Gross alpha and gross beta particle activity in recycled water for augmentation of drinking water supplies

Clemencia Rodriguez, Brian Devine, Angus Cook, Philip Weinstein and Paul Van Buynder

ABSTRACT

An assessment of potential health impacts of radioactive compounds in recycled water to augment drinking water supplies was conducted. Gross alpha and gross beta particle activity was selected for the screening of radioactive species. Samples both pre- and post-reverse osmosis treatment of secondary effluent in Perth, Australia were examined in both a full-scale and in a pilot water reclamation plant. Risk quotients (RQs) were estimated by expressing the mean (RQ mean) and the maximum concentration (RQ max) at each sampling point as a function of the recommended Australian screening levels of 0.5 Bq/L. The results indicate that reverse osmosis (RO) is able to reduce the concentration of gross alpha particle activity (average removal of almost 80%) and gross beta particle activity (average removal of 95%) and produce water of high quality. Maximum gross alpha particle activity in the recycled water was 0.023 Bq/L and maximum gross beta (excluding ^{40}K) particle activity was 0.03 Bq/L, which correspond to an RQ max of 0.07 for gross alpha and 0.06 for gross beta particle activity, respectively. No increased human radiological risk is anticipated if recycled water is used to augment drinking water supplies in Perth.

Key words | Australia, radionuclides, reverse osmosis, water quality, water reuse

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INTRODUCTION

Radioactive materials are a natural part of the environment and are found in varying concentrations depending on extraterrestrial, climatic, geological and geographical conditions. Radionuclides are elements that contain unstable nuclei. They have the potential to decay into a lower energy state by releasing either alpha or beta particles, or gamma rays. Radionuclides and their daughters (often isotopes of another element) progress through a defined "decay series". In nature, the two most common decay series are those of uranium-238 and thorium-232 (Canadian Environmental Protection Act 2000).

Humans are mainly exposed to radiation from naturally occurring radioactive sources and, to a lesser extent, from anthropogenic sources. The US EPA estimates that natural sources contribute on average 80% of the human radiation dose (US EPA 2005). In Canada it is estimated that natural sources contribute on average more than 98% of the human radiation dose, excluding medical exposures (FPTCDW 2008). In North America, the average individual dose of radiation from natural sources is estimated to be 2.4 millisievert (mSv), of which less than 10% comes from ingestion of food and drinking water. Based on Canadian estimates, an individual may receive a total annual dose range from background radiation of 1.2–3.2 mSv, based on geographical location (Canadian Environmental Protection Act 2000). The estimated average annual dose for Australia is estimated to be 2 mSv per year (NHMRC & NRMMC 2004).

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Generally, the presence of radioactive materials is a concern only when concentrations become sufficiently elevated above background levels to pose a potential health risk. Typically, only trace levels of naturally occurring radionuclides are transferred to humans via the various exposure pathways. Although human exposure to radiation sources is derived primarily from background natural radiation, a person's occupation, time spent outdoors, geographical location, past medical treatments and diagnostic testing, time spent traveling in aeroplanes and other activities can greatly impact the relative contributions of natural and man-made radioactive sources.

People can be irradiated either externally or internally through inhaling airborne radioactive substances or consuming food or water which contains radioactive material. Weighting factors are used to provide a measure of the overall effect of any radioactive exposure, because different types of radioactive particles have different penetrating power, and different organs and tissues in the body have different sensitivities to radiation. For example, alpha particles have very low penetration of tissue but will cause considerable cell damage over a short range, whereas beta particles and gamma radiation deposit their energies over a greater range (Canadian Environmental Protection Act 2000).

Occurrence

Radionuclides may enter wastewater by: (1) natural sources, (2) natural sources concentrated or enhanced by human activity and (3) man-made sources. Natural sources of radiation include geologic formations and soils that contain uranium, radium, thorium, radon and other nuclides that are radioactive. Naturally occurring radioactive materials can also be enhanced by human activity and by technologies associated with extraction processes. These materials, referred to as Technologically Enhanced Naturally Occurring Radioactive Materials, may be introduced to the sewerage system from ground and surface water, plants and food, as well as from potential industrial discharges (e.g. water treatment plants, mining and petroleum industries, fertilizers, electronics, ceramics, foundries and paper/pulp mills) (US EPA 2005). Man-made radioactive materials are used in industry, medicine and research. Point sources of contamination include Department of Energy facilities, hospitals, universities and pharmaceutical companies.

In some countries, the largest anthropogenic contributor to wastewater contamination is the nuclear power industry, either in the production of fuel assemblies or controlled releases from reactors. In most regions, the level of radionuclide contamination in wastewater resulting from weapons testing, accidents, industrial applications and research institutions is generally considered to be less than that associated with medical sources (EPA QLD 2000). Medical procedures such as thallium heart stress tests and tumour irradiation therapies are an important source of radionuclides in wastewater. The radionuclides most commonly used in biotechnology, hospitals and medical facilities are ^{14}C, ^{3}H, ^{35}S, $^{32/33}P$, ^{63}Ni, ^{99}Tc and radio iodines. In the US, the average doses from nuclear medicine and x-ray examination procedures are about 0.14 and 0.39 mSv per year, respectively (US EPA 2005). Other sources of anthropogenic radiation include consumer products such as static eliminators (containing polonium-210), smoke detectors (containing americium-241), cardiac pacemakers (containing plutonium-238), fertilizers (containing isotopes from uranium and thorium decay series) and tobacco products (containing polonium-210 and lead-210). It is estimated that the average dose from consumer products to a person living in the US is 0.1 mSv per year (excluding tobacco contributions) (US EPA 2005).

In Australia, direct discharge of radioactive material into the sewerage system is regulated by each state. Some states have adopted limits based on the International Atomic Energy Agency – Basic Safety Standards (BS115) (IAEA 1996). For example, in some states soluble radioactive material may be disposed of via sinks if the specific activity of the material is less than 100 Becquerel per litre (Bq/L). Consequently, risk reduction is based on dilution of the radionuclides by the wastewater stream (EPA QLD 2000). Radioactive elements represent a special case of inorganic pollutants and they show similar environmental behaviours to their stable chemical isotopes. Therefore, during the wastewater treatment process, radionuclides are concentrated in the sewage sludge, similar to solids and toxic metals. Despite the efficiency of wastewater treatment plants (WWTP) to remove radionuclides, maximum concentrations of gross alpha and gross beta particles – above

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the screening value of 0.5 Bq/L – in secondary effluent of 0.7 Bq/L and 1.2 Bq/L, respectively, have been found (NRMMC, EPHC & NHMRC 2008). Secondary effluent treatment using microfiltration (MF) and reverse osmosis (RO) is efficient in removing radioactive contaminants and RO is considered one of the best available technologies for the removal of radionuclides (Electronic-Code of Federal Regulations 2008). Reported removal efficiencies by RO varied from 70–99% and efficiency is influenced by the specific chemical characteristics of the water (Health Canada 2006). For example, removal efficiency for radium and uranium was above 90% in a RO plant in Florida with a TDS range from 895–3373 mg/L. A range of indirect potable reuse projects have reported radionuclide concentrations that fall within the drinking water quality standards stipulated by US EPA and WHO (Singapore Government 2002; OCWD 2006; WBMWD 2006).

Regulations

Cancer is the principal endpoint used to evaluate the health risk from alpha and beta particle emitters, and risk assessment procedures to estimate the cancer risk from radionuclides have been extensively developed. Screening levels for gross alpha and gross beta particle activity have been established based on the carcinogenic potency of the radionuclides included in these categories. Table 1 presents the screening levels for gross alpha and gross beta particles in different countries and institutions. The recommended screening level in the ADWG for each one gross alpha and gross beta particle activities is 0.5 Bq/L. The guidelines recommend that, if the screening level is exceeded, then radium-226 and radium-228 should be determined (NHMRC & NRMMC 2004). For gross beta, the screening value of 0.5 Bq/L excludes the contribution from ^{40}K, a natural beta emitter that is normally absorbed from ingested food and does not accumulate in the body. The total dose of both alpha and beta activity at the screening level corresponds to approximately 0.35 mSv per year, which is one-third of the minimum dose at which intervention is recommended (NHMRC & NRMMC 2004). The guidelines also recommend that the sum of the annual doses from all radionuclides should be less than 0.5 mSv. It is recommended that a value between 0.5–1 mSv should lead to notification of the health authorities, a value above 1 mSv requires operational changes and a value above 10 mSv is unacceptable for drinking. ^{40}K is not considered to be of significance to health because the average contribution of this nuclide to the annual effective dose from background radiation is estimated at 0.18 mSv. For drinking water utilities, the guidelines recommend that radiological quality be assessed when a new supply is brought into service, and then every two years for groundwater supplies, and every five years for surface water supplies.

In Europe, the limit for the uptake of radionuclides via drinking water has been established in the form of a "total indicative dose" of 0.1 mSv per year (European Commission 1998). The total indicative dose needs to be calculated from the contribution of each natural or man-made radionuclide present in drinking water excluding ^{3}H, ^{40}K, radon and radon decay series. However, given the cost and difficulties of measuring each radionuclide, it has been recommended too that screening methods be applied for a first cost-saving assessment of the radioactive content in drinking water. Consequently, if the gross alpha activity in a tap water sample is lower than 0.1 Bq/L and the gross beta activity

Table 1 | Drinking water guideline values for gross alpha and gross beta particles (Bq/L)

Guideline	Gross alpha	Gross beta	Source
Australia	0.5	0.5 (excluding ^{40}K)	NHMRC & NRMMC (2004)
WHO	0.5	1	WHO (2006)
US EPA	0.55	0.04 mSv/yr	US EPA (2006)
Title 22	0.55	1.85	ACWA (2007)
Health Canada	0.1	1	FPTCDW (2008)
New Zealand	0.1 (excluding ^{222}radon)	0.5 (excluding ^{40}K)	Ministry of Health (2005)
European Union	0.1	1	European Commission (1998)

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does not exceed 1.0 Bq/L, it can be assumed that the annual total indicative dose to adults is less than 0.1 mSv per year.

For drinking water, the US EPA has established a maximum contaminant level (MCL) of 0.55 Bq/L for gross alpha particle emitters and an MCL of 0.04 mSv/yr for man-made beta particles and photon emitters (US EPA 2006). The gross alpha particle MCL was originally intended by US EPA to screen drinking water for an unacceptable amount of radioactivity from natural sources: primarily represented by uranium and radium-226, and present mainly in groundwater. However, it also addresses radium-228, a beta emitter. The gross beta particle/photon standard was originally intended to screen drinking water for an unacceptable amount of radioactivity from man-made sources, with the exception of the man-made isotopes strontium-90 and tritium. Some of the natural beta emitters are also excluded from this screening. In the US regulations, drinking water utilities reporting a monitoring result above the MCL for gross alpha or gross beta require quarterly monitoring until four consecutive quarterly samples are below the MCL.

In this paper, gross alpha and gross beta particle activity before and after advanced treatment at Kwinana water reclamation plant (KWRP) and Beenyup pilot plant (BPP) in Perth, Western Australia, was tested. Samples from the secondary effluent at Subiaco WWTP were also tested. Gross alpha and gross beta particle activity was selected for the screening of radioactive species based on the recommendations of the Australian Drinking Water Guidelines (ADWG). The purpose of radionuclide screening was to determine whether wastewater and recycled water contains radionuclide activity greater than the natural background activity. Background concentrations in the groundwater used as source for drinking water supplies was also tested. Risk quotients were calculated to establish whether gross alpha and gross beta particle activity in recycled water was above concentrations of health significance.

METHODOLOGY

Both the KWRP and the BPP have a multiple barrier approach, using MF and RO. They are operated by Water Corporation Western Australia. For KWRP the influent is a mixture of industrial and residential facilities and for BTP the influent is mainly of domestic nature. A summary of the membrane characteristics and operational conditions of the MF/RO treatment is presented in Table 2.

Samples were collected from Beenyup WWTP, Subiaco WWTP and KWRP in three monitoring events in May–June 2007, September 2007 and January 2008. All samples were collected using consistent protocols and procedures to ensure adequate sampling preparation, preservation and transportation to the laboratory. All samples were collected as grab samples and collected on week days (Monday to Friday). Samples from KWRP or BPP were taken at three different points within the MF/RO treatment: before MF, after MF and after RO, as depicted in Figure 1. Data were analysed in Stata version 10 (Stata Corp. 2007). A total of 46 analytical samples were reported, excluding field blanks, trip blanks and replicates taken as part of the QA/QC program. Of 46 samples, 22 (47%) were taken at KWRP and 30 (65%) were analysed during the May–June sampling event. No samples were collected during the November 2006 sampling event. Gross alpha and gross beta at BPP were tested during the September 2007 and January 2008 sampling events. Groundwater was also tested during May–June 2007 and January 2008 to obtain background levels of gross alpha and gross beta particle activity.

Samples from all three monitoring events were collected and analysed by the Australian Radiation Protection and the Nuclear Safety Agency (ARPANSA), a NATA accredited laboratory. Samples sent to the ARPANSA were preserved in nitric acid, stored at 4°C and filtered (0.45 μm). Gross alpha and gross beta radioactivity concentrations were determined by gas flow proportional counting (ERH_RAS_SOP_0100). Limits of detection (LOD) for gross alpha and gross beta particles were variable as they are dependent on the concentration of dissolved solids in the sample. Typical limits of detection were 0.01 Bq/L for both gross alpha and gross beta particle activity. To account for the variability on radiation emissions, the reported uncertainty (coverage factor $K = 2$) was 24% for gross alpha and 12% for gross beta. Potassium-40 (^{40}K) was determined by measurement of potassium by flame AAS with an LOD of 0.05 mBq/L and an uncertainty (coverage factor $K = 2$) of 10% (at 0.5 Bq/L). Alpha measurements were ^{241}Am equivalent and the beta measurements were ^{40}K equivalent.

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Table 2 | Summary of Kwinana and Beenyup plant characteristics

	Kwinana Water Reclamation Plant (KWRP)	Beenyup Pilot Plant (BPP)
Project status	Operating since November 2004	Operating since September 2007
Capacity	17 ML/d	100 kL/d
Inflow	Effluent from Woodman Point WWTP	Effluent from Beenyup WWTP
Membranes	MF material: Polypropylene MF: Memcor CMF-S system RO material: Polyamide and polysulfone RO: Dow – Filmtec FT30	MF material: Polyvinylidene fluoride (PVDF) MF: Memcor CMF-L 6L10V cartridge hollow fibre filters RO material: Composite polyamide RO: Hydranautics ESPA-2 4040, 4 inch membranes
Design flux	MF recovery average rate 90% RO recovery average rate 80% Average permeate flow: 140 m^3/h Average concentrate flow: 35 m^3/h	MF recovery ~ 97% RO recovery average rate 70% Average permeate flow: 2.8 m^3/h Average concentrate flow: l.2 m^3/h RO membrane salt rejection: 98.4%
Biofouling prevention	Feed water dosed with sodium hypochlorite, ammonia and sulfuric acid to reduce biological accretion and membrane damage. Air forced around the membrane walls during the backwash cycle helps to dislodge particles.	Feed water dosed with sodium hypochlorite and sulfuric acid and aqueous ammonia to reduce biological accretion and membrane damage. Hypochlorite and ammonia form monochloramine levels that are maintained throughout the treatment to prevent biological fouling of the RO membranes.
Scaling prevention	Chemical anti-scalant in the RO feedwater used to prevent the supersaturated salts developed from precipitating out onto the surface of the RO membranes. High pH cleaning quarterly to remove fouling. 3 weekly, automated (manually intiated) Clean In Place process uses a re-circulated heated weak acid or caustic/detergent solution to wash the membranes.	Chemical anti-scalant in the RO feedwater used to prevent supersaturated salts developing in the reject water from precipitating out onto the surface of the RO membranes. Clean In Place initiated when the pressure drop across membranes climbs outside of normal range.

Samples from the May–June 2007 monitoring event were also analysed by the Radiation Health Branch (RHB), Department of Health, Western Australia. Samples analysed by the RHB were preserved in nitric acid, filtered (0.45 μm) and evaporated on a stainless steel planchet by Chemistry Centre of Western Australia based upon ISO Method 9697:1992 and following the sampling collection/preservation standards based on AS/NZS 5667.1 (1998). The alpha and beta count rate of each sample was measured (in 2π geometry) using an internal gas flow proportional chamber over a 180 min time period. The reported alpha efficiency was 33.4% ± 1.6% and the beta efficiency was 54.7% ± 2.5%. The beta contribution from ^{40}K was not subtracted from the gross beta counts because potassium was not measured. Alpha measurements were ^{241}Am equivalent and the beta measurements were ^{90}Sr equivalent. In addition to the uncertainty in the counts, the measurement results have an additional total uncertainty of 5%, which is based on the sum of random counting error and the estimated upper limits of systematic error in the measurement. Data were reported in counts per minute and then converted to Bq/L. A total of 20 samples were analysed by the RHB and 26 samples were analysed by the ARPANSA (Table 3).

The RQs were estimated by expressing the mean (RQ mean) and the maximum concentration (RQ max) in each sampling point as a function of the recommended screening levels for gross alpha and gross beta particle activity.

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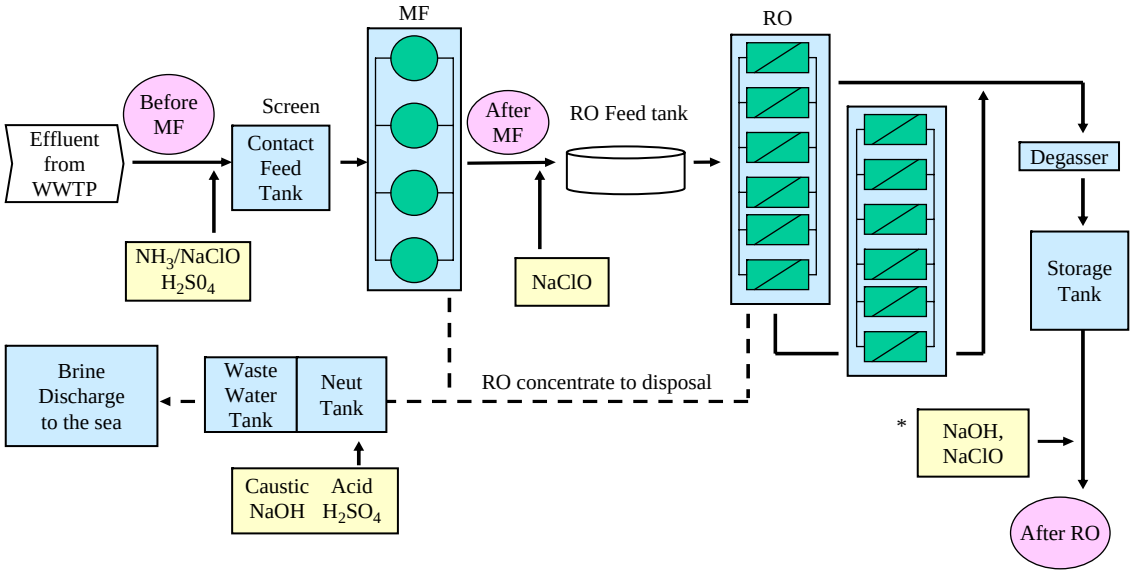

Figure 1 | Schematic representation of the advanced treatment at KWRP and BPP. Sample points represented by circles. Note: *There is no post-RO chemical dosing at the BPP.

RESULTS

The distribution of samples and the summary statistics for the gross alpha particle activity (Table 4) indicate that the highest mean detection for alpha particle activity occurs in the influent of the BPP (0.081 Bq/L) followed by the secondary effluent at Subiaco (0.062 Bq/L) and the influent of the KWRP (0.053 Bq/L). Gross alpha particle activity was lower in the product water of both plants (KWRP = 0.023 Bq/L and BPP = 0.023 Bq/L). In all samples, the gross alpha particle activity was below the screening level of 0.5 Bq/L and the calculated RQs using the maximum concentration observed were one order of magnitude below 1 after the advanced treatment (Table 4).

The results for the gross beta particle activity (Table 5) indicate that the maximum gross beta activity occurs at KWRP (after MF = 1.41 Bq/L and before MF = 1.29 Bq/L) and in secondary effluent at Beenyup (1.06 Bq/L). The maximum detection in the product water at BPP (0.038 Bq/L) and KWRP (0.258 Bq/L) was, respectively, 19 times and 3.5 times lower than the maximum concentrations before MF. The mean gross beta activity in the groundwater was two times higher than in the KWRP product water and almost 13 times higher than in the BPP product water. These results indicate that groundwater contains higher amount of natural background radiation compared to the gross beta particle activity in the recycled water. An RQ was calculated using the screening level of 1 Bq/L proposed by

Table 3 | Frequency of gross alpha and gross beta particle activity samples by event, laboratory and location

				Laboratory				Location WRP			
Event	Month	Year	No. days	ARPANSA	RHB	GW	WW	Before MF	After MF	After RO	No. of samples
1	November	2006	4	-	-	-	-	-	-	-	-
2	May–June	2007	5	10	20	4	8	6	6	6	30
3	September	2007	2	8	0	0	0	4	0	4	8
4	January	2008	2	8	0	4	0	2	0	2	8
Total				26	20	8	8	12	6	12	46

GW, groundwater; WW, wastewater; WRP, Water reclamation plant

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Table 4 | Gross alpha particle activity by type of water and location

Location	Point	*n*	Mean	SD	Min	Max	RQ mean	RQ max
KWRP	Before MF	4	0.053	0.039	0.017	0.09	0.106	0.18
	After MF	3	0.027	0.012	0.018	0.035	0.054	0.07
	After RO	4	0.023	0.010	0.016	0.035	0.046	0.07
Wanneroo	Groundwater	4	0.041	0.018	0.02	0.062	0.082	0.12
Subiaco WWTP	Wastewater	2	0.062	0.051	0.025	0.098	0.124	0.19
BPP	Before MF	4	0.081	0.039	0.023	0.11	0.162	0.22
	After RO	2	0.023	0.015	0.012	0.033	0.046	0.07

n: number of samples, SD Standard deviation, RQ Risk quotient calculated using ADWG guideline value of 0.5 Bq/L

Table 5 | Gross beta particle activity by type of water and location

Location	Point	*n*	Mean	SD	Min	Max	RQ mean	RQ max
KWRP	Before MF	4	0.908	0.291	0.665	1.287	0.91	1.29
	After MF	3	1.313	0.084	1.261	1.409	1.31	1.41
	After RO	4	0.146	0.126	0.03	0.258	0.15	0.26
Wanneroo	Groundwater	4	0.322	0.273	0.149	0.722	0.32	0.72
Subiaco WWTP	Wastewater	2	0.854	0.189	0.72	0.987	0.85	0.99
BPP	Before MF	4	0.743	0.216	0.597	1.058	0.74	1.06
	After RO	2	0.025	0.018	0.012	0.038	0.03	0.04

n: number of samples, SD Standard deviation, RQ Risk quotient calculated using the WHO value of 1 Bq/L

WHO, Health Canada and the EU (Table 1). Using this screening level RQ max was close to or above 1 in all secondary effluent samples. However, the RQ mean is below 1 for all the samples, except after MF at KWRP (RQ mean = 1.31). The lowest RQ max were observed after RO at BPP (RQ mean = 0.04) and KWRP (RQ mean = 0.26).

A total of 13 samples analysed by the ARPANSA reported ^{40}K. The gross beta particle activity, excluding the contribution from ^{40}K, is presented in Table 6. Negative values were reported for some samples, indicating that the amount of natural background radiation (^{40}K) measured by the instrument was larger than the total amount of radioactivity measured in the sample. Negative values are therefore the result of the randomness of radioactive emissions and the very low concentrations of these contaminants in the samples. None of the gross beta samples, excluding the ^{40}K contribution, was above the screening level of 0.5Bq/L and the RQ max in the product

Table 6 | Gross beta particle activity excluding ^{40}K by type of water and location

Location	Point	*n*	Mean	SD	Min	Max	RQ mean	RQ max
KWRP	Before MF	2	−0.055	0.006	−0.059	−0.05	-	-
	After RO	2	0.016	0.008	0.01	0.023	0.032	0.046
Wanneroo	Groundwater	3	0.048	0.008	0.042	0.057	0.096	0.114
Subiaco WWTP	Wastewater	1	−0.026		−0.026	−0.026	-	-
BPP	Before MF	3	0.012	0.034	−0.014	0.05	0.024	0.1
	After RO	2	0.017	0.019	0.003	0.03	0.034	0.06

n: number of samples, SD Standard deviation, RQ Risk quotient calculated using ADWG guideline value of 0.5 Bq/L

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Table 7 | Treatment performance

Parameter	Location	Before MF (Bq/L)	After RO (Bq/L)	Efficiency (%)	Removal*
Gross alpha	KWRP	0.084	0.019	77.4	Moderate
	KWRP	0.09	0.014	84.4	Moderate
	BPP	0.1	0.033	67	Moderate
	BPP	0.11	0.012	89.1	Moderate
Gross beta	KWRP	1.287	0.251	80.5	Moderate
	KWRP	0.665	0.03	95.5	Good
	BPP	0.597	0.038	93.6	Good
	BPP	0.606	0.012	98.0	Good

Note: Removal* (Good >90%, Moderate 50–90%, Intermediate 25–50%, Poor <25%)

water was an order of magnitude below 1 – RQ mean after RO KWRP = 0.032 and BTP = 0.034 (Table 6).

Treatment efficiency was calculated from samples analysed by the ARPANSA before MF and after RO on the same date and location (Table 7, Figure 2). Gross beta particle activity was higher in the plant influent than the gross alpha activity and, as a consequence, the (%) of removal during the advanced treatment was higher for gross beta particle activity ($n = 4$, mean = 91.9%, median = 94.6%, SD = 7.8%, min = 80.5% and max = 98%) than for gross alpha particle activity ($n = 4$, mean = 79.5%, median = 80.9%, SD = 9.6%, min = 67% and max = 89.1%).

Bootstrap simulations were performed in Stata in order to estimate the 95% confidence intervals (CI) of a large sample. Gross alpha and gross beta observations before MF and after RO were re-sampled with 250 and 1,000 replacements and the estimated 95% CI are presented in Table 8. The estimated 95% CI indicates that, if wastewater conditions remain unchanged, the gross alpha particle activity will be between 0.043–0.089 Bq/L in the plant influent, which is well below the screening value of 0.5 Bq/L. Similarly, the estimated upper CI for gross beta particle activity (0.96 Bq/L) will be below the WHO guideline value of 1 Bq/L.

Quality assurance/quality control

There were significant differences between the laboratories in the mean of the gross alpha particle activity, even though alpha measurements were ^{241}Am equivalent for both laboratories (For ARPANSA: $n = 13$, mean = 0.061 Bq/L, SD = 0.036 Bq/L; for RHB: $n = 16$, mean = 0.021 Bq/L, SD = 0.005 Bq/L; Kruskal–Wallis test for comparison of laboratory measurements $\chi 2 = 4.8$, $p = 0.02$). The mean gross beta activity was higher for the RHB but the differences were not statistically significant. (For ARPANSA: $n = 13$, mean = 0.35 Bq/L, SD = 0.30 Bq/L; for RHB: $n = 16$, mean = 0.85 Bq/L, SD = 0.43 Bq/L). The differences may be explained by the variability in radiation

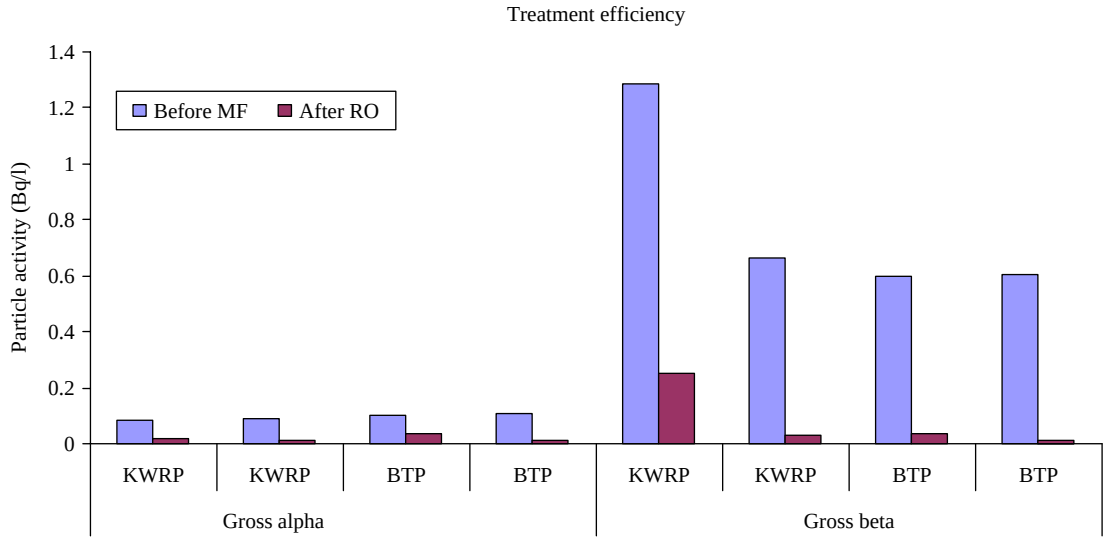

Figure 2 | Removal of gross alpha and gross beta particle activity during advanced treatment.

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Table 8 | Estimated 95% CI for gross alpha and gross beta particle activity (Bq/L)

	Summary of observed data					Bootstrap estimated 95% CI	
	n	Mean	SD	Min	Max	Re-sampling = 250	Re-sampling = 1000
Gross alpha							
Before MF	10	0.657	0.039	0.017	0.11	(0.044–0.088)	(0.043–0.089)
After RO	6	0.02	0.007	0.012	0.033	(0.014–0.026)	(0.014–0.026)
Gross beta							
Before MF	10	0.831	0.232	0.597	1.287	(0.693–0.968)	(0.697–0.964)
After RO	6	0.105	0.116	0.012	0.258	(0.016–0.195)	(0.022–0.189)

emissions and the different methods used by the laboratories. For example, beta measurements were ^{90}Sr equivalent for the RHB and ^{40}K equivalent for the ARPANSA Laboratory.

The Bland–Altman plot for gross alpha and gross beta activity is presented in Figure 3. The graphs display the scatter of the differences between the two laboratories plotted against the averages. For gross alpha activity, the plot indicates that differences are higher when the concentration of gross alpha activity in the samples is low. The plot for gross beta activity is less clear but tends to show that the differences between the laboratories are higher when the gross beta concentration in the samples is high.

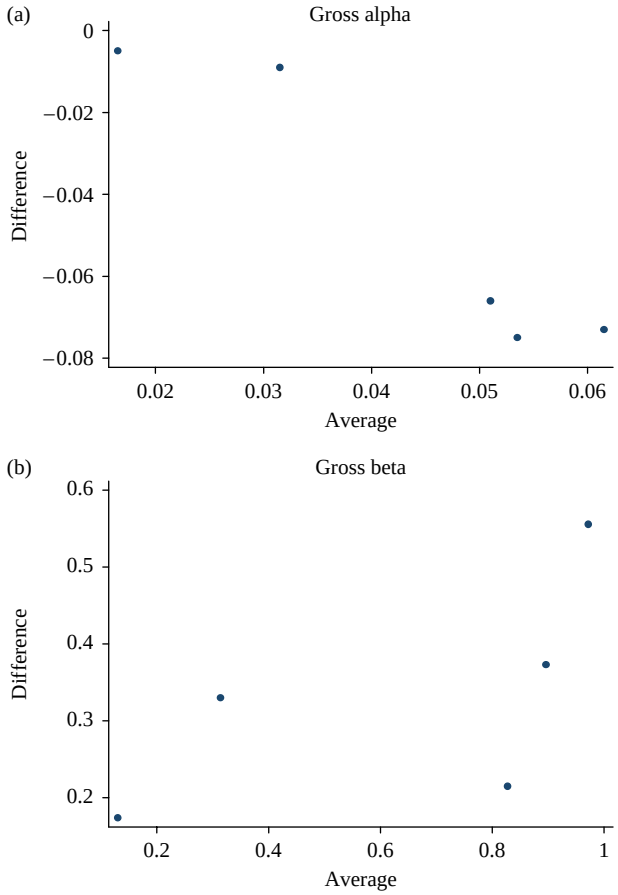

Figure 3 | Bland–Altman plots of the differences between the samples of the two laboratories plotted against the averages for gross alpha (a) and gross beta (b) particle activity.

DISCUSSION

A profile of radiological water quality with data on gross alpha and gross beta activity in different types of water (groundwater, wastewater and recycled water) was presented. The results indicate that none of the samples were above the screening level of 0.5 Bq/L for gross alpha or gross beta (excluding ^{40}K) particle activity. Maximum gross alpha particle in the secondary effluent was 0.11 Bq/L and maximum gross beta (excluding ^{40}K) particle activity was 0.057 Bq/L, which correspond to an RQ max of 0.22 for gross alpha and 0.1 for gross beta particle activity, respectively. The gross alpha and gross beta particles are concentrated in the sewage sludge during the wastewater treatment, similar to solids and metals, and therefore WWTPs are an important treatment barrier for their removal.

The calculated RQs of gross alpha and gross beta particle emitters were also lower in the product water compared to the concentrations in groundwater. For example, the RQ for the gross alpha particle activity in the product water was 0.07, compared to 0.18 for groundwater (i.e. more than 2.5 times higher in the groundwater than in the recycled water). Similarly the RQ

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calculated after the advanced treatment was one order of magnitude below 1 for the gross beta particles. The results demonstrate that reverse osmosis is able to further reduce the concentration of gross alpha and gross beta particle activity and produce water of high quality with an average removal of 95% for gross beta and almost 80% for gross alpha particle activity. These results are also consistent with other studies in which the (%) of gross alpha and gross beta particle removal by membranes is above 90% (Moritz *et al.* 1995; Asano *et al.* 2007) and with the results of water recycling schemes using membranes for indirect potable reuse (Singapore Government 2002; OCWD 2006; WBMWD 2006).

No further nuclide sample characterization was required to identify specific alpha or beta emitters given that no excessive activity levels were found for gross alpha or gross beta particle activity. However, if gross alpha and gross beta are above the screening levels, the steps illustrated on pages 10–12 of the Australian drinking water guidelines (NHMRC & NRMMC 2004) need to be implemented in the first instance to elucidate whether or not increased concentrations of particle emitters are from naturally occurring sources.

As there is no nuclear power industry in WA at the current time (2008), the majority of the anthropogenic radioactivity in domestic wastewater is derived from medical sources, in particular those used for diagnostic and therapeutic procedures. The results also indicate that that the implementation of the state's radiation legislation by the Radiological Council effectively controls industrial and hospital discharges of radioactive materials into the sewerage system. Nevertheless, as part of the risk management strategy, it is important to encourage the implementation of radionuclide point source control measures. For example, current radiation control legislation in WA requires new facilities that may not be able to meet discharge limits to install holding tanks to allow disposable radionuclides to further decay to acceptable levels before discharge. Installation of holding tanks is therefore encouraged not only for new hospitals and industrial facilities using radioactive materials but also for existing facilities to decrease the alpha and beta particle activity in wastewater.

Given that the main anthropogenic source of radionuclides in Perth's sewerage system is through hospital discharges, a reliable and constant update of information on their use including amounts, locations and timing is key to efficient and cost-effective monitoring and assessing wastewater quality changes that may impact on the quality of the recycled water. Water utilities undertaking indirect potable reuse should contact the organisations and government agencies responsible for assessing industrial and environmental sources of radionuclides. Such organisations (e.g. the Radiological Council of WA) may provide information on: (i) natural background levels of radionuclides in soils and groundwater, (ii) facilities that may be permitted to discharge residuals to the sewerage system and (iii) registrations and licenses of radioactive substances.

Differences in the levels reported between the laboratories were observed. The ARPANSA reported higher mean concentration for alpha particles (mean difference = 0.04 Bq/L) and the RHB reported higher concentrations of gross beta particle activity (mean difference = 0.5 Bq/L). These differences may be due to variability in the counting and differences in sample preparation and analysis. Given that the ARPANSA is NATA-accredited for gross alpha and gross beta particle analysis, it is recommended to continue the testing using this laboratory to minimise random and systematic error in the final estimate of the gross alpha and gross beta concentrations. Gross alpha and gross beta methods are significantly compromised when total suspended solid (TSS) levels in wastewater samples are greater than 500 mg/L. In this study, the TSS in secondary effluent ranged between 5–65 mg/L and was below the LOR of 5 mg/L after RO. Thus no decrease in sensitivity of the radioactivity measurement is anticipated at the observed TSS concentrations.

There are some limitations in the use of gross alpha and gross beta particle activity as a screening tool for radioactive material. Firstly, it is only a procedure suited for general radiological monitoring, and thus the identification of individual radioactive species was not performed. Secondly, a screening level for alpha emitter particles would not represent a consistent health protective standard to cover all radionuclides of this class. For example, other less prevalent alpha particle emitting radionuclides (such as polonium-210 and radium-224) are not monitored under the established gross alpha screening method. Similarly a screening level for beta particle activity would not represent a consistent health protective standard that

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would cover all beta emitter radionuclides. In other words, the actual risk at the 0.5 Bq/L level depends on the particular mixture of isotopes and their cancer risk coefficients. However, if the screening level chosen was based on a conservative approach, using the radionuclides with highest risk coefficients, they would be sufficient to ensure public health protection. In addition, lifetime cancer risk depends on the time-averaged concentration in water, rather than specific high occurrences, as reflected by the maximum measured concentrations. Despite these limitations, the total radioactivity present in all analysed samples was low (below 0.5 Bq/L), especially for samples after the RO treatment. These results imply that, for potable supplies through augmentation using recycled water, the human exposure to radioactive compounds is likely to be negligible with a multiple barrier approach and advanced treatment.

CONCLUSION

Based on the results obtained for gross alpha and gross beta particle activity before and after the advanced treatment, no increased human health risk associated with radionuclides is anticipated if recycled water is used to augment drinking water supplies in Perth. It is recommended that quarterly screening for gross alpha and gross beta activity be undertaken during validation monitoring in the water reclamation plants. If the source of the activity is expected to be changing over time, then the sampling frequency should reflect this factor. If, after validation monitoring, there is no reason to anticipate that the source varies with time, then verification monitoring may be done every year. On the other hand, the sampling frequency should be maintained, or even increased, if concentrations are approaching the reference levels. Therefore, a vigilant source control and monitoring programme should be implemented to ensure concentrations of gross alpha and gross beta are within safe limits for human consumption.

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