Assessment of membrane processes for taste and odour removal

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Abstract The occurrence of tastes and odours (T&O) in potable water is considered one of the main problems by the drinking water companies. Thus, several treatment processes have been developed over the years to control T&O, including air stripping, activated carbon and oxidation using ozone. However, little information is available in the literature on the use of membranes for T&O removal. Therefore, the objective of this paper is to present potential of membrane processes for the removal of taste and odour-causing compounds. Several membranes were tested including ultrafiltration (UF), UF combined with powdered activated carbon (PAC), nanofiltration (NF) and low pressure reverse osmosis (LP RO) membranes. Combination of UF with PAC was found to be effective for T&O control. The use of NF or LP RO is still unclear in the objective of T&O control.

Keywords Membrane; powdered activated carbon; reverse osmosis; taste and odour; ultrafiltration

Introduction

Tastes and odours (T&O) in drinking waters represent one of the major causes of consumer complaints, together with water hardness and water turbidity. Different surveys carried out in various French regional centres of the Lyonnaise des Eaux indicate that T&O episodes induce from 30 to 50% of recorded complaints.

The European Drinking Water Directive stipulates a maximum T&O threshold (dilution ratio with a reference water beyond which no odour or taste is perceived) equal to 3. The recent World Health Organization (WHO) recommendations indicate that drinking waters must not present unpleasant tastes but do not include any guideline value. The above mentioned T&O guideline value will probably disappear from the revised drinking water directive. However, beyond these legal considerations, off-flavour episodes strongly contribute to the declining public image of water companies and lead to a worrying paradox: while the price of drinking water is gradually increasing under the influence of the important investment made necessary to comply with the more stringent water quality guidelines, consumption of bottled water by the general public follows the same increasing pattern. In other words, consumers now tend to believe that if their water tastes or smells bad, then it is probably not safe.

Due to their high complexity, taste and odour phenomena continue to challenge both analytical chemists and water treatment engineers. One of the difficulties stems from the fact that T&O are often due to infinitesimal traces of organic compounds such as geosmin or methylisoborneol, the two well known algal metabolites which have odour thresholds concentrations of 4 and 9 ng/L, respectively (Mallevialle and Suffet, 1987) or specific halogenoanisoles such as 2,4,6-trichloro or tribromoanisole with their odour thresholds in the 20–80 pg/L range (Crane *et al.*, 1996). Targeting such low levels of specific compounds in a complex mixture which contains mg/L levels of natural organic matter and hundreds of mg/L of inorganic compounds which may themselves act as precursors for odourous oxidation by-products will remain a challenge. However, enough experience has been gained during the last 15 years to allow water treatment professionals to select the most appropriate treatment processes or combinations of processes in order to solve or minimise T&O problems.

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The objective of the paper was to evaluate the performances of various membrane technologies for control of T&O episodes. The rejection of specific compounds such as trichloroanisole was evaluated with the use of the recently developed Large Volume Gas Chromatography method.

Materials and method

Test conditions: Tests were performed on several ground- and surface waters in France and also in California, USA. Several membrane processes were evaluated including ultrafiltration (Cutoff of 100,000 daltons), UF combined with powdered activated carbon (PAC), tight cutoff UF (500 daltons), nanofiltration (Cutoff of 200 daltons), low pressure reverse osmosis, and reverse osmosis. Trichloroanisole (TCA) and trichlorophenole (TCP) spiking were performed prior NF and RO membranes.

Analysis Methods: The threshold odor and taste number (TON and TTN) consists in diluting several aliquots of the sample which had to be evaluated to varying degrees with odor free water (standard methods 1985)[1]. The highest dilution at which an odor or a taste can be perceived is an indirect expression of that sample's threshold odor or taste concentration (TOC or TTC), and the dilution is reported as a "threshold odor or taste number".

The Closed Loop Stripping Analysis (CLSA)[1] was employed to extract and concentrate "earthy-musty" volatile compounds such as 2,4,6-TCA present in the water. To concentrate 2,4,6-trichlorophenol (2,4,6-TCP), Liquid/Liquid extraction was employed. Both concentrates were analyzed by Large Volume Gas Chromatography (LVGC-MS; ULTRATRACE).

Results and discussion

The efficacy of UF, NF and UF combined with PAC (WPH PAC, Calgon Carbon Corp.) for odour removal was evaluated on Delta water, California. Selected results are presented in Figure 1 (Jacangelo *et al.*, 1994). The results showed that UF, as expected, was not efficient in removing T&O causing compounds; no significant differences were observed even for low cutoff UF membrane (500 daltons), suggesting that the molecular weights of some compounds producing the musty odour were less than 500 daltons. However, the use of PAC combined with UF (CRISTAL process, Baudin and Anselme, 1995) was found to be effective in controlling T&O. The TON was reduced from 9 to 2.

In come cases, the CRISTAL process was found to be more efficient than the conventional ozone/granular activated carbon filtration process. Today, 13 full-scale plants, with

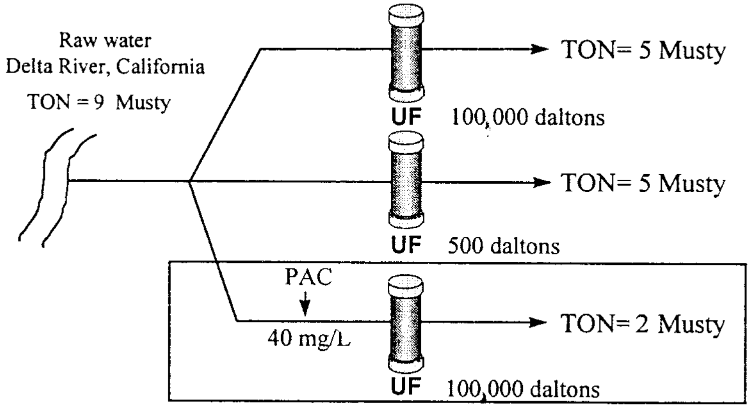

Figure 1 Impact of membrane cutoff on odour removal

Table 1 Aquasource UF full-scale plants using the CRISTAL process

Plants and on-line date	Capacity	Source water	Average PAC dose	Water quality objective
Châtel-Gérard, France, 1993	660 m^3/d	groundwater	6–8 mg/L	colour
Chermisey, France, 1994	200 m^3/d	groundwater	10 mg/L	TOC
Apié, France, 1996	28,000 m^3/d	reservoir water	8 mg/L	TOC–T&O
Saint-Quentin, France, 1997	250 m^3/d	groundwater	5 mg/L	pesticide
Vigneux, France, end 1997	35,000 m^3/d	clarified surface water	8 mg/L	TOC – T&O – pesticide
Kopper, Slovenia, end 1997	35,000 m^3/d	reservoir water	10 mg/L*	pesticide
Lausanne, Switzerland, 1999	65,000 m^3/d	lake water	5–10 mg/L*	T&O
Glendal, Texas, USA, 1999	1 900 m^3/d	surface water	NA	T&O
Saint Julien, France, 1999	400 m^3/d	groundwater	5 mg/L *	T&O
Bucey le Gy, France, 1999	1,200 m^3/d	groundwater	5 mg/L *	pesticides
Avranche, France, 2000	1,200 m^3/d	clarifed surface water	NA	pesticides
La Terrisse, France, 2000	6,000 m^3/d	clarified reservoir water	NA	TOC –T&O – pesticide
La Teysonne, France, 2000	2,000 m^3/d	reservoir water	NA	TOC – T&O

*Occasional additon of PAC

NA: Not available

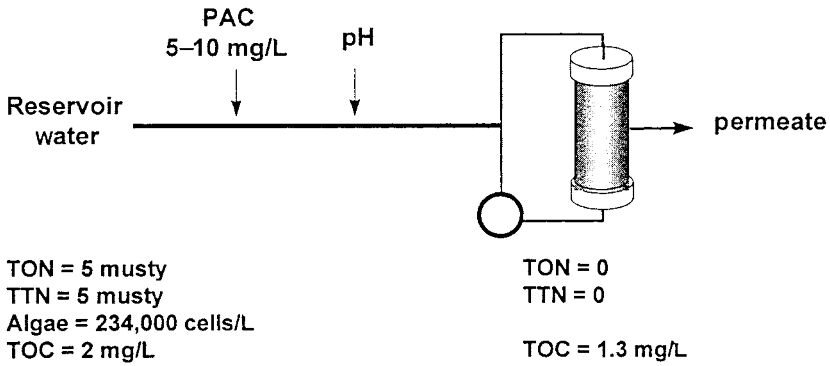

Figure 2 Example of full-scale application, Apié plant, 28,000 m^3/d

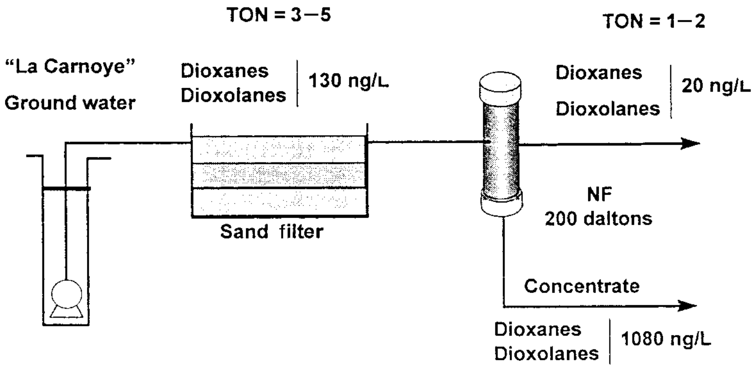

Figure 3 Example of TON reduction using NF filtration

capacities ranging from 200 to 65,000 m^3/d, are using this process, and at least 6 of them for controlling T&O episodes as a primary objective as reported in Table 1.

Figure 2 illustrates typical water quality for the Apié full-scale plant (28,000 m^3/d). The TON and threshold taste number (TTN) of 5 present in the raw water were reduced to 0 using the combination of UF and PAC process. The average carbon dose is currently 8

Table 2 Odour thresholds of various chlorination by-products or their biological transformation metabolites

Compound	Odour threshold (µg/L)	Reference
Phenol	9.5	Young *et al.* (1996)
2-chlorophenol	0.088	Young *et al.* (1996)
4-chlorophenol	10	Young *et al.* (1996)
2,4-dichlorophenol	5.4	Young *et al.* (1996)
2,6-dichlorophenol	5.9	Young *et al.* (1996)
2-bromophenol	0.024	Crane *et al.* (1996)
2,6-dibromophenol	0.016	Crane *et al.* (1996)
2-iodophenol	<0.4	Crane *et al.* (1996)
iodoform	0.5	*
chloroiodomethane	2	*
chloroform	7500	Young *et al.* (1996)
bromoform	5	*
2,4,6-trichloroanisole	0.00008	Young *et al.* (1996)
2,4,6-tribromoanisole	<0.00003	Crane *et al.* (1996)

*determined at 45°C, estimated from a Weber-Fechner plot

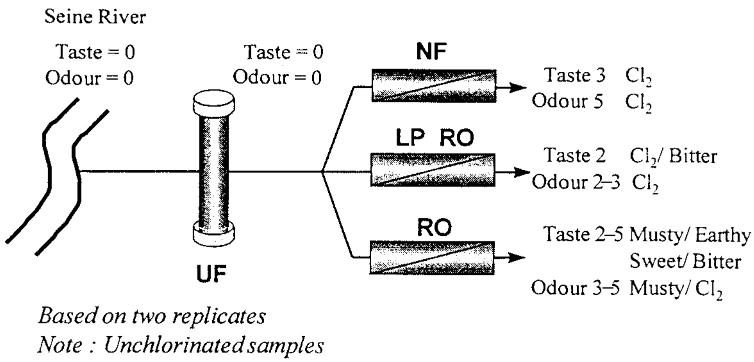

Figure 4 Example of TON increase using RO filtration

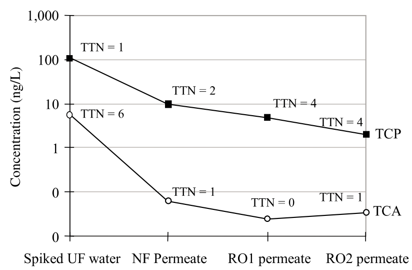

Figure 5 TCA and TCP removal by NF RO membranes

mg/l. It is suspected that most of the T&O causing compounds for this reservoir water come from algae by-products. Similar results are observed for the Vigneux plant as described elsewhere (Anselme *et al.*, 1999). The integration of adsorption (PAC) in the UF process makes it possible to use the UF process for treatment of surface or groundwater in the case of T&O occurrences. The place of the CRISTAL process (polishing treatment or single treatment) and the choice of the associated specific treatment (i.e. ozonation) was found to depend on the source water quality and odour causing potential (Laîné *et al.*, 1997).

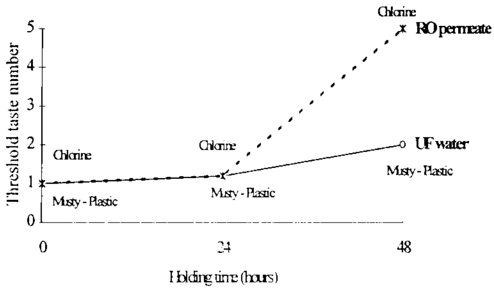

Figure 6 Impact of holding time for T&O occurrence on UF and RO treated waters

In terms of taste and odor control, surprising results were obtained when NF and RO membranes were used. Although the concentration of dioxanes and dioxolanes dropped significantly from 130 to 20 ng/L (85% removal) when NF was used, an odor was still present in the treated water (TON 1-2) as illustrated in Figure 3. However, it should be noted the fairly high removal of these low molecular weight compounds (88 et 74 respectively) despite the NF membrane cut-off of approximately 200 daltons.

More surprising results were obtained when water from the Seine River was filtered through NF, low pressure RO and RO membranes. The water, which presented no taste and odor initially, exhibited as significant degradation in terms of taste and odor (chlorinous and musty at TON of up to 5) as shown in Figure 4. Several hypotheses can be suggested :

1. The odorous compounds already present in the source water may have been masked and could be perceived only after softening and TOC removal.
2. Some non odorous compounds were oxidized to form odorous by-products.

Regarding the first hypotheses, compounds with low odour thresholds such as trichloroanisole could be responsible in the cases of no detection or unsolved T&O episodes. It should be noted, as shown in Table 2, that trichloroanisole has a very low odour threshold ranging from 0.03 to 0.08 ng/L. Therefore, TCA and TCP spiking were performed on prior NF and two RO membranes.

TCA and TCP seeding upstream NF and RO composite membranes (single element at 90% feed water recovery) indicated that both membranes were efficient to remove to a low level these musty-smelling compounds (Figure 5). However, an increase of the TON with time was observed. As presented in Figure 6, after 48 hours holding time, the threshold taste number of the RO permeate increased from 1 to 5 (chlorine taste), whereas the UF permeate threshold remained below 2 without any change in terms of taste descriptors. The cause of the chlorinous discriptor has not been elucidated at this time. Work is in progress to assess chlorinous T&O formation.

Conclusion

The present paper summarizes effects of various membrane processes on T&O reduction. It is clear from the examples shown that low-pressure membrane processes only (i.e. UF) would not reduce T&O in the treated water. On the other hand, combination of UF with PAC was found to be effective for T&O control. The use of NF, low pressure RO, or RO remains unclear for T&O control. Some surprising results were observed with the low cutoff membranes. These data still need to be confirmed on the full-scale pilot plant. However, work is still required in order to better characterise the T&O causing compounds, especially those associated with chlorinous odours.

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