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Dose assessment for marine biota and humans from discharge of ^{131}I to the marine environment and uptake by algae in Sydney, Australia

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A B S T R A C T

Iodine-131 reaches the marine environment through its excretion to the sewer by nuclear medicine patients followed by discharge through coastal and deepwater outfalls. ^{131}I has been detected in macroalgae, which bio-accumulate iodine, growing near the coastal outfall of Cronulla sewage treatment plant (STP) since 1995. During this study, ^{131}I levels in liquid effluent and sludge from three Sydney STPs as well as in macroalgae (*Ulva* sp. and *Ecklonia radiata*) growing near their shoreline outfalls were measured. Concentration factors of 176 for *Ulva* sp. and 526 for *E. radiata* were derived. Radiation dose rates to marine biota from ^{131}I discharged to coastal waters calculated using the ERICA dose assessment tool were below the ERICA screening level of 10 μGy/hr. Radiation dose rates to humans from immersion in seawater or consumption of *Ulva* sp. containing ^{131}I were three and two orders of magnitude below the IAEA screening level of 10 μSv/year, respectively.

1. Introduction

In Australia and globally, radioisotopes are routinely discharged to the environment following their uses in medicine, veterinary medicine, research and industry. Within nuclear medicine, which is an important industry in terms of its discharge of radioactivity to the marine environment, there are a wide range of radioisotopes that are used to diagnose and treat a variety of disorders. In general radioisotopes are chosen for use in medicine for their ease of detection, low radiotoxicity, short half-lives and appropriate chemical or biological behaviour (Titley et al., 2000). These radioisotopes will decay while in the body, but some of their activity will also be discharged to sewer as the patient excretes the radioisotope. The major radiopharmaceuticals used in the Sydney area include ^{51}Cr, ^{18}F, ^{125}I, ^{131}I, ^{89}Sr, ^{99m}Tc and ^{90}Y.

Iodine-131 is considered by the International Commission on Radiological Protection (ICRP) to be the most critical medical radionuclide in terms of the potential radiological dose it provides to medical staff, the public and their relatives following procedures involving therapeutic administration of unsealed radionuclides (ICRP, 2004). Iodine-131 is critical in part because dosages of ^{131}I used for treatment of thyroid cancers are typically the largest dosages administered in nuclear medicine (OSPAR Commission, 2002). It is also the most commonly used therapeutic radioisotope in Sydney (Davis, 2006). With a half-life (8.04 days) longer than the time it takes to be excreted from the body, transported to and treated at a sewage treatment plant (STP), ^{131}I is often still present when it reaches the marine environment and because the gamma radiation it emits is relatively high energy (364 keV) it is easy to detect and measure.

Iodine-131 is used for both diagnostic and therapeutic procedures. Diagnostic dosages range from 0.2 to 400 MBq and treatment dosages range from 50 MBq to 10 GBq, with a typical ablation dosage being approximately 7 GBq (ARPANSA, 2007; Davis, 2006; Titley et al., 2000). Not all of the ^{131}I administered to patients is excreted as some radioactive decay occurs while it is in the body. The percentage of, and rate at which, ^{131}I is excreted from the body is dependent upon the nature of the treatment. The levels of ^{131}I excreted decrease over time after treatment.

In Sydney, all STPs discharge their treated effluent to the marine environment either directly through coastal or deep ocean outfalls, or indirectly via outfalls into rivers. Some STPs also recycle some of their treated effluent as water for industrial, residential or municipal reuse (ARPANSA, 2007). Release of effluent containing radionuclides to the marine environment provides a potential route by which the public can be exposed to radionuclides. An investigation into the sources and fate of radioactive discharges to public sewers in the UK by Titley et al. (2000) found that the discharge of effluent

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containing radionuclides to the marine environment provides a higher radiological dose to members of the public than sludge containing radionuclides being re-used on agricultural land or disposed to landfill.

Marine algae are well known for their ability to concentrate iodine from seawater. One way to characterise the extent to which iodine is concentrated above ambient (seawater) concentrations is through use of a concentration factor (CF), which is the ratio of iodine in the algae over the concentration in the surrounding seawater. The recommended CF for iodine in macroalgae is 10 000 L/kg fresh weight (IAEA, 2004). Measuring the ^{131}I content of marine algae is a good way of monitoring ^{131}I levels in the local environment as it is concentrated to above ambient levels and continuously integrates changes in exposure. However, the use of CFs assumes the system in question to be at equilibrium and, since ^{131}I levels in algae are a net value from uptake, loss and radioactive decay of ^{131}I, this is not always the case. In addition, CFs for marine algae are affected by exposure time and algal condition. Thus, it can be difficult to infer ^{131}I concentrations in the environment surrounding algae by measuring its ^{131}I content (Druehl et al., 1988).

In this study we sought to gain an understanding of the relative contributions of direct sewage discharges from the Australian Nuclear Science and Technology Organisation ("ANSTO") and discharges from nuclear medicine facilities and outpatients, to the levels of ^{131}I found in effluent from Cronulla STP. Also, in order to determine whether there is any radiological risk to marine biota or humans from the current levels of ^{131}I discharged to the coastal marine environment in Sydney, we measured the levels of ^{131}I in effluent from three STPs in the Sydney region (including Cronulla STP) and in algae from their corresponding shoreline outfalls. Using these data, we performed an environmental dose assessment for marine biota living near the sewage outfalls, as well as a human dose assessment for people swimming or eating algae from the sewage outfalls.

2. Study area

The population of Sydney is approximately 4.3 million. Sales of ^{131}I to nuclear medicine facilities in the Sydney region are approximately 2500 GBq per year. On average, approximately 70% of this activity would be expected to be excreted based on biokinetic studies on people undergoing various ^{131}I medical procedures (Barrington et al., 1996; Hilditch et al., 1991; ICRP, 2004), contributing about 1750 GBq of ^{131}I to the sewer system each year. During 2007–2008, 427 GL of wastewater was discharged to the marine environment through ocean outfalls and approximately 42 600 dry tonnes of biosolids were produced, corresponding to a biosolids removal rate of about 10%. However, these statistics are dominated by the three large high rate primary treatment STPs with offshore deep ocean outfalls, not the smaller secondary and tertiary treatment STPs focussed on in this study (Sydney Water, 2008b).

There are many hospitals and clinics in Sydney that provide nuclear medicine services. Of these, there are 20 hospitals within Sydney Water's area of operations that administer therapeutic nuclear medicine (Davis, 2006). Therapeutic procedures often involve administration of large activities. Therefore, some nuclear medicine patients are required to remain in hospital for up to one week after treatment in order to prevent exposure of family members, caregivers or members of the public to any sizeable radiological dose (ICRP, 2004). The majority of the activity excreted by these patients therefore travels to the receiving STP of the hospital, although some is also excreted into alternate sewerage systems after the patient returns home. Sydney Water's current trade waste policy requires all nuclear medicine units to have holding tanks, in which the radioactive effluent from patients is stored and allowed to decay before it is discharged to the sewer. The required detention time of at least 16 days waste must be discharged to sewer at a controlled rate to achieve appropriate dilution.

Of the Sydney hospitals that provide nuclear medicine, 17 have their effluent treated by one of the three major Sydney STPs (Bondi, Malabar, North Head), all of which provide high flow rate primary treatment and have deepwater ocean outfalls between 2 and 4 km offshore (Sydney Water, 2008a). As this paper is investigating levels of ^{131}I in inter-tidal algae growing near shoreline ocean outfalls, these STPs are not discussed further here. Sutherland Hospital and a private nuclear medicine clinic (Southern Radiology) are the only providers of ^{131}I treatments within the Cronulla STP catchment. Neither of these nuclear medicine facilities provide inpatient dosages, therefore the activity they administer will not necessarily be received by Cronulla STP.

ANSTO discharges its treated liquid effluent to Cronulla STP and the Environmental Monitoring group at ANSTO has measured ^{131}I levels in algae from its ocean outfall at Potter Point since 1995 (Hoffmann et al., 1996, 1997, 1998, 1999, 2000, 2001; Hoffmann and Loosz, 2002; Hoffmann and Ferris, 2002; Hoffmann et al., 2003, 2004, 2005, 2006, 2008, see Section 5.3.1). For this reason, Cronulla STP and Potter Point ocean outfall were chosen as one of the study sites for this investigation. Two other STPs with shoreline outfalls of comparable size to Cronulla STP were also chosen for study and are shown in Table 1. Cronulla STP is the largest of the three STPs investigated and provides tertiary treatment, whereas STP A and STP B provide secondary treatment.

The location of ANSTO and its receiving STP at Cronulla are shown in Fig. 1.

3. Materials and methods

Treated sewage effluent and sludge from Cronulla STP, STP A and STP B, raw effluent from ANSTO and algae from Potter Point, outfall A and outfall B were collected and analysed for ^{131}I.

3.1. Sampling

3.1.1. Effluent and sludge

Iodine-131 treatments are administered irregularly and at most nuclear medicine clinics, infrequently. Hence, levels of ^{131}I in STP effluent vary widely. It was therefore necessary to collect samples over a period of time and calculate an average value to establish a representative ^{131}I concentration. The advantage of sludge sampling is that its longer residence time means a single sample provides a more long-term representation of ^{131}I levels in that particular STP. In addition, it can accumulate ^{131}I and thus demonstrate the presence of low levels of ^{131}I passing through an STP when they are too low to be detected in the effluent.

All effluent samples were collected in 1 L HDPE bottles from the final treated effluent stream so that levels of ^{131}I in the samples reflected what was discharged to the marine environment. Sludge samples were collected either in plastic or glass jars. Gloves were worn at all times when collecting and processing each sample type.

Six 1 L grab samples of liquid effluent from ANSTO holding tanks (HT, non-active trade waste) and three 1 L samples of liquid effluent decanted from ANSTO mixing tanks (MT, low level waste waters) were collected between 31/3/2008 and 17/4/2008.

From Cronulla STP, fourteen 1 L 24 h composite effluent samples, sampled hourly from 09:00 to 08:00, and two digested sludge samples were collected between 3/4/2008 and 17/4/2008. Eleven 1 L 24 h composite effluent samples, sampled hourly from 09:00 to 08:00 from Cronulla STP were also collected between

Table 1
Characteristics of Cronulla STP, STP A and STP B.

STP	Cronulla	A	B
Treatment level	Tertiary	Secondary	Secondary
Discharge volume (ML/day)	54	36	16
Discharge location	Potter Point (shoreline)	Outfall A (shoreline)	Outfall B (shoreline)
Effective population	200 000	140 000	59 000