Integrating membrane technologies and blending options in water production and distribution systems to improve organoleptic properties. The case of Consorci d'Aigües de Tarragona.

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Abstract
The drinking water supply in the Tarragona area has recently evaluated the integration of membrane desalination technology to improve the organoleptic water quality of the distributed water potabilized from the Ebro River. The aim of the study was to predict the organoleptic water improvement and the potential reaction of consumers when blends between membrane desalinated and conventionally treated source were supplied. This paper presents the water quality improvement in terms of both chemical and organoleptic properties. Regarding the relationship between the salinity represented by the total dissolved solids (TDS) and water preference, the overall trend indicated that water with lower salinity was preferred. These results confirm that the assessment of water is primarily driven by TDS even though other factors (e.g. pH, mineral composition) can play a significant role in the liking of water, which is consistent with previously published studies.

Keywords
Membrane technologies, drinking, organoleptic quality, desalination, blends, reverse osmosis.

INTRODUCTION

In recent decades, the Tarragona Metropolitan Area (TMA) drinking water supply network has been primarily based on surface water resources from the Ebro River. The drinking water treatment plant of the Consorci d' Aigües de Tarragona (CAT) is located in L'Ampolla (Spain) and supplies drinking water in the province of Tarragona (355.000 m^3/d) (Ruana et al., 2013). The raw water intake is located at the low basin of Ebro river, which has moved from sporadically high values in salinity to continuous episodes of high content of sulfate ions. These resources are suffering the effects of agricultural and industrial discharges, as well as a reduction in quantity, decreasing the quality of the raw river water (Valencia, 2007). Additionally, due to the Mediterranean climate, the natural water resource availability is periodically lower than the water demand in the area. In this direction, a semi-industrial pilot plant was used to quantify the quality improvement by means of reverse osmosis (RO) and thus, validate the potential implementation of a large-scale plant, with a capacity of 250.000 m^3/d. The main objective of these tests were to maximize the salt rejection and the water recovery as well as to minimize the costs for the full scale pilot plant with the integration of those membrane technologies within the current conventional treatment scheme. Ultrafiltration (UF) and microfiltration (MF) as pretreatment steps of RO and nanofiltration (NF) have been validated.

High and medium pressure membrane techniques (NF, RO) can greatly reduce the minerals of water and alter the salt content (Yeh et al., 2000; Greenlee et al., 2009). Therefore, the product requires remineralisation to mitigate the aggressiveness of the product, which typically involves the addition of calcium and bicarbonate salts and sometimes carbon dioxide. In addition, this remineralisation process provides two advantages. First, the remineralisation contributes to improve the water flavour (Whelton et al., 2007;). Second, the remineralised water is healthier for the human body because research suggests that alkaline-earth salts protect against disfunctions of the heart and vascular system (Withers, 2005). Water taste strongly depends on its mineral composition. For example, very high or very low concentrations of total dissolved solids (TDS) have been demonstrated to affect water taste (Teillet et al., 2010). Also some cations, anions (Whelton et al., 2007), the residual chlorine (from the disinfection process) and the evolution of water in the distribution system can significantly affect the taste (Dietrich, 2006). Other minor compounds can also produce undesirable effects such as unpleasant odour and taste for consumers.

Generally, water tasting with trained panelists or volunteers is preferred in laboratory experiments over other instrumental techniques such as the electronic tongue. Despite the fact that panel sensory evaluations are subjective and that the experiments required are time-consuming and involve high costs, they provide a deeper understanding of the consumer preferences (Devesa et al., 2007). The experiments performed with untrained volunteers usually lead to conclusions which are very close to the profile and expectations of real consumers. On the other side, instrumental techniques can only be used for quality control. In this sense, it has been shown that human senses are sometimes more sensitive than the available analytical equipment (Devesa et al., 2004). The objective of this work was to characterize the organoleptic improvement of the membrane treated water when blended with water following the traditional treatment line from L'Ampolla drinking water treatment plant (DWTP), including sedimentation-coagulation, ozonization and activated carbon filtration. The final aim was directed to provide recommendations and limits for the blending options.

MATERIALS AND METHODS

Drinking water pilot plant (ELSA) treatment description.

L'Ampolla DWTP treats water from the Ebro River. At that point, the river has received effluents from industrial and urban wastewater treatment plants (WWTPs), as well as pollution from agricultural and industrial activities. The high anthropogenic influence on the water quality made it necessary to add an ozone stage to remove several organic and inorganic compounds that cannot be efficiently removed by the more common treatments, such as flocculation or activated carbon. Therefore, currently the Ebro DWTP treatment scheme (referred as conventional treatment) is composed of a pre-oxidation (permanganate) – coagulation /flocculation/settling (ferric chloride) – sand filtration – activated carbon and final chlorination, presenting a maximum treatment capacity of 4 m^3/s (Ruana et al., 2013).

An improved treatment line has been evaluated using a pilot plant fully automated. The evaluated process used membrane technology to treat a percentage of the total flow (up to a 50%) from the conventional treatment from L'Ampolla DWTP with a pre-treatment via micro-coagulation and UF or MF as protection for the subsequent RO step. Product water from the conventional treatment fed the pilot plant (12 m^3/h) where pH was corrected to pH 7 dosing CO_2(g)/L. From this point, the water was pumped and treated by means of a MF or UF system (pressurised; outside-in; dead-end). Permeate water was stored and fed by a high pressure pump to the RO racks. Doses of 1 mg/L sodium bisulphite and 2 mg/L of antiscalant were added to assure the membrane operation requirements. A configuration with 3 stages with 5, 2 and 1 pressure vessels was used. Between the second and third stage, a booster pump was used to assure the hydraulic requirements of the stages. The pilot plant incorporated a water recovery system from the generated brine (2 m^3/h) including a lamella settler equipped with flocculation followed by another UF system to remove the particulate matter escaping from it with a water production of 40 L/(m^2·h). The water was send to a new RO or NF stage after dosing sodium bisulphite (1 mg/L) and antiscalant (7 mg/L). The new permeate obtained was mixed with the previous RO step

Water samples for organoleptic characterization

Samples from the conventional treatment train of L'Ampolla DWTP and from the RO pilot plant were collected. Two blends were prepared at the Aigües de Barcelona laboratory, at 50/50 % and 70/30 % (RO water/conventional treatment water) ratios. In addition, two tap water samples from the metropolitan area of Barcelona were also considered in both experiments. These waters have traditionally been taken as reference in several previous studies (García et al, 2015). Cardedeu DWTP treats good quality surface water (from Ter River) by means of a conventional treatment, with a TDS about 300 mg/L; Sant Joan Despí DWTP water is obtained from a high salinity surface (Llobregat River) and a groundwater resources, of about 1 g/L of TDS.

Panelists and tasting sessions

Sensory experiments were carried out by an expert panel (14 subjects, 5-7 per session) from Aigües de Barcelona, trained according to the Flavour Profile Analysis (FPA) method (AWWA, 2010). The sensory tests used in this study were not of a descriptive nature (the taste-and-odour wheel is not used) and, therefore, required a lower degree of training. This panel was used due to availability reasons. On the other hand, its evaluation allowed the response of the most sensitive consumers to be extrapolated. The tasting took place in a room specifically intended for this purpose, comfortable and free from interfering odours. The encoded samples were always presented without giving the panelists any information which could influence their appreciation. Flavour test was carried out in glasses on samples thermostatized at 25º C. The sensory technique used (Meilgaard et al., 1991) was the rating, scoring and triangle tests. The subjects were asked to rate the overall liking of samples on a 0–10 scale.

Sensory test methods. Both affective (scoring and ranking test) and difference (triangle test) techniques were used (Morran et al., 2004). For the ranking test, a series of n different samples were presented and the tasters had to order them in accordance with a certain characteristic, in this case their overall assessment of the flavour. The sample perceived as the best received n points, n-1 for the following, etc., until the last one which is granted 1 point. For the scoring test, the tasters were asked to give a score, using a scale from 1 (extremely bad) to 10 (excellent) units. For the triangle test, groups of three samples were presented, two of which were identical and one was different. Tasters were told to identify the odd sample. The comparison of the number of correct responses with a critical number allowed to deduce if the difference between the samples studied could be considered as statistically significant or not, at a certain confidence level (α =0.05).

Data analysis. Analysis of variance was used to assess whether the observed differences in the scoring test (average liking) were statistically significant. If the F-test exceeded the critical value (confidence level 0.05) for the corresponding degrees of freedom, then the null hypothesis assumption of the equivalent mean rating among the rated waters was rejected. A multiple comparison procedure (Fischer's Least Significant Difference (LSD)) was used to determine which of the means were significantly different. The data from the ranking test was analysed using Friedman-type statistics in a non-parametric test. If the data was significant, then the LSD, which is the non-parametric analogue to Fisher's LSD for the rank sum was calculated (Meilgaard et al., 1991). Any two samples whose rank sum differed by more than the LSD rank were declared significantly different at the confidence level considered.

RESULTS AND DISCUSSION

Characterisation of the organoleptic properties of treated surface water sources: relationship between physico-chemical properties and taste

Table 1 lists the physico-chemical properties of conventionally treated Ebro River water at L'Ampolla DWTP and the two reference water from Barcelona Metropolitan Area (Ter and Llobregat Rivers). The Ebro River source is transferred through a 60 km pipe. Its composition is revealing an increasing anthropogenic impact in the last decade as this lower part of the Ebro basin is receiving a salt run-off from agriculture areas at the upper part of the basin, as well as industrial and domestic discharges additionally to other diffuse pollution problems (Fernandez-Turiel et al., 2000). The composition of the conventionally treated water showed low to medium concentrations of bicarbonate (182±11 mg/L) and an electrical conductivity of 988±10 (µS/cm), for a total dissolved solids of 734±11 mg/L. Values of the ions presenting an impact on the water taste are chloride (145 ±10 mg/L), sodium (95±7 mg/L), calcium (102±8 mg/L) and magnesium (24±3 mg/L). The Ter River water, after being treated in Cardedeu DWTP presented lower concentrations of cations and anions, particularly sodium and chloride, and much lower conductivity values (396±35 mg/L) than the surface water from the Llobregat River (1672±201 µS/cm) conventionally treated in Sant Joan Despí DWTP.

Table 1. Mean value of physico-chemical properties of water after conventional treatment (n=4). Standard deviation in brackets

	DWTP1 (Ebro River)	DWTP2(Ter River)	DWTP3 (Llobregat River)
Hardness (mg$CaCO_3$/L)	350 (12)	165 (10)	452 (28)
Calcium (mg/L)	102 (8)	51 (3)	120 (8)
Magnesium (mg/L)	24 (3)	9 (1)	37 (2)
Sodium (mg/L)	95 (7)	16 (1)	175 (37)
Potassium (mg/L)	2,7 (0,3)	2,8 (0,2)	26,0 (3,0)
Bromide (mg/L)	< 0,1 (loq[1])	< 0,1 (loq)	0,7 (0,1)
Sulphate (mg/L)	205 (20)	45 (1)	199 (10)
Chloride (mg/L)	145 (10)	27 (1)	301 (67)
Conductivity (µS/cm) (20ºC)	988 (34)	396 (35)	1672 (201)

[1] loq: limit of quantification

Ranking and triangular tests results for the three water types, collected in Table 2, indicated that the panelists preferred water from the Ter River, assigning an average score of 7,06±0,3, while the water from the Ebro and Llobregat Rivers received average scores of 3,6±0,9 and 2,8±0,4, respectively. Then, clear different organoleptic properties were perceived between the three facilities. The differences agreed with previous studies (Devesa et al., 2010; Raich-Montiu et al., 2014) using water samples obtained directly from the DWTP where the water origin was well defined in comparison with samples from the distribution network, when blending options were applied as it was the case of the Barcelona Metropolitan Area. The three ranking results and also the scorings were found to be significantly different by multiple comparison procedure (LSD). The capacity of discrimination between waters L'Ampolla DWTP and reference Llobregat water was evaluated on a joined panel of 26 trained tasters and volunteers, 14 and 12 respectively. The number of correct answers was 16 and, therefore, the two samples were considered different (confidence level: 5%).

Table 2. Results of ranking and scoring tests of samples of produced water from the L'Ampolla DWTP compared to two references.

Water	Ranking	Scoring
Reference A (Ter River DWTP2)	2.8	7.0
L'Ampolla (Ebro River DWTP1)	1.8	5.6
Reference B (Llobregat River DWTP3)	1.3	3.6

Influence of membrane treatments (UF-RO) and water source on the organoleptic properties.
The results presented in Table 3 showed that conventionally treated water (sample 1) was given a lower score than that treated with RO membrane technology (samples 1 and 2), even when they were from the same source. Tasters penalized the source of sample 5, which contained the highest salinity. These results agreed with previously published studies, in which taste analysis seemed to indicate a worse organoleptic perception for RO desalinated seawater in front of RO desalinated surface water (Devesa et al., 2010, Garcia et al., 2015). Results of ranking tests and scoring test are presented in Table 2 and Figure 1, and Table 3 and Figure 2, respectively. The LSD grouping leaded to slight different structures.

Table 3. Ranking and scoring tests (waters with the same letter (a-d) were not significantly different). Blend refers to a mixture between the conventional treatment (*) from Ebro River DWTP (L'Ampolla) (coagulation-sedimentation-ozonization-activated carbon filtration) and the UF/RO treatment scheme assessed at pilot plant level with the reported ratios; Ter River DWTP and Llobregat River DWTP refer to water conventionally treated by Cardedeu and Sant Joan Despí DWTP, respectively.

S#	Water	Rank	LSD groups	Score	LSD groups
1	Blend 50% (Conv)/50% (UF/RO)		a	8.1	a
2	Blend 50% (Conv)/50% (UF/RO)	4.1	a	7.9	a b
3	Reference A (Ter River DWTP)	3.4	b	7.0	b
4	Conventional* (Ebro River DWTP)	2.2	c	4.5	c
5	Reference B (Llobregat DWTP)	1.0	d	1.4	d

Triangular tests to find out if the blend 50/50 could be differentiated from the conventional water and, also if the two blends (50/50 and 70/30) between them would be discriminated were performed using a joined panel of trained and untrained tasters. The results, presented in Table 4, indicated that the blend 50/50 was differentiated and outperformed the other samples, forecasting a better organoleptic perception among consumers. When blended samples 50/50 and 70/30 were compared, no differences were found and thus, the increase of the ratio of blending would not be justified from the organoleptic point of view of improving water quality perception.

Table 4. Results of the triangular tests between conventional water from L'Ampolla DWTP and blends with water produced by the RO-UF pilot plant.

Water	Panellists	Correct answers	Significant difference?
Conventional* vs Blend 50/50	40	27	Yes
Blend 50/50 vs Blend 70/30	36	11	No

Optimization of organoleptic taste: production recommendations
Figure 1 shows the scores of the different blends and unblended water samples compared to TDS. The overall trend was that water with increased salinity was less desirable. This result is consistent the effect of TDS on the water assessment (Devesa et al., 2004; Fabrellas et al., 2004). However, other factors can play a significant role in liking water (e.g. pH, mineral composition), which would explain the behavior of each water sample (Devesa et al., 2010; Glueckstern et al., 2005). As can be seen, Llobregat River sample, scored as 1.4 with the present panel, had been evaluated in previous studies with values between 3.0 and 3.4.
Table 5 summarizes the data of the organoleptic characterization of blended samples from four DWTPs using surface waters (Ebro River, Llobregat, Abrera and Cardedeu) and from a DWTP using

seawater. With exception of the Ebro River and Cardedeu DWTP, where water is produced with conventional treatments not involving membrane processes, the other three plants include the use of RO and electrodialysis reversal. It is also plotted samples produced by blending some of the previously mentioned water resources, either sampled at the treatment plant itself or prepared at the laboratory mixing, at given rations, the original source and the permeate produced through the membrane step. As it can be seen, blending of water turned into an enhancement of the samples' organoleptic perception.

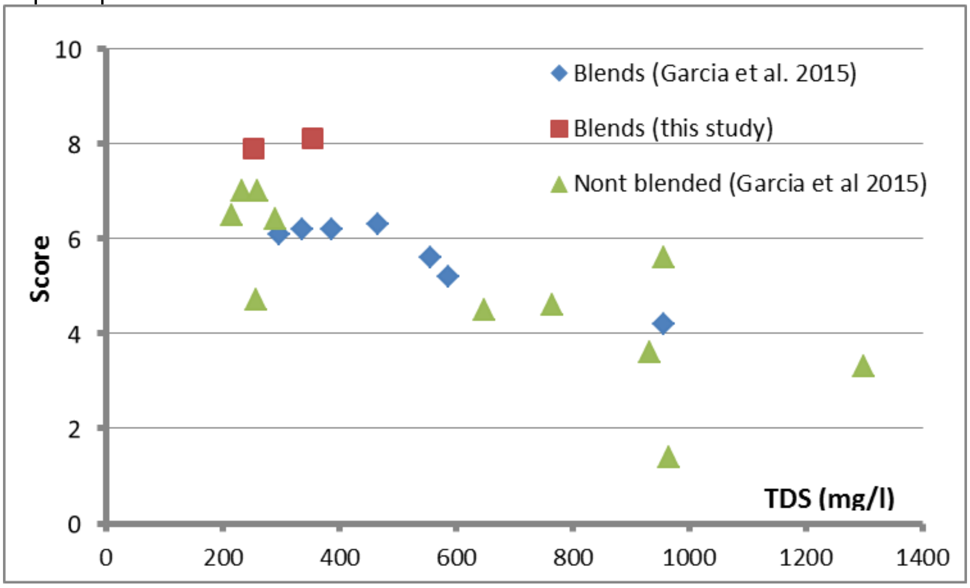

Figure 1. Variation in the scores as a function of TDS content for the water samples analysed within this study, as well as data from literature (Garcia et al., 2015).

Table 5. TDS, scores and LSD grouping from different water samples (blended and non-blended; from this work and from literature). Waters with the same latter (a to d) are not significantly different.

Membrane Technology	Sample	TDS (mg/L)	Score	LSD groups	Ref
EDR[1]	Abrera DWTP	383	6.5	a	Garcia et al., 2015
None[2]	Cardedeu DWTP	302	6.4	a	Garcia et al., 2015
S-RO[3]/EDR	Blend 4	445	6.3	a	Garcia et al., 2015
RO/EDR	Blend 2	352	6.2	a b	Garcia et al., 2015
RO/EDR	Blend 3	305	6.2	a b	Garcia et al., 2015
RO/EDR	Blend 5	257	6.1	a b	Garcia et al., 2015
RO/EDR	Blend 0	522	5.6	a b	Garcia et al., 2015
RO/EDR	Blend 6	524	5.2	b c	Garcia et al., 2015
SWRO[4]	El Prat seawater plant	243	4.7	c d	Garcia et al., 2015
None	Llobregat Conventional	1077	4.6	d	Garcia et al., 2015
RO/EDR	Blend 1	884	4.2	d	Garcia et al., 2015
None	Llob-Conventional	1163	3.3	d	Garcia et al., 2015
RO/UF[5]	Ebro DWTP Blend 50/50	353	8.1	e	This study
RO/UF	Ebro DWTP Blend 30/70	254	7.9	a	This study
None	Ebro Conventional*	956	5.6	a	This study

*conventional: refers to water produced at the Ebro River DWTP (L'Ampolla) using the existing treatment (coagulation-sedimentation-ozonization-activated carbon filtration, [1] EDR refers to Electrodialysis reversal, [2] SRO refers to a surface river water treated by RO, [3] SW-RO refers to seawater desalinated using RO, [5] RO/UF: refers to Ebro River water treated with UF and RO . .

Figure 1 shows that the general tendency was the improvement of the organoleptic water quality with blends prepared using water obtained both by RO and electrodialysis. In the case of the samples of the Ebro River, the scores were slightly higher than those obtained with samples from the Ter River, initially presenting (non-blended samples) higher score.

CONCLUSIONS

This study enabled the collection of valuable information regarding the influence of new membrane treatment technologies using UF and RO in the drinking water treatment scheme. In particular, the organoleptic water characteristics of water produced by this scheme were compared to the current conventional treatment from L'Ampolla DWTP (coagulation-sedimentation-ozonization-activated carbon filtration), as well as various blends produced with both treatment lines.. In this scenario, the effectiveness of mixing water treated by UF/RO membranes with water undergoing a filtration step through granular activated carbon before leaving the DWTP to improve the organoleptic properties of water was clearly demonstrated. Two blends of water from membrane treated and conventional sources, as well as three unblended water samples, from three different surface origins, were assessed using ranking, scoring and triangular tests. Several conclusions can be drawn from the results of the tasting with trained and untrained panelists.

The ranking and scoring results of both panels were similar. These results indicated that the flavour of the blends was significantly better than the water previously distributed in the Tarragona area. Therefore, membrane treatment with UF-RO would positively contribute to consumer perception. This improvement was quite considerable for both blends. The detection in the improvement is favorable due to the high percentage (75 %) of Ebro water that the blends contained. In agreement with these results, a multiple comparison test performed on the normalized scores of the aggregated results of both panels resulted in defining a grouping structure of water samples that were significantly different from the others. With respect to the relationship between TDS and liking, the overall trend observed was that the taste of water was less appealing when the salinity increased. These results confirmed that the water assessment is primarily determined by TDS even though other factors (pH and mineral composition) can play a significant role in how water tastes. More research is needed on this subject.

Triangle test results for both panels (trained and volunteers) indicated that all the blends could be differentiated from the Llobregat water (produced by the conventional treatment). This test was also used to compare the sensitivity of the two panels. These results suggest that the number of tap water consumers may increase in the Tarragona Metropolitan Area with the implementation of UF/RO stages. However, in the future, further studies regarding the relationship of the physico-chemical properties of water and its evolution along the distribution network, as well as on the blending of water from the seawater desalination treatment plant, should be undertaken in the context of water taste preferences.

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