Evaluating the effects of granular and membrane filtrations on chlorine demand in drinking water

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Abstract

In this study, chlorine decay experiments were conducted for the raw water from Nakdong River that is treated by Chilseo Water Treatment Plant (CWTP) situated in Haman, Korea as well as the effluents from sand and granular activated carbon (GAC) filters of CWTP and fitted using a chlorine decay model. The model estimated the fast and slow reacting nitrogenous as well as organic/inorganic compounds that were present in the water. It was found that the chlorine demand due to fast and slow reacting (FRA and SRA) organic/inorganic substances was not reduced significantly by sand as well as GAC filters. However, the treated effluents from those filters contained FRA and SRA that are less reactive and had small reaction rate constants. For the effluents from microfiltration, ultrafiltration, and nanofiltration the chlorine demand because FRA and SRA were further reduced but the reaction rate constants were larger compared to those of sand and GAC filter effluents. This has implications in the formation of disinfection by products (DBPs). If DBPs are assumed to form due to the interactions between chlorine and SRA, then it is possible that the DBP formation potential in the effluents from membrane filtrations could be higher than that in the effluents from granular media filters.

Key words: chlorine decay model; initial concentration; nitrogenous compounds; organic and inorganic compounds; reaction rate constants

Introduction

Drinking water starts its journey within catchments, and is subsequently purified at treatment plants and delivered through distribution systems. The water quality generally deteriorates during its passage through the catchment due to the accumulation of organic and inorganic dissolved and suspended pollutants, but the subsequent treatment processes produce water of high quality. However, within distribution systems, the quality of the water generally deteriorates again. A major problem that occurs in distribution systems is the decay of disinfectant due to the residual dissolved substances including natural organic matters (NOM) present in the water after the treatment as well the physical status such as pipe materials, pipe age, stagnant regions etc. of the distribution systems. This allows the re-growth of microorganisms in the distribution systems. A model that describes the changes in the quality of water that occur in distribution systems is needed to determine how much to enhance the treatment processes or to improve distribution systems so that microbiological criteria are met. Improvement of a distribution system can be in several forms, such as installing re-chlorination stations and cleaning mains or service reservoirs (Jegatheesan *et al*., 2004; Piriou *et al*., 1997; Butterfield *et al*., 2002).

NOM that are present in water bodies generally enter from catchment runoffs. NOM could be subdivided into dissolved organic matters (DOM) and particulate organic matters (POM) (Eikebrokk *et al*., 2006). The molecules of DOM are less than 0.20 to 0.45 μm and that of POM are above 0.45 μm. DOM is composed of humic and non-humic substances. Humic substances are hydrophobic and are easily removed by conventional water treatment processes (coagulation/flocculation/sedimentation and filtration). However, the non-humic substances which are also called as biodegradable organic materials (BOM) are hydrophilic and not easy to be removed by conventional water treatment processes. Some of the hydrophilic components (hydrophilic neutrals) are hardly removed by coagulation and flocculation but the others (charged hydrophilic compounds) are removed when the water is under-dosed or over-dosed with coagulants (Chow *et al*., 2004; Bolto *et al*., 1999). Furthermore, the charged hydrophilic compounds were found to contribute significantly for the formation of disinfectant by products (DBPs) (Weragoda, 2005). BOM are also composed of biodegradable dissolved organic carbon (BDOC) and assimilable organic carbon (AOC). While the BDOC is mineralized by the heterotrophic bacteria present in the water distribution system, AOC is converted to cell mass by the same heterotrophic bacteria to form biofilm on the inner walls

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of the distribution system pipes. POM are composed of algae, bacteria, particulate detritus, organic matter within flocs and inorganic particles covered by NOM and could be removed by conventional water treatment processes. Thus, evaluating the efficiency of a water treatment plant in treating NOM is essential in order to optimize the chlorine dosage for disinfection as well as to minimize the formation for DBPs. A rapid and simple method to analyze the NOM present in the water would help most of the treatment plants which may have limited analytical facilities.
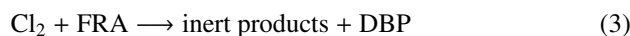
In this article the chlorine decay kinetics is modeled to compare the chlorine demand in the raw and treated water and thus provide an indication on the level of NOM present in those waters. This model can estimate the magnitude of fast and slow reducing (organic and inorganic) compounds and nitrogenous compounds present in the water that consumes chlorine. The common inorganic substances in the water are Fe^{2+}, Mn^{2+}, and H_2S (Jegatheesan *et al*., 2003). The model can also estimate the reaction rates of those compounds when they react with chlorine. Thus, the model will be able to predict the high initial chlorine demand (due to its interaction with fast reducing organic/inorganic and nitrogenous compounds) and the continuing demand (due to the interaction of chlorine with slow reducing organic/inorganic and nitrogenous compounds) of raw or treated water. The model prediction of initial chlorine demand of the treated water and the actual initial chlorine demand at the treatment plant could be used in finding the elements that are contributing to the consumption of chlorine at the treatment plant other than the water itself (such as pumps and pipes at the treatment plant); this allows better control of chlorine dosing at the treatment plant, which in turn will reduce the formation of DBPs. In addition, the model will accurately predict the decay due to the organic/inorganic and nitrogenous compounds that are remaining in the water at any point in the distribution system, if the travel time of water to the point from the treatment plant is known. Thus, the difference between the chlorine level predicted by the model and the actual chlorine level at the point will indicate the status of the distribution system with respect to its chlorine consumption (due to biofilm and corrosion). Further, if re-chlorination is introduced in the distribution system at the downstream of the treatment plant the model will predict the chlorine decay due to the slow reacting organic/inorganic and nitrogenous compounds accurately.

This study was conducted to evaluate the performances of sand and granular activated carbon (GAC) filters of Chilseo Water Treatment Plant (CWTP) in Masan, Korea in removing chlorine demand compounds from source water. The effectiveness of microfiltration (MF), nanofiltration (NF) and reverse osmosis (RO) in removing chlorine demand compounds that were present in the treated water from CWTP was also evaluated by passing the treated water through MF, NF, and RO separately.

1 Chlorine decay model

There are several models for chlorine decay and disinfectant by-product formation available in the literature (Biswas *et al*., 1993; Boccelli *et al*., 2003; Clark, 1998; Sohn *et al*., 2004). In this study, a simple and effective model proposed in the literature is used (Kastl *et al*., 1999). The following expressions are used in the model to represent the reactions of chlorine with fast reducing agents (FRA), slow reacting agents (SRA), fast reducing nitrogenous compounds (FRN) and slow reacting nitrogenous compounds (SRN). Chlorine in the form of free chlorine (denoted as Cl_2) is considered to react with the above four agents. FRA would generally consist of inorganic compounds associated with Fe^{2+} and Mn^{2+} as well as organic compounds. SRA consist of organic compounds, as most of the inorganic compounds will react with chlorine rapidly. FRN will consist of compounds associated with NH_4^+ as well as other organic-nitrogen compounds and SRN will mostly consist of organic-nitrogen compounds. Both FRN and SRN will react with chlorine and produce combined chlorine (denoted as CC). The sum of free and combined chlorine will be the total chlorine (denoted as TC). The organic compounds of FRA and SRA are considered as the DBP formation precursors.

$$Cl_2 + FRN \longrightarrow CC \tag{1}$$

$$Cl_2 + SRN \longrightarrow CC \tag{2}$$

$$Cl_2 + FRA \longrightarrow \text{inert products} + DBP \tag{3}$$

$$Cl_2 + SRA \longrightarrow \text{inert products} + DBP \tag{4}$$

$$CC \longrightarrow \text{inert products} \tag{5}$$

While Cl_2 indicates the available free chlorine present in the water, the sum of Cl_2 and CC indicates the total chlorine present in the water. The reaction rates for the above five equations are k_1, k_2, k_3, k_4, and k_5 respectively. Thus the equations governing the decay of free and combined chlorine can be written as:

$$\frac{dC_{Cl_2}}{dt} = -k_1 C_{Cl_2} C_{FRN} - k_2 C_{Cl_2} C_{SRN} - \\ k_3 C_{Cl_2} C_{FRA} - k_4 C_{Cl_2} C_{SRA} \tag{6}$$

$$\frac{dC_{CC}}{dt} = -k_5 C_{CC} + k_1 C_{Cl_2} C_{FRN} + k_2 C_{Cl_2} C_{SRN} \tag{7}$$

$$C_{TC} = C_{Cl_2} + C_{CC} \tag{8}$$

These processes were constructed in AQUASIM which contains a dynamic equation solver that can perform parameter estimation to find the best fit of the model output to the experimental data (Reichert, 1994). In this study the experimental chlorine decay data were used to estimate the initial concentrations of fast and slow reducing organic/inorganic and nitrogenous compounds along with the reaction rates k_1 through k_5. The weighted error between experimental and model (χ^2) data can be used as a measure of goodness of fit of the model to the experimental data and can be defined as in Eq. (9)

$$\chi^2(p) = \sum_{i=1}^{n} \left(\frac{f_{\text{meas},i} - f_i(p)}{\sigma_{\text{meas},i}} \right)^2 \tag{9}$$

where, $f_{meas,i}$ is the ith measured value, $f_i(p)$ is the calculated value from the model, using parameter values p. $\sigma_{meas,i}$ is the estimated standard deviation of $f_{meas,i}$. During the fitting of the model to the experimental data, the initial concentrations of fast and slow reducing organic/inorganic and nitrogenous compounds along with the reaction rates k_1 through k_5 were adjusted by the AQUASIM software until χ^2 reached a minimum value. The initial concentrations of FRA, FRN, SRA, and SRN were obtained as mg/L of chlorine equivalent.

2 Materials and methods

Water samples for chlorine decay experiments were collected from CWTP, which takes raw water from nearby Nakdong River and treats it by pre-ozonation, coagulation, flocculation, sedimentation, filtration, intermediate ozonation, and GAC adsorption. Two liter of samples were collected from (i) raw water, (ii) treated water after sand filtration, (iii) treated water after GAC filtration, (iv) treated water by MF, (v) treated water by NF, and (vi) treated water by RO. The MF membrane is made of PVDF (Kolon) with pore size of 0.1 μm. Both NF and RO membranes are made of polyamide (Saehan). The NaCl rejection rates of NF and RO are 85%–95% and 99.2%, respectively. The raw water samples were dosed with 3 and 5 mg/L of chlorine and other samples were dosed with 1 and 2 mg/L of chlorine. Dark brown bottles were used to prevent light penetration or algal growth and thus to obtain the decay of chlorine due to reducing compounds present in water samples. Free and total chlorine concentrations were

measured immediately after dosing (1 min), then at short intervals for the first 1 h and at longer intervals for until the total chlorine concentration decreases to 0.1 mg/L. Chlorine concentrations were determined using the DPD colorimetric method according to the standard methods (APHA, 1998).

3 Results and discussion

3.1 Reaction rate constants and initial concentrations

Table 1 shows the water quality of raw water and the effluents from sand filter, GAC filter, MF, NF, and RO. As can be seen from Table 1, the water quality, in terms of turbidity, total organic carbon (TOC), conductivity and UV_{254} absorption, improves significantly after the treatment. When raw water was dosed with 3 mg/L, after 7 h the free and total chlorine concentrations were 0.19 and 0.21 mg/L, respectively. Similarly, when the raw water was dosed with 5 mg/L of chlorine, after 23.5 h the free and total chlorine concentrations were again 0.19 and 0.21 mg/L. When the sand filter effluent was dosed with 1 and 2 mg/L chlorine, the free (or total) chlorine concentrations were 0.02 and 0.08 mg/L after 48 and 96 h, respectively. For the GAC filter effluents that were dosed with 1 and 2 mg/L of chlorine, the free (or total) chlorine concentrations were 0.08 and 0.01 mg/L after 30 and 144 h, respectively. Fig. 1 shows these experimental data of chlorine decay along with the model fittings.

The raw water that enters CWTP obviously consisted of all four compounds (FRA, SRA, FRN and SRN) as the total chlorine measurements were due to both free
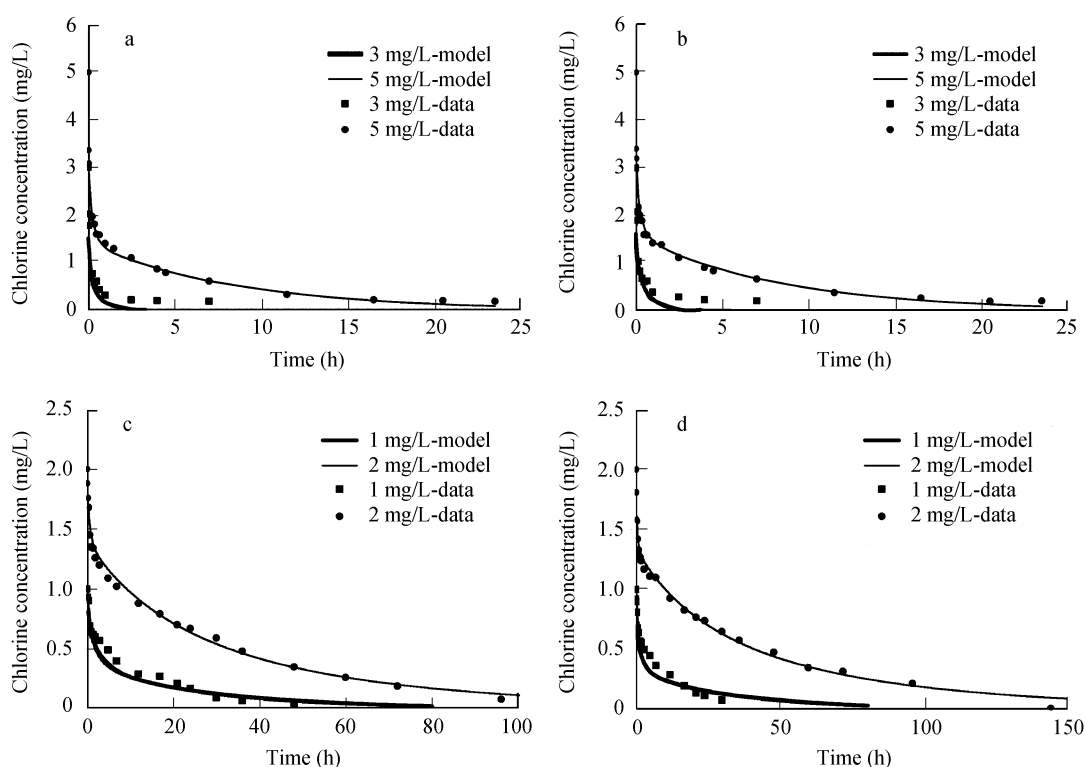
Fig. 1 Model fitting to experimental chlorine decay data. (a) free chlorine of raw water; (b) total chlorine of raw water; (c) free (or total) chlorine of sand filter effluent; (d) free (or total) chlorine of GAC effluent.

Table 1   Quality of raw water and effluent from granular media and membrane filtrations

Test water	pH	Turbidity (NTU)	Conductivity (µS/cm)	TOC (mg/L)	UV_{254}
Raw water	7.7	15.6	223	2.80	0.121
Sand filter effluent	7.1	0.08	234	1.85	0.031
GAC effluent	7.1	0.10	237	1.41	0.024
MF effluent	7.5	0.23	218	NA	NA
NF effluent	7.4	0.31	88	NA	NA
RO effluent	5.8	0.55	48	NA	NA

GAC: granular activated carbon; MF: microfiltration; NF: nanofiltration; RO: reverse osmosis.
NA: not analysed.

and combined chlorine. As can be seen from Table 2, the model estimated a total chlorine demand of 61.3 mg/L out of which 57.8 mg/L were due to SRN. The demand of chlorine due to FRA and SRA together was 3.40 mg/L. The FRA reaction rate constant was the highest at 171.80 L/(mg·h) while the second highest reaction rate constant was 23.61 L/(mg·h) due to the reaction of FRN (Table 2).

After the treatment, however, there was no combined chlorine concentration in the samples, indicating there were no nitrogenous compounds present in the treated water. Thus the concentrations of FRN and SRN were zero for all the samples tested except the raw water sample and the free and total chlorine concentrations were same for each time a sample was analysed. From the model it could be found that the chlorine demand due to both FRA and SRA was not reduced by sand or GAC filtration significantly and was around 3.18 (sand filter effluent) and 2.87 mg chlorine equivalent/L (GAC

effluent). However, the reaction rate constants of those compounds were reduced significantly. For example, the reaction rate constant of FRA compounds was reduced from 171.80 (raw water) to 1.00 (sand filter effluent) and 1.22 L/(mg·h) (GAC effluent). Similarly, the reaction rate constant of SRA compounds was reduced from 1.93 (raw water) to 0.015 (sand filter effluent) and 0.014 L/(mg·h) (GAC effluent). Thus, the rate of decay of chlorine in the GAC treated water would be slow due to small reaction rate constants rather than the initial concentrations of FRA and SRA. These values are summarized in Table 2.

When the MF effluent was dosed with 1 and 2 mg/L of chlorine, free (or total) chlorine concentration was 0.06 and 0.18 mg/L after 60 and 72 h, respectively. Similarly, when the NF effluent was dosed with 1 and 2 mg/L of chlorine, free (or total) chlorine concentration was 0.14 and 0.78 mg/L after 72 h. For the RO effluent that was dosed with 1 mg/L of chlorine, free (or total) chlorine concentration was 0.18 mg/L after 72 h. Fig. 2 shows these experimental data of chlorine decay along with model fittings.

When the MF, NF, and RO treated effluents were analysed for chlorine demand it was found the reaction rate constants for FRA compounds were large (89.20, 208.0, and 62.70 L/(mg·h), respectively). The reaction rate constants for SRA compounds presented in those waters were 0.070, 0.076 and 0.18 L/(mg·h), respectively. However, the initial concentrations were reduced significantly when the MF, NF, and RO treated effluents were analysed and they were 1.77, 1.17, and 0.76 mg chlorine equivalent/L, respectively (Table 2).
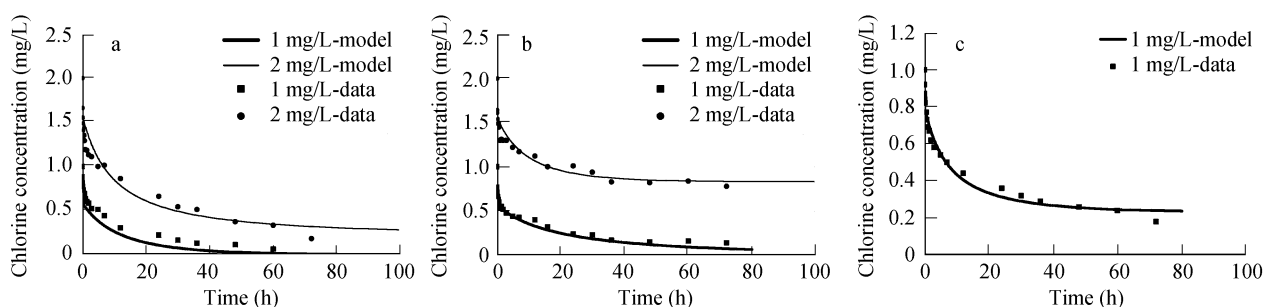
Fig. 2   Model fitting to experimental chlorine decay data. (a) free (or total) chlorine of MF effluent; (b) free (or total) chlorine of NF effluent; (c) free (or total) chlorine of RO effluent.

Table 2   Reaction rate constants of Reactions (1) to (5), initial concentrations of fast and slow reacting agents and nitrogenous compounds

Parameter	Raw water	Sand filter effluent	GAC filter effluent	MF effluent	NF effluent	RO effluent
k_1 (L/(mg·h))	23.61	–	–	–	–	–
k_2 (L/(mg·h))	0.002	–	–	–	–	–
k_3 (L/(mg·h))	171.80	1.00	1.22	89.20	208.0	62.70
k_4 (L/(mg·h))	1.93	0.015	0.014	0.07	0.076	0.18
k_5 (L/(mg·h))	1.194	–	–	–	–	–
FRN-init (mg/L)	0.135	–	–	–	–	–
SRN-init (mg/L)	57.76	–	–	–	–	–
FRA-init (mg/L)	1.35	0.58	0.64	0.42	0.43	0.20
SRA-init (mg/L)	2.05	2.60	2.23	1.35	0.74	0.56
χ^2	1.30	0.29	0.36	1.26	0.37	0.04

–: the combined chlorine did not form during the chlorination of effluents from the granular media and membrane filtration. Therefore, the reaction rate constants as well as the initial concentrations of FRN and SRN and the reaction rate constant of combined chlorine do not have any values.

3.2 Formation of disinfectant by-products (DBPs)

The model used in this study would also help to estimate the DBPs formed in different samples due to chlorination. In this study two cases were assumed: (i) both FRA and SRA are responsible for the formation of DBPs and 5% of each FRA and SRA reacted would produce DBPs, and (ii) only SRA are responsible for the formation of DBPs and 5% of SRA reacted would produce DBP (Weragoda, 2005). Figures 3 and 4 show the concentrations of DBPs for raw water dosed with 1 and 3 mg/L of chlorine and for the effluents from media and membrane filters dosed with 1 mg/L of chlorine. When both FRA and SRA contribute to the formation DBPs, both the sand and GAC filter effluents could produce 50 μg/L of DBPs over a period of 80 h (Fig. 3a), while MF, NF, and RO effluents could produce 50, 47, and 38 μg/L of DBPs (Fig. 3b), respectively. On the other hand, when only SRA contributes to the formation of DBPs, as can be seen in Fig. 4a, raw water produced only a small amount of DBPs as most of the chlorine dosed is consumed by FRA and FRN and little interacted with SRA. Over a period of 80 h sand filter effluent and GAC filter effluent generated around 20 and 15 μg/L of DBPs, respectively. However, the effluents produced by MF, NF, and RO generated more than 25 μg/L of DBPs over a period of 80 h. High SRA reaction rate constants obtained for membrane effluents are one of the reasons for this prediction. Another reason is due to the assumption that the SRA present in all samples were similar and 5% of this SRA reacted was assumed to produce DBPs in all effluents. However, this may not be true and therefore further experiments should be conducted to evaluate the amount of DBPs produced by different effluents.

3.3 Simulations of chlorine decay and the formation of DBPs

The model used in this study would also allow simulating the chlorine decay due to FRA and SRA in granular media and membrane filter effluents when they are put into a distribution system. The first set of simulations were carried out supposing that all the effluents are dosed with 1 mg/L of chlorine initially and re-chlorinated with another 1 mg/L of chlorine at a service reservoir after 24 h. The corresponding chlorine decay and the formation of DBPs are shown in Fig. 5. It can be seen from Fig. 5a, that the RO effluent had the highest residual chlorine (of 1.24 mg/L) after 80 h followed by NF effluent (0.84 mg/L). Effluents of MF, GAC, and sand filter had similar residual chlorine after 80 h (0.34, 0.35 and 0.27 mg/L, respectively). Similarly, the DBPs due to 5% of each FRA and SRA by-products provided the highest value for MF, GAC, and sand filter effluents (83.1, 82.3, and 86.7 μg/L, respectively) after 80 h. NF and RO effluents produced 58.1 and 38.0 mg/L of DBPs, respectively during the same time period (Fig. 5b). Thus, RO and NF effluents could be re-chlorinated with less than 1 mg/L of chlorine to obtain residual chlorine concentrations that are similar to the residual chlorine concentrations in MF, GAC, and sand filter effluents after 80 h. Fig. 6 shows the residual chlorine and DBP concentrations in the effluents after 80 h where MF, GAC, and sand filter effluents were re-chlorinated
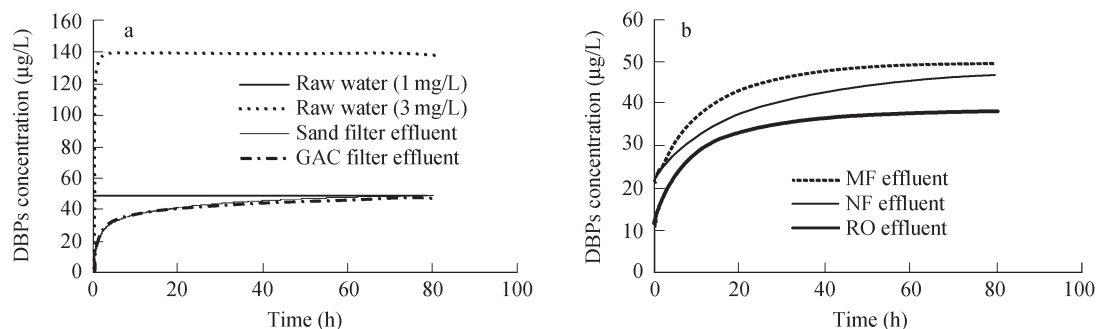

Fig. 3   Model prediction of disinfectant by-product (DBP) formation due to both FRA and SRA.
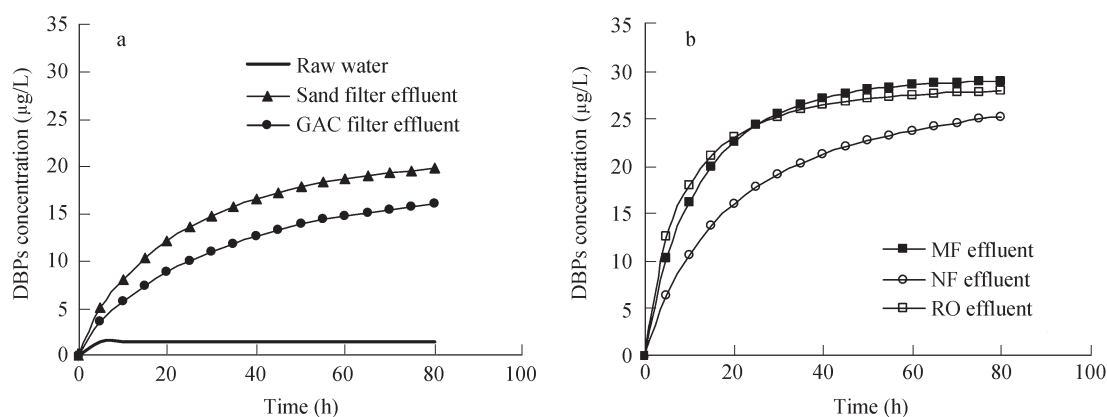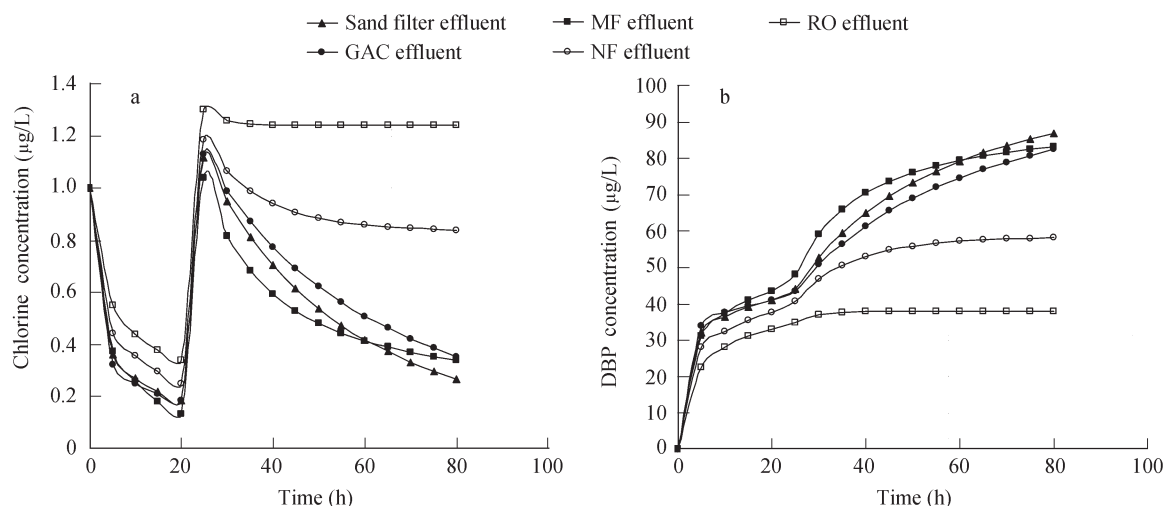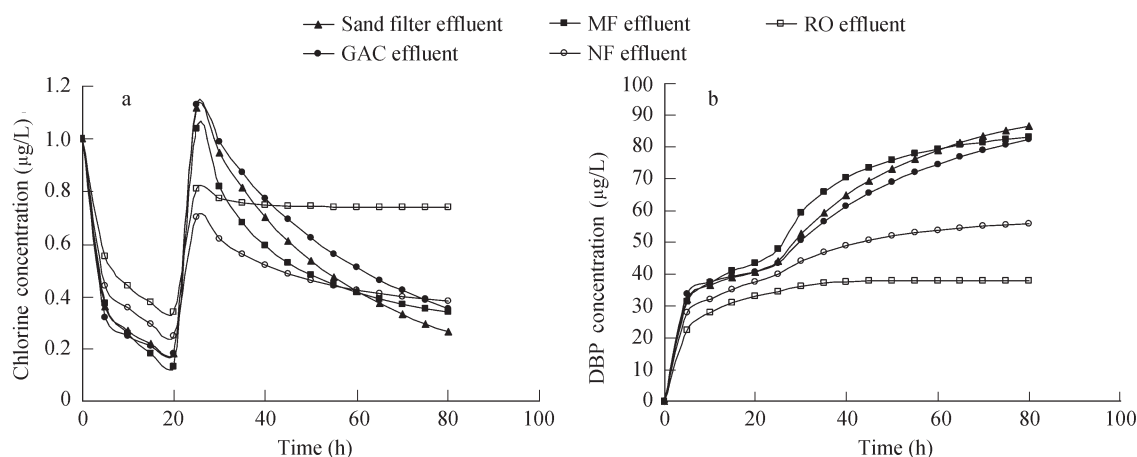

Fig. 4   Model prediction of disinfectant by-product formation due to SRA only.

Fig. 5 Model prediction of residual chlorine (a) and DBP (b) concentrations in a distribution system due to re-chlorination (1 mg/L) after 80 h.

Fig. 6 Model prediction of residual chlorine (a) and DBP (b) concentrations in distribution system due to re-chlorination (1 mg/L for MF, GAC and sand filter effluents and 0.5 mg/L for NF and RO effluent) after 80 h.

with 1 mg/L of chlorine and NF and RO effluent were re-chlorinated with 0.5 mg/L of chlorine. In this case, after 80 h, the RO and NF effluents had 0.74 and 0.38 mg/L of residual chlorine, respectively. However, there was no significant reduction in residual DBP concentrations in RO and NF effluents after 80 h as there was sufficient concentration of chlorine dose was used (0.5 mg/L) during re-chlorination to react with most of the SRA.

4 Conclusions

The performances of sand and granular activated carbon (GAC) filters of Chilseo Water Treatment Plant (CWTP) situated in Masan, Korea were evaluated for their effectiveness in removing chlorine demand compounds from source water. The effectiveness of microfiltration (MF), nanofiltration (NF) and reverse osmosis (RO) in removing chlorine demand compounds that were present in the treated water from CWTP was also evaluated. Chlorine demand was estimated by fitting the chlorine decay data to a model that describes the chlorine decay due to fast and slow reacting nitrogenous and organic/inorganic compounds.

This study found that chlorine demand due to both fast and slow reacting organic/inorganic compounds was not reduced by sand or GAC filtration significantly and was around 3.40 (raw water), 3.18 (sand filter effluent), and 2.87 mg chlorine equivalent/L (GAC effluent). However, the reaction rate constants of those compounds were reduced significantly. For example, the reaction rate constant of fast reacting organic/inorganic compounds was reduced from 171.80 (raw water) to 1.00 (sand filter effluent) and 1.22 L/(mg·h) (GAC effluent). Similarly, the reaction rate constant of slow reacting organic/inorganic compounds was reduced from 1.93 (raw water) to 0.015 (sand filter effluent) and 0.014 L/(mg·h) (GAC effluent). Thus, the rate of decay of chlorine in the GAC treated water would be slow due to small reaction rate constants rather than the initial concentrations of the fast and slow reacting organic/inorganic compounds.

When the MF, NF, and RO treated effluents were analysed for chlorine demand it was found the reaction rate constants for fast reacting compounds were large (89.2, 208.0 and 62.70 L/(mg·h), respectively); the reaction rate constants for slow reacting compounds presented in those

waters were 0.07, 0.076 and 0.18 L/(mg·h), respectively. However, the initial concentrations were reduced significantly when the MF, NF, and RO treated effluents were analysed and they were 1.77, 1.17 and 0.76 mg chlorine equivalent/L, respectively. This has implications in the formation of disinfection by products (DBPs). If DBPs are assumed to form due to the interactions between chlorine and SRA, then it is possible that the DBP formation potential in the effluents from membrane filtrations could be higher than that in the effluents from granular media filters. However, the assumption here is that the percentage of slow reacting organic compounds that is converted to DBP is the same for all effluents, which needs further investigation.

Acknowledgments

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