Received: 30 September 2022 | Revised: 22 December 2022 | Accepted: 23 January 2023

DOI: 10.1002/wer.10840

RESEARCH ARTICLE

water
ENVIRONMENT RESEARCH

Analysis of the evolution of ultra-filtered water quality in a drinking water distribution system by particle size distribution: Influence of pre-ozonation

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Funding information
Spanish Ministry of Science and Innovation, Grant/Award Number: CTM2010-18899-TECNO

Abstract

An experimental drinking water distribution system (DWDS) was used to evaluate the evolution of particle size distribution (PSD) and basic quality parameters of ultrafiltered water with or without pre-ozonation. An ultrafiltration (UF) module was set up, associated with a pre-ozonation system (3.7 g O_3/m^3). The permeate was circulated in the DWDS (300 m; 0.9 m/s) with 0.4 mg/L of chlorine, and the analysis of the PSD was performed using a β-variable mathematical model. A better control of membrane fouling was obtained with pre-ozonation, and PSD was necessary to observe water quality differences between permeates and in the DWDS. A decrease in particle concentration of 1.8 logarithms was obtained with the application of UF membranes, while a decrease of only 1.2 logarithms was obtained with pre-ozonation. The system without pre-ozonation showed a higher efficiency at removing smaller particles (around 2 μm), with the absence of particles larger than 23 μm during both stages. The PSD revealed a worsening of water quality in the DWDS with an increase of particles smaller than 5 μm during the application of UF membranes, while with pre-ozonation, all particle sizes analyzed increased their concentration.

Practitioner Points

- Pre-ozonation led to a better control of membrane fouling, but a worsening of permeate quality according to particle size distribution.
- Pre-ozonation does not improve the turbidity, dissolved organic carbon or UV254 removal capacity of ultrafiltration during drinking water treatment.
- Particles size distribution reveals the deterioration of water quality in a drinking water distribution system better than turbidity or DOC.
- Ozone prior to ultrafiltration membranes led to a worsening of permeate quality, more significant in the drinking water distribution system.

KEYWORDS
drinking water distribution system, particle size distribution, pre-ozonation, UF membranes

INTRODUCTION

The use of ultrafiltration (UF) membranes for water potabilization has become a real alternative to conventional systems due to the capacity of the membranes for screening out microorganisms and particulate matter (Guo et al., 2010). However, their low capacity for dissolved organic matter removal and membrane fouling are two of the main drawbacks of drinking water production by UF processes (Rojas-Serrano et al., 2015; Shi et al., 2014).

Several mechanisms take place during membrane fouling, such as concentration polarization, adsorption, pore blocking, and cake layer formation. The matter in the feed solution can form deposits on the membrane surface or in membrane pores due to several physicochemical mechanisms, which can lead to the development of reversible or irreversible fouling (Shi et al., 2014). The main consequence of these processes is a decrease of water flux through the membrane and loss of performance efficiency.

To overcome drawbacks of drinking water production by UF processes, pre-treatments such as ozonation have been applied (Li et al., 2022; Rojas-Serrano et al., 2016). Due to the high oxidative capacity of ozone, pre-ozonation can reduce the deposition of organic matter on or within membranes and can suppress the proliferation of microorganisms on the membrane surface. Therefore, pre-ozonation slows down membrane cake formation, acting as an effective technology for UF membrane fouling control (Li et al., 2022). However, pre-ozonation could negatively influence the permeate quality of UF membranes. The membrane cake layer acts as a prefilter that screens out particles and molecules, thus improving permeate quality (Farahbakhsh & Smith, 2004; Szymanska et al., 2014). In addition, due to molecular fragmentation caused by ozone (Zhong et al., 2017), its application increases the presence of smaller and more metabolizable organic compounds, which can pass easily through the UF membranes.

The final quality of drinking water in the consumers' tap depends not only on the drinking water treatment but also on its route through the drinking water distribution system (DWDS). In general, the quality of drinking water deteriorates in the DWDS (Vreeburg & Boxall, 2007). The accumulation of particles in the DWDS can be re-suspended or carried by the flow, affecting water quality (Verberk et al., 2007; Vreeburg et al., 2008; Liu et al., 2013). The most important source for particles in the DWDS is water from the treatment plant due to incomplete removal, addition, or degradation of materials. The DWDS itself can also produce particles from materials' erosion or corrosion and chemical reactions (Vreeburg & Boxall, 2007).

Biofilm formation in the pipe wall or associated with deposits of particles and their detachment leads to deterioration of the water quality in the DWDS (Álvarez-Arroyo et al., 2022; Liu et al., 2013). The occurrence of these processes depends on several factors such as the presence of bacteria, availability of organic matter and nutrients, presence of residual disinfectants, and the characteristics and hydraulic conditions of the DWDS (Liu et al., 2013).

Membrane technology has been shown to have a high bacteria removal capacity (Guo et al., 2010). However, several authors have observed microbiological contamination in the permeate zone of UF membrane due to the absence of sterile conditions during backwashing or failure of membrane integrity (Guo et al., 2010; Pérez et al., 2021). Biological stability of water in DWDS also depends on the presence of nutrients, and low dissolved organic carbon (DOC) removal is one of the main drawbacks of UF membranes (Rojas-Serrano et al., 2015). In this respect, it has been widely reported that an increase in organic carbon in DWDS stimulates the growth of bacteria in both transported water and biofilm (Liu et al., 2013).

One of the most widespread strategies to prevent biofilm development in DWDS is to maintain a constant chlorine concentration. Chlorine is effective in controlling the bacteriological quality of water or biofilm development, but while its application slows down such development, it does not prevent it altogether (Álvarez-Arroyo et al., 2022).

Both ultrafiltration and ozonation–ultrafiltration processes are suitable alternatives for drinking water production. However, when working with UF membranes only, faster membrane fouling can be expected than when membranes are combined with pre-ozonation (Rojas-Serrano et al., 2016). On the other hand, the effluent quality obtained by UF membranes may be better than that obtained when pre-ozonation is applied. Therefore, a different evolution of the quality of water in the DWDS may be expected depending on whether or not the membrane process is combined with pre-ozonation.

Organic matter concentration in water samples is usually represented by the measurement of parameters such as UV absorbance at 254 nm wavelength (UV_{254})

and total or DOC. These parameters, together with turbidity, can be considered as basic for drinking water quality determination, since their analysis allows us to assess the quality of the raw water or the effectiveness of both conventional treatments of potabilization and membrane processes (Matilainen et al., 2011). These parameters have also been applied to assess the evolution of drinking water quality in DWDS (Álvarez-Arroyo et al., 2015; Rojas-Serrano et al., 2016).

Deterioration of drinking water quality in a DWDS has been associated with the mobilization of accumulated particles from within the DWDS (Vreeburg et al., 2008). The analysis of the presence of particles may therefore be a good complement to the basic drinking water quality parameters to evaluate the evolution of the water quality in a DWDS.

Particle size distribution (PSD), which is closely related to the presence of particulate matter, is commonly used for the characterization of water when its quality is high (Byrne et al., 2014; Ceronio & Haarhoff, 2005). The PSD provides information about the absolute count of the various size ranges of particles present in water that is adequate to describe sudden changes in quality, mainly when membrane processes are applied (Guo et al., 2010).

Analyses of PSD in DWDS can be an important tool for water quality determination since they advise about aesthetic quality, problems derived from microbiological growth or chemical transformations (Byrne et al., 2014). Particles in the water of the DWDS have different origins, such as the treatment plant or through direct contribution, re-suspension, or detachment of particles from the pipeline walls or biofilm, so PSD could be useful for assessing the quality of the water and its temporal evolution (Verberk et al., 2007).

Accordingly, the aim of this study is to compare the quality of drinking water obtained by UF membranes with or without pre-ozonation and to determine its evolution in an experimental DWDS according to basic parameters (UV_{254}, DOC, and turbidity) and PSD.

MATERIALS AND METHODS

Experimental facility

The experimental facility (Figure 1) comprised a ring-filter macrofiltration pre-treatment phase (150 μm), a pre-ozonation, an ultrafiltration membrane system, and a

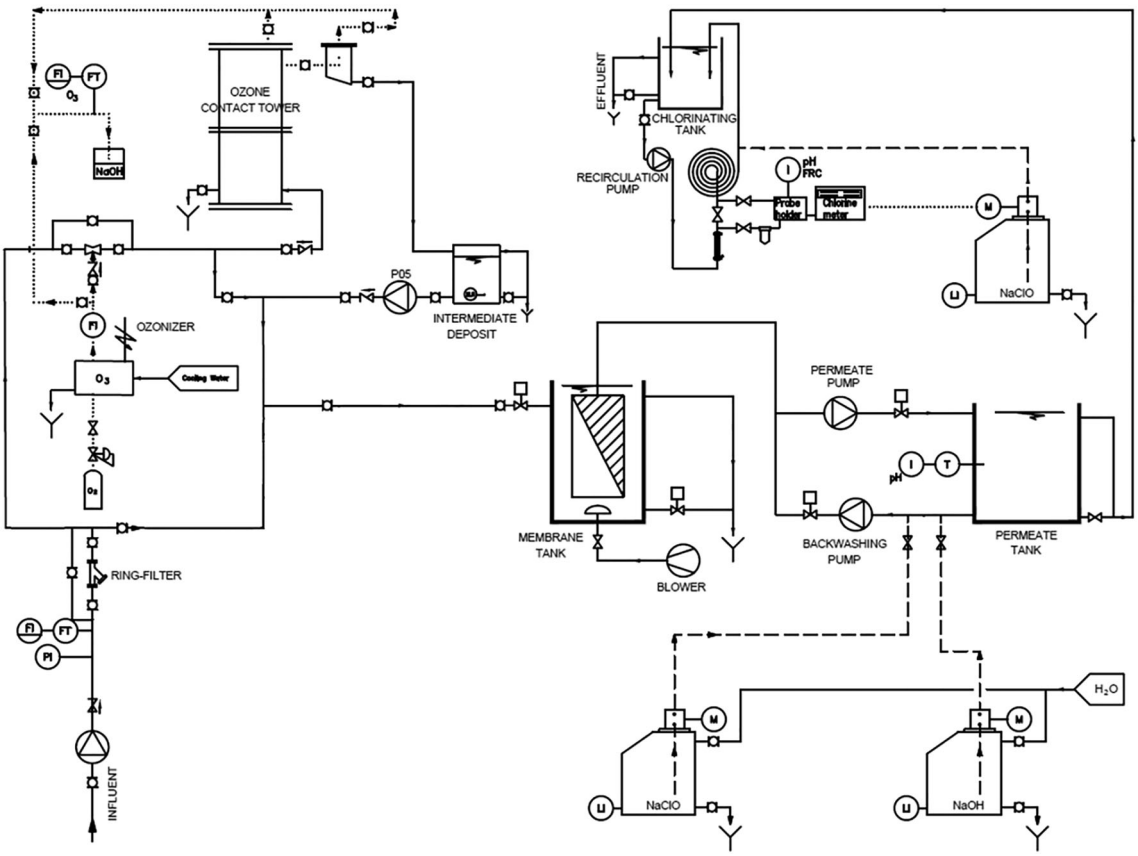

FIGURE 1 Schematic diagram of the experimental facility.

post-chlorination system, after which the treated water was re-circulated through an experimental DWDS.

The pre-ozonation system consisted of an ozone generator (C-L010DTI, AirTree Ozone Technology Co., Ltd., Taiwan) with a maximum ozone generation capacity of 10 g/h working with pure oxygen (Air Liquide, S.A.). The generated ozone was injected into the water by a Venturi system prior to a contact column. The water and gas were separated after the contact column by an overflow, and residual ozone was removed by a saturated solution of NaOH. The ultrafiltration module was equipped with submerged spiral-bound polyvinylidene fluoride membranes (SpiraSep 960, TriSep Corporation), with an effective pore size of 0.03 μm and 20.9 m^2 filtration area.

The post-chlorination system was composed of a chlorination tank, which received the ultrafiltrated water, an experimental DWDS simulated with 300 m of polyethylene pipe (internal diameter 14 mm) in which the permeate water was circulated by a centrifugal recirculation pump, and a chlorine management system (Kontrol800, Seko) which maintained the free residual chlorine (FRC) concentration according to the pH values by dosing NaClO (5%).

Experimental procedure

The whole study was conducted over 49 weeks and divided into two different stages: ultrafiltration (UF) and ozonation, followed by ultrafiltration (O_3-UF). After each stage, the membrane module in operation was removed and replaced with a new one, and the experimental DWDS was likewise replaced. The experimental facility treated surface water from the Canales reservoir in the province of Granada, Spain.

During the UF stage, the system worked continuously filtering the water by means of a permeate vacuum pump with a capacity of 1 m^3/h. Every 30 min of filtration, membranes were backwashed for 30 s with a flow 1.5 m^3/h of permeate. The membrane was continuously aerated by means of an air blower (15 Nm^3/h). Chemical cleaning (NaClO and NaOH) was carried out according to the threshold transmembrane pressure (TMP) established at −0.7 bar.

During the O_3-UF stage, the influent was injected continuously with O_3 and the ozonated water passed through the contact column working with a hydraulic retention time of 12 min. The ozone generator operated at 1.2 bar with an oxygen flow rate of 0.2 Nm^3/h. An in-line ozone analyzer (Mini-HiCon, In USA Corp., USA) was used to measure the ozone concentration in the gas line prior to injection and after overflow. The transferred ozone dose was adjusted to obtain a transferred ozone dose of 3.7 g O_3/m^3 according to Rojas-Serrano et al. (2016).

The UF membrane permeate water was circulated in the experimental DWDS at a velocity of 0.9 m/s, and a constant concentration of 0.4 mg FRC/L was maintained.

Throughout the experimental study, samples of raw water (influent), permeate water, and distributed water were taken daily to determine turbidity, UV_{254}, and DOC. Weekly samples were obtained for PSD analysis.

Analytical methods

Samples were taken in thoroughly cleansed plastic bottles (rinsed with particle-free water) and analyzed immediately. Turbidity was obtained by measurement of the diffused radiation (DINKO D-112). For the determination of UV_{254}, water samples were filtered through a filter of 0.45 μm prior to measurement by UV-visible spectrophotometer (Heλios γ) with a 1 cm quartz cell. DOC was measured using a combustion TOC Analyzer (FormacsTH, SKALAR).

The analysis of PSD was carried out with a particle counter for liquids (LiQuilaz-E20, Particle Measuring Systems). The equipment took a sample volume of 5 mL with a syringe and injected it through an optical particle sensor, which counts the particles according to size, with a total range of 2 μm to 125 μm, obtaining cumulative counts in the sample volume. All samples were analyzed with a constant scanning sensitivity of 1 μm until the particle cumulative count became zero.

The data obtained from the particle analyzer were adjusted to the β-variable model described by Ceronio and Haarhoff (2005), a mathematical model based on the power law model but with variable exponent based on the adjustment of the PSD to Equations (1) and (2), whereby the normalized particle counts (PC_n) were calculated by the ratio of the increment of cumulative counts (N) of two consecutive particle diameters (d_p) between the interval of these sizes.

$$PC_n = \left(\frac{\Delta N}{\Delta d_p}\right) = A \cdot d_p^{-\beta} \quad (1)$$

$$\beta = b \cdot \log d_p \quad (2)$$

The calculations consisted in the determination of parameters "A" and "b" of this adjustment, according to the methodology described by Ceronio and Haarhoff (2005). In the β-variable model, the coefficient "A" is related to the number of particles in suspension in the water sample (specifically in 0.5 μm < d_p < 1.5 μm),

while the parameter "b" is related to the extension of particle size and the curvature of the fit (related at the same time to the homogeneity of the distribution).

Statistical analysis

The data bank was treated and analyzed by Excel for the determination of parameters "A" and "b," as well as the calculation of Pearson correlation coefficient (r^2) of the fits, by programming a macro. IBM® SPSS® Statistics (v.21) for Windows was used for the analysis of variances (ANOVA) in order to determine the existence of statistically significant differences between the stages with a significance level of 5% under algorithms from the Student–Newman–Keuls (SNK) post hoc test.

RESULTS AND DISCUSSION

To carry out the comparative analysis between the two stages, a continuous operation time of 220 days was proposed for each one. During the O_3-UF stage, the membrane worked normally, but the UF stage lasted only 130 continuous days. During the last days of operation of the UF stage, the TMP reached the maximum operating values, which proved impossible to correct by means of intense chemical cleaning operations carried out with 1 g Cl_2/L at pH = 12. A better control of membrane fouling was achieved by pre-ozonation, as was previously observed by Rojas-Serrano et al. (2016). In contrast, for this type of influent and under the established operational conditions, irreversible membrane fouling occurred when working with the UF membrane as a single treatment.

Throughout the period of study, the quality of the influent was constant between stages with respect to turbidity and DOC, while differences were observed with respect to UV_{254} (Tables 1 and 2). This indicates differences in the chemical structure of the organic matter, with higher presence of hydrophobic aromatic compounds when the system was operating with pre-ozonation (Jegatheesan et al., 2009).

Turbidity of water improved between 50% and 89% both with single-treatment UF and with pre-ozonation. A similar removal turbidity rate was obtained by Rojas-Serrano et al. (2016), working with the same installation and the same influent. However, in their study, higher removal rates were obtained with the application of pre-ozonation, probably due to the higher transferred ozone dose applied. Although the potabilization of surface water by UF may guarantee constant water turbidity (Rojas et al., 2008), the characteristics of the influent and pre-treatment can affect the removal rates.

For most of the samples, permeate DOC concentrations were slightly lower than influent DOC concentrations, while in a small number of samples, permeate DOC concentrations were higher than influent DOC concentrations. This circumstance was observed in 5% of the sampling during the UF stage, with certain samples standing out due to their significantly high permeate DOC concentrations. These specific cases cause the mean

TABLE 1 Mean values of the water quality parameters of influent, permeate, and in the DWDS during the UF stage (n = 78).

		Influent				Permeate				DWDS				
Parameter	**Units**	**Mean**	**S.D.**	**Max.**	**Min.**	**Mean**	**S.D.**	**Max.**	**Min.**	**Mean**	**S.D.**	**Max.**	**Min.**	***p* value**
Turbidity	NTU	4.4[a]	4.1	18.5	2.3	1.6[b]	0.5	2.7	1.0	1.5[b]	0.3	2.1	1.1	0.0020
DOC	mg L^{-1}	7.1	1.9	10.8	5.1	8.6	5.1	20.0	4.6	7.8	2.0	12.4	4.8	0.4630
UV_{254}	m^{-1}	12.7[a]	3.9	17.3	1.3	9.9[b]	2.7	13.9	1.8	9.9[b]	3.2	15.1	2.2	0.0350

Note: A different letter (a, b) indicates statistically significant differences (SNK 0.05) between groups: influent, permeate, and DWDS.

TABLE 2 Mean values of the water quality parameters of influent, permeate, and in the DWDS during the O_3-UF stage ($n = 111$).

		Influent				Permeate				DWDS				
Parameter	**Units**	**Mean**	**S.D.**	**Max.**	**Min.**	**Mean**	**S.D.**	**Max.**	**Min.**	**Mean**	**S.D.**	**Max.**	**Min.**	***p* value**
Turbidity	NTU	4.0[a]	1.5	9.7	2.6	1.7[b]	0.4	2.3	1.1	2.0[b]	0.6	4.0	1.2	0.0001
DOC	mg L^{-1}	7.0	2.2	12.0	3.4	6.7	3.4	13.3	1.9	6.8	3.4	14.1	3.1	0.9480
UV_{254}	m^{-1}	18.3[a]	10.2	60.4	7.3	6.6[b]	2.1	12.7	3.6	5.9[b]	2.1	11.3	3.8	0.0001

Note: A different letter (a, b) indicates statistically significant differences (SNK 0.05) between groups: influent, permeate, and DWDS.

DOC concentration of the permeate to be higher than that of the influent during the UF stage. In spite of this, statistically significant differences between the concentrations of DOC were not observed in either stage indicating that DOC removal capacity was irrelevant. A low retention capacity for low molecular weight compounds can be expected for the UF system working as a single treatment (Rojas-Serrano et al., 2015; Shi et al., 2014). In addition, ozone causes fragmentation of organic matter, generating lower molecular weight compounds (de Vera et al., 2016), which can pass through the UF membrane more easily (Rojas et al., 2008).

A mean removal yield of 60% was observed for UV_{254} during the O_3-UF stage, while only 18% was achieved during the UF stage, with statistically significant differences in both cases (Tables 1 and 2). The powerful oxidant capacity of ozone causes a break of double bonds and aromatic cycles (de Vera et al., 2016), measured by UV_{254}, to produce smaller aliphatic compounds that are not measured by UV_{254}. This capacity is the reason for the decrease of UV_{254} values but not in values of DOC in the permeate obtained during the O_3-UF stage. Regarding the UV_{254} removal capacity during the UF stage, this may be due to the presence in the influent of high molecular weight aromatic compounds, which can be retained by the UF membrane.

Turbidity of permeate produced by O_3-UF worsened in the DWDS, which was not observed during the UF stage. However, no statistically significant differences between the values of permeate, and the water of the DWDS were observed in either of the stages (Tables 1 and 2).

As observed in previous studies (Álvarez-Arroyo et al., 2015), there were no statistically significant differences between the DOC and UV_{254} values for permeates and the DWDS during the two stages (Tables 1 and 2). However, during the O_3-UF stage, UV_{254} values in the DWDS decreased with respect to the permeate, although not significantly. A possible cause was the presence of residual ozone in the DWDS, which reduces the aromaticity of organic matter (de Vera et al., 2016).

On the basis of these results and the corresponding statistical analysis, parameters such as turbidity, DOC, and UV_{254}, do not allow us to detect significant variations in water quality in the DWDS after potabilization by UF membranes with or without ozone as pretreatment, despite the different mechanisms of action of the two applied processes.

In contrast to the results observed through turbidity or DOC, PSD showed a different quality of the influent during the two stages. A more heterogeneous particle count with higher particle numbers was observed during the UF stage. In addition, maximum particle sizes of 46 μm were detected during the UF stage, while particles over 42 μm in size were not detected during the O_3-UF stage (Table 3).

The influent PSD fitted a logarithmic regression with an increase in particle concentration as size decreases for both stages (Figure 2). Higher values of parameter "A" of the β-variable model were observed during the UF stage (Table 4), with a heterogeneous evolution over time in both stages (Figure 3). In addition, a statistically significant correlation (0.581 Pearson's coefficient) was obtained between turbidity and parameter "A" in influent samples during the O_3-UF stage but not during the UF stage. This result confirms the differences in the quality of the influent between the two stages. In contrast, the homogeneity of the PSDs obtained throughout the analysis time was similar during the two stages, so the values of parameter "b" of the β-variable model were similar over time (Figure 4).

TABLE 3 Average, minimum, and maximum particle count (total particles between 2 and 125 μm) and large particle size (μm) for each water stream in the experimental stages.

		Influent		Permeate		DWDS	
		Count	Max. size	Count	Max. size	Count	Max. size
UF ($n = 15$)	Mean	11,711[a]	29.9[a]	188[b]	17.3[b]	300[b]	17.4[b]
	S.D.	10,405	8.2	101	2.7	140	1.6
	Min.	79	13	42	11	83	15
	Max.	37,841	46	399	23	558	21
O_3-UF ($n = 24$)	Mean	9014[a]	27.5[a]	563[b]	17.2[b]	1872[b]	17.8[b]
	S.D.	7216	7.2	1103	3.8	5725	5.9
	Min.	712	14	81	10	6	3
	Max.	31,240	42	5027	23	28,549	35

Note: A different letter (a, b) indicates statistically significant differences (SNK 0.05) between groups: influent, permeate, and DWDS.

PSD analysis of the permeate showed the high efficiency of the tested processes in particulate matter removal, with a decrease of 1.8 logarithms with the application of UF membranes, while a decrease of 1.2 logarithms was obtained with pre-ozonation. A significant decrease in the number of particles for the smaller sizes (Figure 2) and the absence of particles larger than 23 μm during both stages was observed (Table 3). UF membranes have a high particle retention capacity achieved by sieving or adsorption mechanisms, which result in the accumulation and deposition of particles on the membrane surface. However, these mechanisms of action cause an increase of concentration polarization, thus promoting the development of a cake layer (Guo et al., 2010; Shi et al., 2014).

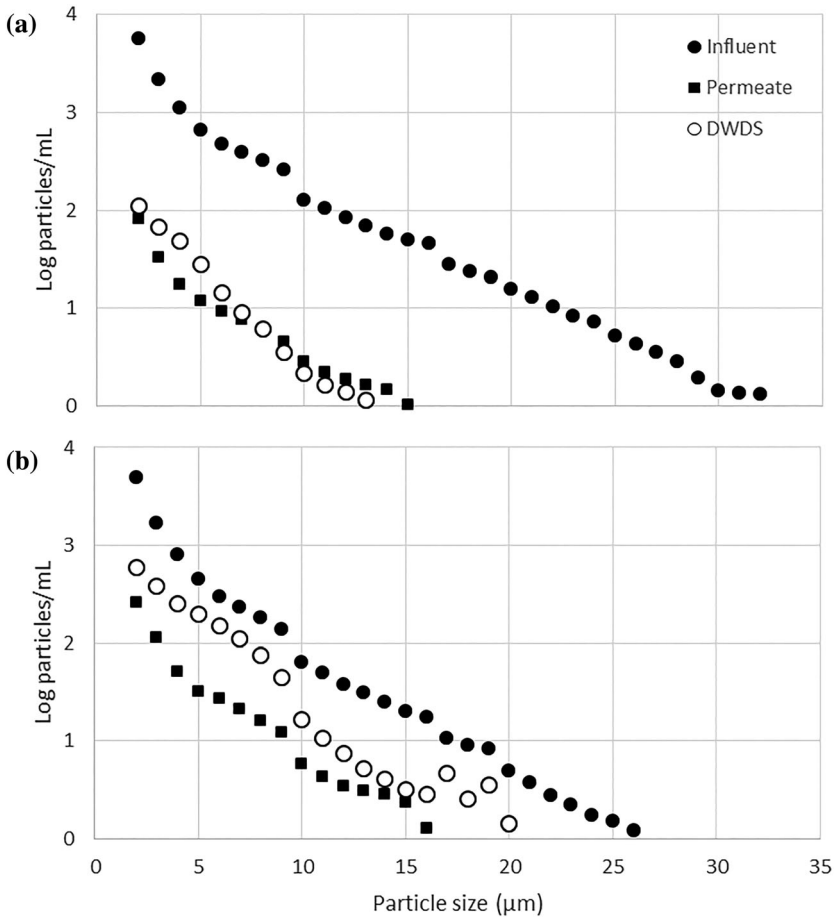

FIGURE 2 Influent (●), permeate (■), and DWDS (○) mean PSD (2–125 μm), during UF stage (a) and O_3-UF stage (b).

TABLE 4 Average, minimum, and maximum coefficients A and b of PSD β-variable model for each water stream in the experimental stages.

		Influent		Permeate		DWDS	
		A	b	A	b	A	b
UF ($n = 15$)	Mean	3856.7^{a}	2.1^{a}	26^{b}	1.5^{c}	59.5^{b}	1.7^{b}
	S.D.	3067.0	0.1	16	0.2	43.5	0.2
	Min.	952.5	1.9	6	1.2	10.0	1.4
	Max.	10298.0	2.3	58	1.8	148.1	2.2
O_3-UF ($n = 24$)	Mean	2635.9^{a}	2.1^{a}	56.9^{b}	1.6^{c}	205.4^{b}	1.9^{b}
	S.D.	2219.1	0.2	75.1	0.2	211.8	0.3
	Min.	150.4	1.9	11.1	1.4	23.7	1.5
	Max.	8149.4	2.4	273.2	2.0	834.9	2.7

Note: A different letter (a, b, c) indicates statistically significant differences (SNK 0.05) between groups: influent, permeate, and DWDS.

Due to the sieving capacity of UF membranes, aided by the cake formed during the ultrafiltration process, a high removal capacity of particles larger than 2 μm can be expected. This particle retention capacity is more noticeable for large particles. However, contrary to the conclusions of Vreeburg et al. (2008), ultra-filtered water cannot be considered particle-free (Lousada-Ferreira et al., 2016; Rojas et al., 2008). This fact was confirmed in the present study, where particles larger than the membrane pore size were observed in the permeate during both stages.

The origin of the particles present in permeates can be highly diverse. Membrane pore size should be perceived as a pore size distribution fraction with a wide variety of sizes, including some abnormally large or defective pores (Lousada-Ferreira et al., 2016). Permeate quality from UF membranes can also be compromised due to membrane failure (Guo et al., 2010). In addition, wear and tear of membrane materials can alike contribute to the presence of particles in the permeate.

According to parameter "A" of the β-variable model, a significant removal of particles was achieved with similar statistical significance for both stages (Table 4). However, the evolution of parameter "A" in the permeate during the UF stage was more homogeneous than during the O_3-UF stage (Figure 3). The concentration of particles in the permeate was independent of its concentration in the influent for both stages (Pearson's coefficient 0.256 and −0.091 for stages UF and O_3-UF, respectively). Therefore, a better control of particle concentration in the permeate was obtained with single-treatment UF than with pre-ozonation.

The values of parameter "b" decreased between influent and permeate for both stages with significant statistical differences (Table 4) but without correlation (Pearson's coefficient 0.118 and −0.08 for stages UF and O_3-UF, respectively). Similar "b" values over time were observed for both stages, but with slightly lower values for permeate obtained during the UF stage than for the O_3-UF stage (Figure 4). The maximum particle size detected in the permeates of the two stages was the same (Table 3), but a lower presence of particles around 2 μm in size was obtained with single-treatment UF. These results explain the lower values of "b" and higher permeate quality. Pre-ozonation enables particles to pass more easily through the membrane (Szymanska et al., 2014), which explains the better quality of the permeate obtained with single-treatment UF. Since the UF membrane is a physical barrier to particulate material, the application of ozone causes an increase in the concentration of particles in the permeate, but without affecting their maximum size.

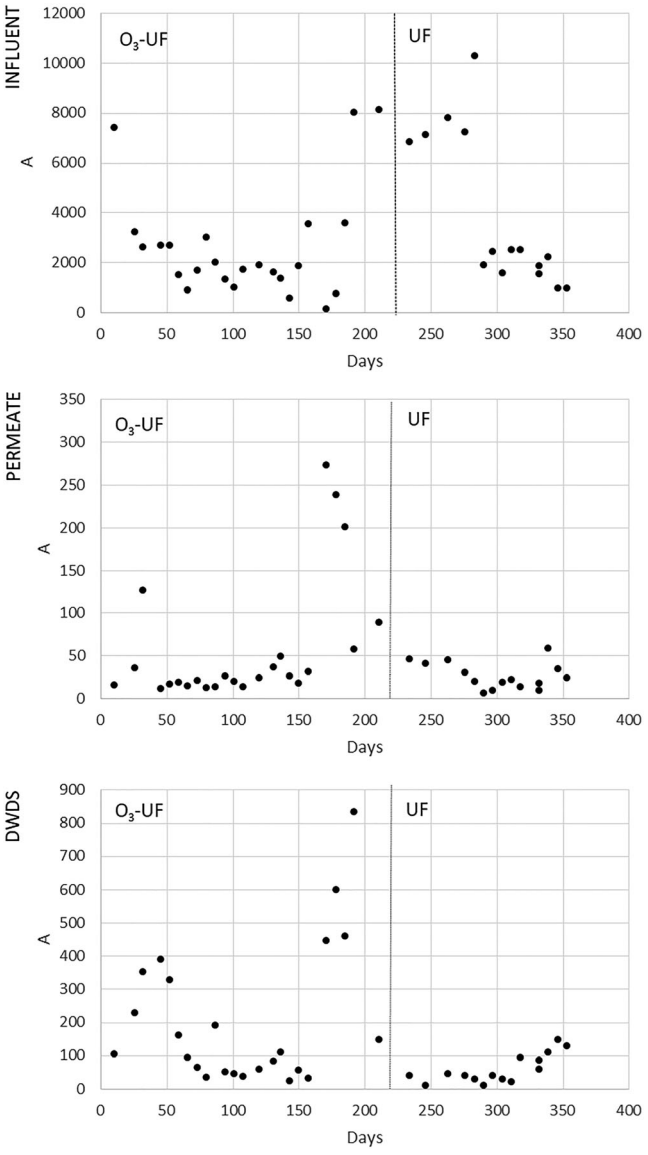

FIGURE 3 Parameter "A" time course of influent, permeate, and DWDS water during UF and O_3-UF stages.

PSD analysis of the water samples from the DWDS showed a worsening of water quality, with a clear increase in the concentration of particles, although without statistically significant differences (Table 3). According to logarithmic regression (Figure 2), during the O_3-UF stage, a clear increase of particle concentration was observed for each of the measured particle sizes. However, during the UF stage, this increase was observed only for the smaller particle sizes. Particles over 21 μm were not observed during the UF stage, whereas during the O_3-UF stage, particles of up to 35 μm were observed.

Particles are deposited in the DWDS as a function of the water velocity, with the concentration decreasing at lower water velocity. In this regard, Byrne et al. (2014) observed that the presence of particles in DWDS with a

water velocity between 0.0141 and 0.0031 m/s depends on the input of particles into the system. However, in the present study, the concentration of particles in the DWDS increased with respect to the permeate during both stages. A constant velocity of 0.9 m/s was established during our experiments, which limited particle deposition. Moreover, due to the service age of the pipe (130–220 days) and the use of polyethylene, which does not produce corrosion, the increase of the number of particles could not come from the wear and tear of the pipe.

Values of both β-variable model constants experienced a generalized increase in the DWDS with respect to permeate during both stages, with statistically significant differences for parameter "b" (Table 4). The time course of parameter "A" (Figure 3) during O_3-UF was heterogeneous and correlated with the values of the permeate (Pearson's coefficient 0.605). In contrast, a clear tendency to increase over time was observed during the UF stage without correlation with the values of the permeate (Pearson's coefficient 0.221). This shows a greater dependence of the particle concentration in the DWDS on those supplied by the permeate during the O_3-UF stage than during the UF stage. Unlike the values observed for influent and permeate, the values of parameter "b" in the water samples from the DWDS were also heterogeneous during both stages (Figure 4). Similarly, no statistical correlation was found for parameter "b" between the permeate and DWDS values (Pearson's coefficient −0.020 and 0.104 for stages UF and O_3-UF, respectively).

During the UF stage, the particles were contributed to the DWDS in the lowest number, and due to the water velocity, they remained re-suspended. As a result, in the first days of the UF stage, the values of parameter "A" in the permeate and DWDS were similar. However, over time, an increase in the DWDS of the concentration of particles of around 2 μm in size was observed. This phenomenon caused an increase not only in the values of parameter "A" without statistical correlation with values of the permeate but also in the values of parameter "b," by affecting the curvature of the PSD. During this stage, the average maximum size of particles detected in the DWDS decreased with respect to the size observed in the permeate.

By contrast, during the O_3-UF stage, the contribution of particles to the DWDS was higher and showed a different evolution in the DWDS. In this stage, the concentration of particles increased for all registered sizes, although more significantly for lower sizes. This particle behavior caused a higher increase in the values of parameters "A" and "b," with a very heterogeneous evolution over time (Figures 3 and 4). During this stage, the

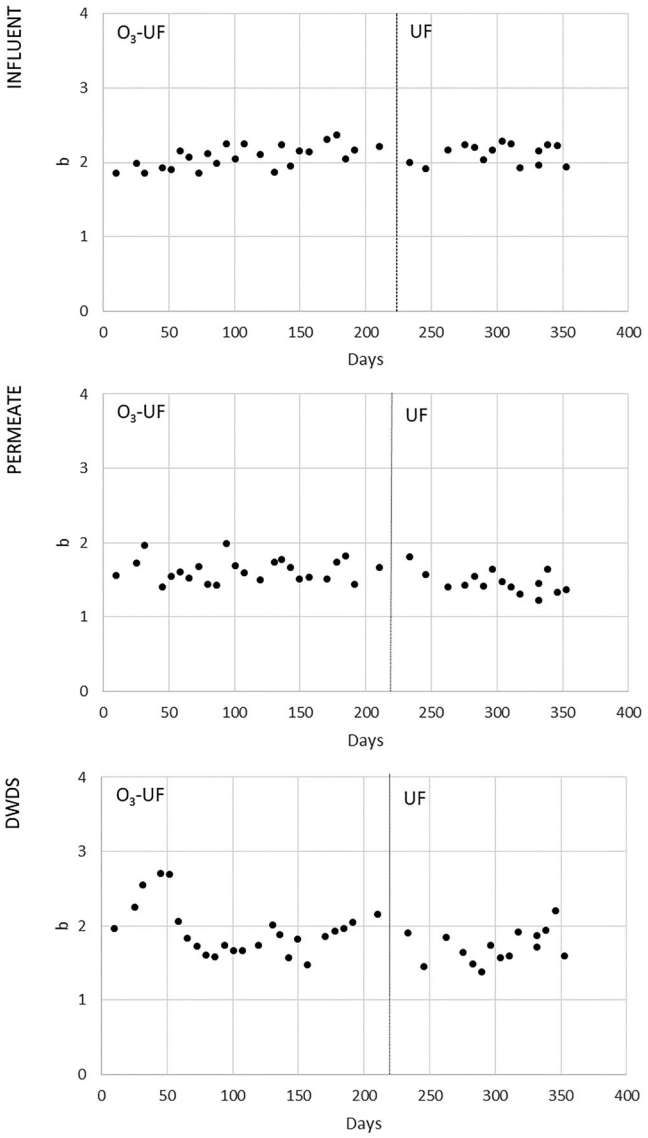

FIGURE 4 Parameter "b" time course of influent, permeate, and DWDS water during UF and O_3-UF stages.

average maximum size of particles detected in the DWDS increased with respect to the size observed in permeate.

The presence of particles in the DWDS was influenced by the contribution of the permeate, especially during the O_3-UF stage. However, bearing in mind that the water velocity in the DWDS, particle deposition, aggregation, and re-suspension do not seem to be the most likely cause of the deterioration of water quality, but rather the development of biofilms. Certainly, biofilm developed in the DWDS during both stages as described in a simultaneous study carried out with the same experimental installation and operational conditions (Álvarez-Arroyo et al., 2022).

A proportion of the particles that pass through the membranes are organic matter, which can favor the development of biofilm during both stages (Liu

et al., 2013). Furthermore, due to the fragmentation of organic compounds by ozone oxidation, the biodegradability of organic matter reaching the DWDS was higher during the O_3-UF stage (de Vera et al., 2016; Szymanska et al., 2014). Accordingly, a higher increase in the concentration of different sizes of particles in the DWDS can be expected during the O_3-UF stage due to the detachment of the biofilm developed on the wall of the pipe, dragged by the water flow. However, further research is needed to confirm the potential for more biofilm development in DWDS when ozone is applied prior to UF membranes during water potabilization.

CONCLUSIONS

An experimental DWDS was used to evaluate the evolution of drinking water quality of ultrafiltered water with or without pre-ozonation. The application of ozone prior to UF membranes resulted in a better control of membrane fouling, and the UF membranes were effective in water turbidity removal with no significant increase in the yield due to the pre-ozonation. The analyzed processes were not able to correct DOC concentration, showing only a certain capacity for the removal of hydrophobic aromatic compounds by the O_3-UF process. The following conclusions can be obtained from the study:

- Under our test conditions, it was not possible to observe quality differences between the permeates obtained with or without pre-ozonation, or between the permeates and the water quality in the DWDS, by means of turbidity, DOC or UV_{254}.
- PSD analysis of the permeate and its fit to the β-variable model demonstrated the higher efficiency of UF membranes at removing smaller particles (around 2 μm) compared with the system with pre-ozonation.
- PSD analysis of the water samples from DWDS showed a worsening of water quality, which was more pronounced when pre-ozonation was used.
- With pre-ozonation, a significant increase of particle concentration in the DWDS was observed for each of the measured particle sizes, particularly for lower sizes, while during the UF stage, only particles smaller than 5 μm increased in number.

AUTHOR CONTRIBUTIONS

Rocío Álvarez-Arroyo: Conceptualization; methodology; writing - original draft; investigation; formal analysis; data curation. **Jorge Ignacio Pérez:** Methodology; investigation; formal analysis. **Luz Marina Ruiz:** Methodology; investigation; formal analysis. **Miguel Ángel Gómez:** Conceptualization; methodology; investigation; formal analysis; supervision; funding acquisition; project administration; writing - review and editing.

ACKNOWLEDGMENTS

This study was funded by the Spanish Ministry of Science and Innovation (CTM2010-18899-TECNO). It was conducted at the Department of Civil Engineering and Institute of Water Research of the University of Granada and the Parque de las Ciencias (Science Park) of Granada (Spain).

CONFLICT OF INTEREST STATEMENT

The authors declare that there are no competing interests.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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How to cite this article: Álvarez-Arroyo, R., Pérez, J. I., Ruiz, L. M., & Gómez, M. Á. (2023). Analysis of the evolution of ultra-filtered water quality in a drinking water distribution system by particle size distribution: Influence of pre-ozonation. *Water Environment Research*, *95*(2), e10840. https://doi.org/10.1002/wer.10840