Journal of Environmental Radioactivity 125 (2013) 86–92

Contents lists available at SciVerse ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Removal of radionuclides in drinking water by membrane treatment using ultrafiltration, reverse osmosis and electrodialysis reversal

M. Montaña [a,*], A. Camacho [a], I. Serrano [a], R. Devesa [b], L. Matia [b], I. Vallés [a]

[a] Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, ETSEIB Diagonal 647, 08028 Barcelona, Spain
[b] Aigües de Barcelona, AGBAR. Laboratory, General Batet, 5-7, 08028 Barcelona, Spain

ARTICLE INFO

Article history:
Received 31 May 2012
Received in revised form
17 December 2012
Accepted 6 January 2013
Available online 29 January 2013

Keywords:
Removal radioactivity
Gross alpha activity
Ultrafiltration
Electrodialysis reversal
Reverse osmosis

ABSTRACT

A pilot plant had been built to test the behaviour of ultrafiltration (UF), reverse osmosis (RO), and electrodialysis reversal (EDR) in order to improve the quality of the water supplied to Barcelona metropolitan area from the Llobregat River. This paper presents results from two studies to reduce natural radioactivity. The results from the pilot plant with four different scenarios were used to design the full-scale treatment plant built (SJD WTP). The samples taken at different steps of the treatment were analysed to determine gross alpha, gross beta and uranium activity. The results obtained revealed a significant improvement in the radiological water quality provided by both membrane techniques (RO and EDR showed removal rates higher than 60%). However, UF did not show any significant removal capacity for gross alpha, gross beta or uranium activities. RO was better at reducing the radiological parameters studied and this treatment was selected and applied at the full scale treatment plant. The RO treatment used at the SJD WTP reduced the concentration of both gross alpha and gross beta activities and also produced water of high quality with an average removal of 95% for gross alpha activity and almost 93% for gross beta activity at the treatment plant.

1. Introduction

Clean water, free of toxic chemicals and pathogens, is essential to human health. Common treatment methods used at waterworks are a combination of chemical oxidation, coagulation-flocculation, sand filtration and disinfection. However, in recent years, membrane technology has become an extraordinarily useful tool for the desalination of seawater, and this technology is also being increasingly used in the production of freshwater. Recent advances suggest that many issues involving water quality could be resolved or greatly ameliorated by using ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), electrodialysis (ED) or electrodialysis reversal (EDR) processes. UF and NF membrane filtration processes work by excluding contaminants using pore size constrains when water under pressure is forced to pass through a semi-permeable membrane with different pore sizes. Both the pore size and applied pressure must be adequate for the required purposes.

The RO membrane works as a molecular filter that rejects positively and negatively charged ions based on molecular weight when pressurized water is forced through the membrane. In contrast, the driving force for separation in ED and EDR processes is an electric potential, and an applied current is used to transport ionic species across selectively permeable membranes. The principal difference between ED and EDR is that EDR includes the additional step of a change in electrode polarity every 15–20 min, thus causing a reversal in ion movement. This step minimizes scale buildup on the membranes which means that EDR can operate for longer time periods between cleanings.

Van der Bruggen and Van der Casteele (2003) have reviewed the use of NF to remove cations, natural organic matter, biological contaminants, organic pollutants, nitrates and arsenic from groundwater and surface water. The UF process is also used for purification of water contaminated by toxic metal ions, radionuclides, organic and inorganic solutes, bacteria and viruses. For example, ultrafiltration assisted by complexation has been used to reduce uranium concentration (Kryvoruchko et al., 2004). An NF pilot plant experiment was set up to determine the uranium removal efficiency and for most experiments the uranium removal was about 95% (Raff and Wilken, 1999). In another study (Favre-Réguillon et al., 2008) demonstrated that uranium rejection depended on the uranyl species. In addition, RO effectively removes many inorganic contaminants, including many toxic metals and radionuclides, such as radium and uranium (Huikuri et al., 1998). RO can remove 87–98% of radium from drinking water and similar elimination can be achieved for alpha, beta and photon emitters

* Corresponding author.

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http://dx.doi.org/10.1016/j.jenvrad.2013.01.010

(EPA, 1998). Uranium and its complexes are very heavy, which allows the RO process to effectively remove (95–99%) uranium complex such as uranyl carbonate (Hansen, 2004). In principle, removal of radionuclides by ED/EDR is similar to RO. ED/EDR does not remove neutral species, such as UO_2CO_3, as it removes relatively small amounts of ions that have low mobility. As with RO, a prefiltration step may be necessary before both membrane processes (Ardnt, 2010).

There are some studies about the influence of conventional treatments applied at water treatment plants to reduce radioactivity (Gafvert et al., 2002; Jiménez and De la Montaña Rufo, 2002; Baeza et al., 2006, 2008; Palomo et al., 2010). In these works it was found that uranium and radium removal were very sensitive to the pH and coagulant used and also showed important influence of other ions presented in the waters.

Whether or not a particular treatment technology effectively removes radionuclides from drinking water depends on the contaminant's chemical and physical characteristics as well as the water system's characteristics (e.g., the source water quality and the water system size). Other considerations such as cost, service life and co-treatment compatibility are also important and must be taken into account.

The aim of this study was to evaluate the effectiveness of UF, RO and EDR for the removal of radioactivity in water by considering different scenarios from a pilot plant (Devesa et al., 2010) in order to obtain an initial estimation of the possible elimination of radioactivity by membrane technology. This paper also reports results using the final RO treatment applied at Sant Joan Despí Water Treatment Plant (SJD WTP) which supplies drinking water to the city of Barcelona.

2. Materials and methods

2.1. Water characteristics

The supply of drinking water to the metropolitan area of Barcelona has two main sources, surface water and occasionally water from wells. One of the plants that supplies water to Barcelona is the Sant Joan Despí Water Treatment Plant (SJD WTP) which catches water from the Llobregat River and on some occasions, as described in detail below, includes water from wells.

Mean daily water consumption is about 900,000 m^3, with about 50% coming from the Llobregat River and 45% from the Ter River.

The Llobregat river basin, flows through rural or industrialized areas. Its water has high salinity, with a dry residue of around 900 mg/L. This is mainly due to the nature of the upper part of the basin, which has had a long tradition of mining (sodium and potassium chlorides).

As previously mentioned, surface water catchment from the Llobregat is occasionally complemented by water from wells in the aquifers of the river delta. These wells are near the SJD WTP and are used mainly in periods of drought or on isolated days when river flow is low. They are also used when episodes of river water pollution prevent water catchment, in that it fails to reach the standards laid out in legislation on human drinking water or water company quality control requirements. These episodes are usually due to rains causing some rivers and collectors of polluted water to overflow. Under normal conditions, this polluted water by-passes the treatment plant and is returned to the river down- stream. Occasionally, pollution is caused by industrial dumping.

The origin of alpha and beta activity in the Llobregat river basin has been previously investigated and it was established that it was mainly due to ^{234}U, and ^{238}U for alpha activity and ^{40}K for beta activity (Ortega et al., 1996; Camacho et al., 2010). Other isotopes of a natural or artificial origin were not normally found. Routine monitoring of the resources and the supply system have shown that conventional treatments at the SJD WTP do not significantly decrease the amount of alpha and beta total radioactivity.

2.2. Scenarios of the study

This paper presents radioactivity results from two studies. Firstly, results from a pilot plant with four different designs or scenarios are presented. Secondly, results from the full-scale treatment plant built according to the findings of the pilot study are commented on.

2.2.1. Pilot plant

The pilot plant was located within the SJD WTP which catches water from the final stretch of the Llobregat river basin. The conventional full-size SJD WTP has the following treatment stages: pre- chlorination, coagulation-flocculation, sand filtration, ozonation, granular activated carbon (GAC) filtration and post-chlorination.

This pilot plant was set up with three membrane modules: UF, RO and EDR (Table 1). In addition, it also had a module which reproduced the polishing treatment (ozonation plus GAC filtration) used at the SJD WTP. The operating conditions of the latter were adapted to coincide, as far as possible, with those of the full-size plant (ozone dosage, 3 mg/L; GAC, Chemviron F-40; contact time in carbon filters, 9 min).

Four different scenarios were studied in the pilot plant using the following configurations (Fig. 1):

Scenario 1: composed of the following stages: pumping of raw water from the Llobregat River, UF of the whole flow, and subsequently two parallel treatments: RO and conventional (ozonation and GAC filtration) treatments. This scenario produced two final effluents, one after the RO treatment and another after GAC filtration. For the radiochemical study, four effluents were sampled: raw water and after UF, RO and GAC treatments.

Scenario 2: was analogous to the previous one but RO was replaced by EDR. The effluents were the same: raw water and after UF, EDR, and GAC treatments.

Scenario 3: is arranged completely in series: catchment of raw water, UF, EDR and GAC filtration.

Scenario 4: analogous to Scenario 3 but UF was replaced by sand filters, which is the conventional treatment at the SJD WTP: catchment of raw water, sand filters, EDR and GAC filtration.

Two samples per sampling point (1, 2, 3 and 4 in Fig. 1) for each scenario were taken, at the raw water, after UF, after RO, after EDR and after GAC filtration. All samples were analysed for gross alpha activity and gross beta activity. Uranium content was tested in some samples. There was a sequential arrangement of the scenarios in time, so feed water characteristics suffered from some fluctuations.

2.2.2. Full-scale treatment plant

The study of the performance of the different scenarios of the pilot plant showed that the best results (radiological, chemical,

Table 1
Main characteristics of the membrane modules (pilot plant).

	UF	RO	EDR
Capacity (L/s)	3.5	1.7	1.5
Membrane	ZeeWeed 500B	BW30LE-440	AQ-X
Manufacturer	Zenon	Filmtec	Ionics
Stages	1	1	2