 (2005) 399A–406A.

[2] X. Li, H.P. Chu, Membrane bioreactor for the drinking water treatment of polluted surface water supplies, Water Res. 37 (2003) 4781–4791.

[3] S. Cho, N. Fujii, T. Lee, S. Okabe, Development of a simultaneous partial nitrification and anaerobic ammonia oxidation process in a single reactor, Bioresour. Technol. 102 (2011) 652–659.

[4] J. Tian, H. Liang, Y. Yang, S. Tian, G. Li, Membrane adsorption bioreactor (MABR) for treating slightly polluted surface water supplies: As compared to membrane bioreactor (MBR), J. Membr. Sci. 325 (2008) 262–270.

[5] Y. Miura, Y. Watanabe, S. Okabe, Membrane biofouling in pilot-scale membrane bioreactors (MBRs) treating municipal wastewater: Impact of biofilm formation, Environ. Sci. Technol. 41 (2007) 632–638.

[6] K. Kimura, N. Yamato, H. Yamamura, Y. Watanabe, Membrane fouling in pilot-scale membrane bioreactors (MBRs) treating municipal wastewater, Environ. Sci. Technol. 39 (2005) 6293–6299.

[7] A. Andersson, P. Laurent, A. Kihn, M. Prévost, P. Servais, Impact of temperature on nitrification in biological activated carbon (BAC) filters used for drinking water treatment, Water Res. 35 (2001) 2923–2934.

[8] A. Joss, D. Salzgeber, J. Eugster, R. König, K. Rottermann, S. Burger, P. Fabijan, S. Leumann, J. Mohn, H. Siegrist, Full-scale nitrogen removal from digester liquid with partial nitritation and ANAMMOX in one SBR, Environ. Sci. Technol. 43 (2009) 5301–5306.

[9] J. Dosta, I. Fernández, J.R. Vázquez-Padín, A. Mosquera-Corral, J.L. Campos, J. Mata-Álvarez, R. Méndez, Short- and long-term effects of temperature on the ANAMMOX process, J. Hazard. Mater. 154 (2008) 688–693.

[10] X. Hao, J. Heijnen, M. Van Loosdrecht, Model-based evaluation of temperature and in flow variations on a partial nitrification–ANAMMOX biofilm process, Water Res. 36 (2002) 4839–4849.

[11] Z. Wang, H. Liang, F. Qu, Start up of a gravity flow CANON-like MBR treating surface water under low temperature, Chem. Eng. J. 217 (2013) 466–474.

[12] S. Rysgaard, R.N. Glud, N. Risgaard-Petersen, T. Dalsgaard, Denitrification and ANAMMOX activity in Arctic marine sediments, Limnol. Oceanogr. 49 (2004) 1493–1502.

[13] V. Ravindran, H. Tsai, M.D. Williams, M. Pirbazari, Hybrid membrane bioreactor technology for small water treatment utilities: Process evaluation and primordial considerations, J. Membr. Sci. 344 (2009) 39–54.

[14] Z. Wang, R.W. Field, F. Qu, Y. Han, H. Liang, G. Li, Use of threshold flux concept to aid selection of sustainable operating flux: A multi-scale study from laboratory to full scale, Sep. Purif. Technol. 123 (2014) 69–78.

[15] R.W. Field, G.K. Pearce, Critical, sustainable and threshold fluxes for membrane filtration with water industry applications, Adv. Colloid Interface Sci. 164 (2011) 38–44.

[16] K.A. Third, A.O. Sliekers, J.G. Kuenen, M.S.M. Jetten, The CANON system (completely autotrophic nitrogen-removal over nitrite) under ammonium limitation: Interaction and competition between three groups of bacteria, Syst. Appl. Microbiol. 24(4) (2011) 588–596.

[17] APHA, WEF, AWWA, Standard Methods for the Examination of Water and Wastewater, twentieth ed., American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC, 1998.

[18] W. Liu, H. Wu, Z Wang, S.L. Ong, J.Y. Hu, W.J. Ng, Investigation of assimilable organic carbon (AOC) and bacterial regrowth in drinking water distribution system, Water Res. 36 (2002) 891–898.

[19] D. Van der Kooij, A. Visser, W.A.M. Hijnen, Determining the concentration of easily assimilable organic carbon in drinking water, J. Am. Water Works Assn. 74 (1982) 540–545.

[20] P. Aimar, P. Bacchin, Slow colloidal aggregation and membrane fouling, J. Membr. Sci. 360 (2010) 70–76.

[21] J. Zhang, H.C. Chua, J. Zhou, A.G. Fane, Factors affecting the membrane performance in submerged membrane bioreactors, J. Membr. Sci. 284 (2006) 54–66.

[22] R.W. Field, K. Zhang, Z. Cui, B.-K. Hwang, Flat sheet MBRs: Analysis of TMP rise and surface mass transfer coefficient, Desalin. Water Treat. 35 (2011) 82–91.

[23] B.K. Hwang, C.-H. Lee, I.-S. Chang, A. Drews, R. Field, Membrane bioreactor: TMP rise and characterization of bio-cake structure using CLSM-image analysis, J. Membr. Sci. 419–420 (2012) 33–41.

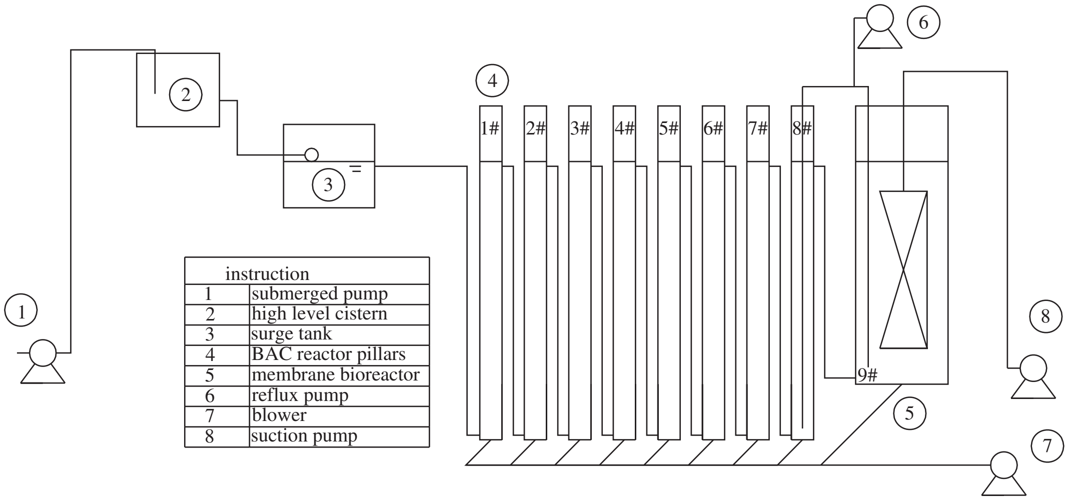
[24] T. Jiang, M.D. Kennedy, B.F. Guinzbourg, P.A. Vanrolleghem, J.C. Schippers, Optimising the operation of a MBR pilotplant by quantitative analysis of the membrane foulingmechanism, Water Sci. Technol. 51 (2005) 19–25.

[25] S.M. Hunt, E.M. Werner, B. Huang, M.A. Hamilton, P.S. Stewart, Hypothesis for the role of nutrient starvation in biofilm detachment, Appl. Environ. Microbiol. 70 (2004) 7418–7425.

[26] A. Joss, N. Derlon, C. Cyprien, S. Burger, I. Szivak, J. Traber, H. Siegrist, E. Morgenroth, Combined nitritation-ANAMMOX: Advances in understanding process stability, Environ. Sci. Technol. 45 (2011) 9735–9742.

Appendix

A brief description of the pilot plant follows: Each biological activated carbon reactor column (1–8)has a volume of $0.1^2 \times 2.5 \times 3.14 = 0.0785\ \text{m}^3$. The system was designed to be flexible and up to eight columns were used to adjust the HRT. When not required, each column can be bypassed (piping not shown). The membrane reactor (9#) has an operating volume of $0.83 \times 0.47 \times 2.1 \times 30\%\text{m}^3$ (The factor of 30% arises because of the volume taken by the membrane module $0.805\ \text{m} \times 0.45\ \text{m} \times 1.77\ \text{m} = 0.246\ \text{m}^3$.) Upon optimization, it was found that just one reactor column (8#) combined with membrane reactor (9#) was good enough for removal of the impurities. So, initially this study was conducted with only one reactor column(8#) and membrane reactor (9#). Sustainable flux was found during a prior laboratory study to be less than 10 L/m^2 h. When the MBR was operated at 9.17 L/m^2 h, the HRT was ≈34 min.(7.13mins (8#) + 26.83 min (9#)). This increased as flux declined and additional columns were introduced. Initially, 317 g PAC was added to the reactor. Sludge retention time (SRT) was 20 d maintained by discharging 1/20 of the total volume every day. PAC (16 g) was initially added after discharge to balance the PAC of 20 mg/L and changed as the flux decline. Recycling flux was set at 1 m^3/h. Before the particular study reported here was conducted starting on 28 May 2010, the system had been operated from 15 January 2010 to optimize operating conditions (reflux, PAC addition, HRT (adjust by changing number of reactor columns), SRT, and aeration mode.) The details of the preliminary work can be found in [11].



instruction	
1	submerged pump
2	high level cistern
3	surge tank
4	BAC reactor pillars
5	membrane bioreactor
6	reflux pump
7	blower
8	suction pump

Fig. A.1. Schematic of the pilot plant MBR system setup.

Table A.1
Water quality and efficiency of impurities removal from 28 May to 30 December

Constituent	Influent	Effluent (treated by MBR)	Removal efficiency (%)
DOC (mg/L)	3.22 ± 0.37 (2.85–3.59)	1.61 ± 0.27 (1.34–1.88)	50 ± 9.2
UV_{254} (cm^{-1})	0.052 ± 0.003 (0.049–0.055)	0.021 ± 0.004 (0.017–0.025)	60 ± 9
SUVA	0.0165 ± 0.0025 (0.014–0.019)	0.0125 ± 0.0015 (0.011–0.014)	24.2 ± 12.3
THMFP (μg/L)	74.5 ± 9.5 (65–84)	11.3 ± 7.6 (3.7–18.9)	84.9 ± 9.8
HAAFP (μg/L)	43 ± 11.5 (32.5–54.5)	4.3 ± 2.6 (1.7–6.9)	90 ± 6.9
AOC (μg/L)	531.1 ± 89.8 (441.3–620.9)	126.5 ± 37.2 (89.3–163.7)	75.8 ± 9.8
pH	8.2 ± 0.2 (7.8–8.4)	8.0 ± 0.2 (7.8–8.4)	

Table A.2
Duration and rate of temperature corrected permeability loss of each pseudo steady state

Pseudo steady state	Temperature (℃)	Flux (L/m^2 h)	J_{20} (L/m^2 h)	Duration (d)	Permeability loss [(L/m^2 h) per bar per day]
PSS I	20–28.6	9.17	9.17–7.53	51 (5/28–7/17)	0.322
Transition 1	28.6–28.2	9.17–5.83	7.95–4.83	8 (7/17–7/24)	3.9
PSS II	28.2–28.5	5.83	4.83–4.8	11 (7/24–8/3)	0.027
Transition 2	28.5–29.1	5.83–5	4.8–4.06	1 (8/3–8/4)	7.4
PSS III	29.1	5	4.06	21 (8/4–8/24)	0
Transition 3	29.1–25.8	5–4.17	4.06–3.64	3 (8/24–8/26)	1.4
PSS IVa	25.8–26.1	4.17	3.643.62	23 (8/26–9/17)	0.009
PSS IVb	26.1–6.5	4.17	3.62–6.02	72 (9/17–11/28)	−0.333
Transition 4	6.5–5.5	4.17–2.5	6.02–3.72	9 (11/28–12/6)	0.256
PSS Va	5.5–1	2.5	3.72–4.31	11 (12/6–12/16)	−0.536
PSS Vb	1	2.5	4.31	25 (12/16–12/30)	0