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Detection and tracing of the medical radioisotope ^{131}I in the Canberra environment

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Abstract. The transport and radioecology of the therapeutical radioisotope ^{131}I has been studied in Canberra, Australia. The isotope has been detected in water samples and its activity quantified via characteristic γ-ray photo peaks. A comparison of measurements on samples from upstream and downstream of the Canberra waste water treatment plant shows that ^{131}I is discharged from the plant outflow into the local Molonglo river. This is consistent with observations in other urban environments. A time-correlation between the measured activities in the outflow and the therapeutical treatment cycle at the hospital identifies the medical treatment as the source of the isotope. Enhanced activity levels of ^{131}I have been measured for fish samples. This may permit conclusions on ^{131}I uptake by the biosphere. Due to the well-defined and intermittent input of ^{131}I into the sewage, the Canberra situation is ideally suited for radioecological studies. Furthermore, the ^{131}I activity may be applied in tracer studies of sewage transport to and through the treatment plant and as an indicator of outflow dilution following discharge to the environment.

1 Introduction

The radioisotope iodine-131 (^{131}I) with a radioactive half-life of $t_{1/2}|^{rad}$ = 8.02 d is routinely and successfully applied in the medical treatment of thyroid cancers. Typically activities of several GBq are given orally to patients. This significantly exceeds activities given in other nuclear medicine procedures. Patients are kept in hospital for several biological half-lives $t_{1/2}|^{bio}$ = 14 h, thus excreting most of this activity into the hospital sewage [1]. In many hospitals, in agreement with recommendations by the International Atomic Energy Agency [2], such waste is directly discharged into the public sewage system. Global practice varies considerably. In Europe, the maximally permitted release rate for ^{131}I ranges from 3.7 MBq/L in Greece to less than 5 Bq/L in Germany and Luxembourg [3].

Data on the pathways of ^{131}I into the environment and possible uptake by the biosphere are scarce, despite it being readily detectable via a characteristic γ-ray line. It bio-accumulates in algae [4,5] and the uptake by other *flora* and *fauna* may be expected. Geven *et al.* showed that fish, including carp, accumulate radioactive iodine isotopes in their active thyroid tissue [6].

In Canberra typically only a single patient is treated per week with ^{131}I at the Canberra Hospital. Consequently, the frequency of discharge into sewage and the attrition of the isotope due to radioactive decay are well matched. This may permit the tracing of individual discharges of ^{131}I through the sewage system and assist with the interpretation of measured ^{131}I activities in the outflow from the wastewater treatment plant. In addition, the well-calibrated, but sporadic nature of the input activities may permit ^{131}I to be used as a tracer of wastewater flow and dilution.

2 Study Area

Canberra has a population of approximately 350,000 people. Sewage is treated at the Lower Molonglo Water Quality Control Centre (LMWQCC) that is located on the western fringe of the city and it releases treated water into the Molonglo River, which is a subsidiary of the Murrumbidgee River. An activated sludge-type treatment of sewage is used, enabling shorter treatment times in comparison to alternative methods.

The wastewater treatment plant services all Canberra hospitals, however, nuclear medicine procedures are only conducted at the Canberra Hospital in Woden, at a distance of 20 km from the wastewater treatment plant.

Patients that undergo treatment with ^{131}I are admitted into hospital on Friday morning. During their treatment in hospital they are kept in an isolation room in the minor procedures ward to ensure the radiation safety protection of personnel and public. At approximately noon on Friday, the patients is given a calibrated dose of ^{131}I orally. The patient is then typically kept until Monday. The isolation room has a dedicated lavatory. Since almost five biological half-lives pass before the

patient is discharged, most of the ^{131}I given enters the sewage from this lavatory.

Figure 1 shows a schematic of the wastewater flow from the Hospital to the treatment plant and, following treatment, its discharge via the Molonglo and Murrumbidgee rivers. The sampling points used in this study are indicated. Due to their upstream location sampling points ② and ④ should show no evidence of ^{131}I.

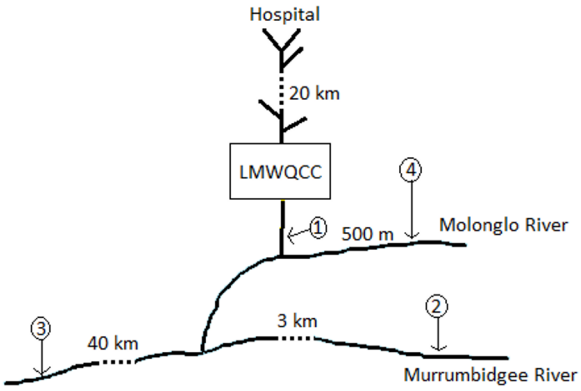

Fig. 1: Schematic illustration of the wastewater flow from the nuclear medicine unit at the Canberra Hospital to and through the waste water treatment plant (LMWQCC). The four sampling points and relevant distances are indicated.

3 Measurement Details

3.1 Water samples

Following initial sampling at all four points to establish feasibility, water samples were collected regularly over a period of 10 days at sampling point ①. Figure 2(a) displays a photo of this sampling point that is on the 30 m long outflow stream from the treatment plant.

At each collection 10 L of surface water were allowed to flow into HDPE containers. 1 L of this volume was decanted into a beaker and acidified with 40 mL of 5 molar H_2SO_4 to prevent the adhesion of particulates to container walls during preparation and measurement. The decanted volume was reduced by evaporation at 70°C over typically 6 hours to 50 mL. At this temperature the volatility of iodine can be avoided [7].

The remaining 50 mL were then decanted into a 80 mm diameter glass beaker with a 2 mm thick floor for γ-ray spectroscopy. A lead-shielded high purity germanium detector was used. The setup is shown as a cross-sectional drawing in figure 2(b). Consistent detection geometry was ensured. In order to fit the glass beakers, the lid of the lead shield was propped up.

3.2 Fish samples

Three european carp (*Cyprinus carpio carpio*) were caught from the Molonglo River at the point where the outflow stream of the treatment plant enters the river, about 20 m downstream of sampling point ①. An example is shown in figure 2(c). Two of these carp were caught during the 10 days when regular water samples were taken at point ①. The carp weighed between 2.0 and 2.2 kg and were between 43-48 cm long. As the carp's kidney thyroid tissues has been found to accumulate iodine [6], entrails taken from each fish were subjected to γ-ray spectroscopy.

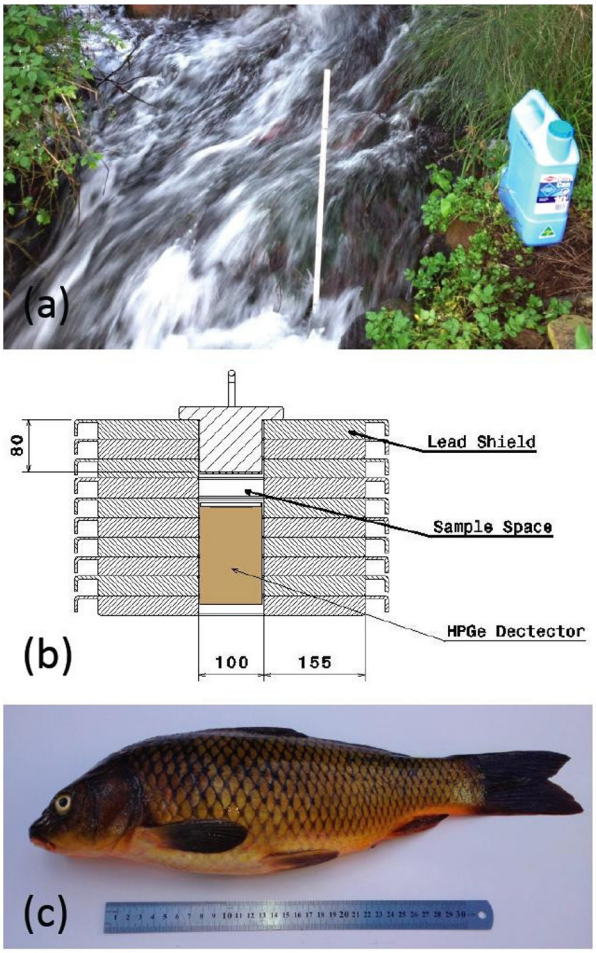

Fig. 2: (a) Photo of sampling point ① (white stick) on the treatment plant outflow stream indicating the flow conditions. (b) Cross-sectional drawing of the shielded γ-ray spectrometer used. Dimensions are given in mm. (c) European carp caught in the Molonglo river, where the outflow from the treatment plant meets the river.

3.3 Gamma-ray spectroscopy

Typically the γ-ray spectroscopy of a sample was performed over 24 hours. Figure 3 shows typical γ-ray energy spectra from these measurements over the energy range of 250 – 650 keV. Several background lines are present between 500 keV and 620 keV.

The radioisotope ^{131}I decays via β-decay and the characteristic 364.5 keV γ-ray line in its daughter can be used to identify it in the spectrum. γ-ray lines at 284.3 keV and 637.0 keV are also characteristic for the isotope, however, less intense. The 364.5 keV line is absent in figure 3(a), however, dominates the spectrum over the background lines in figure 3(b) and (c).

The energy spectra were calibrated using several known radioactive sources and a linear calibration could be achieved. The photo peak intensity of the 364.5 keV line was integrated and the background subtracted.

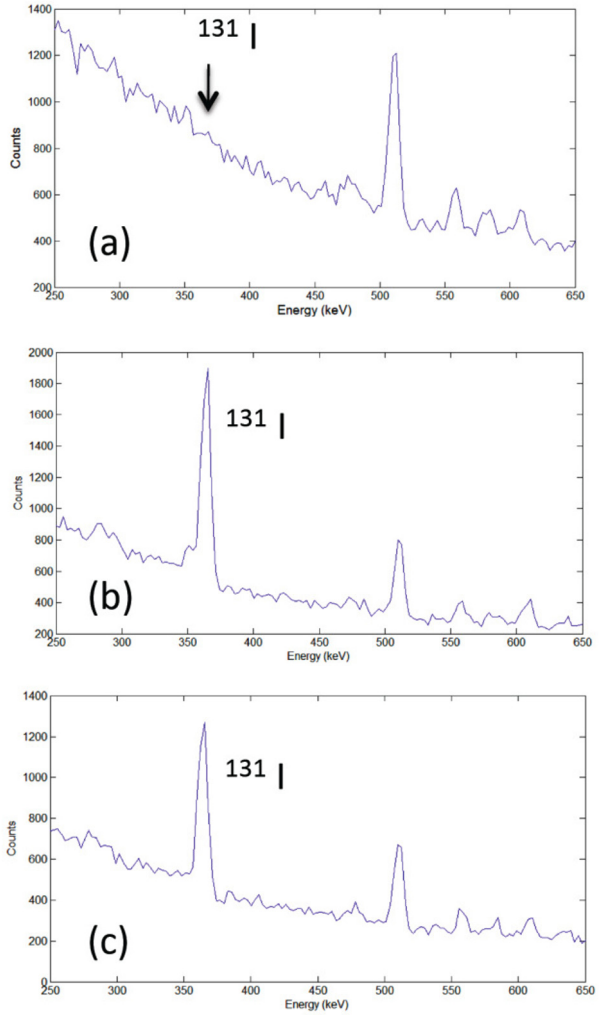

Fig. 3: Gamma-ray spectroscopy of water samples from sampling point ① on the outflow from the treatment plant. The detected number of counts are plotted versus γ-ray energy. (a) Sample taken before an ^{131}I therapy session at the hospital. (b) Sample taken about 2 days after a therapeutic dose was given (c) Sample taken 3 days after a dose was given.

4 Results and Discussion

As expected, water samples from upstream from the Murrumbidgee river (point ②) and the Molonglo river (point ④) did not show any detectable signal of ^{131}I. In the sample taken at point ③ from the Murrumbidgee river also no ^{131}I was detected. Given that this sampling point is 40 km downstream from the wastewater treatment plant, this is not a surprising result.

In contrast, the levels of ^{131}I detected in the outflow water at point ① varied greatly over the 10 days of regular sampling, as it is apparent from figure 3. During this period two ^{131}I therapies where performed at the Hospital on consecutive Fridays. The respective doses were 2.22 GBq and 5.55 GBq. The times of therapy are indicated in figure 4. In the same figure the measured activity concentrations for the water samples from the outflow of the treatment plant are displayed. For each sample the detected number of counts was corrected for the radioactive decay of ^{131}I between sampling and the time of measurement. Results have also been normalised in regard to measurement time and sample volume.

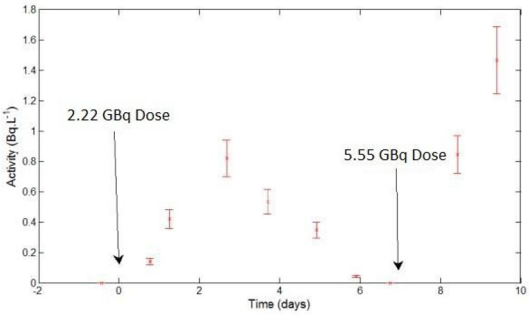

Fig. 4: The activity concentration of water samples collected at the outflow ①, corrected for radioactive decay as described in the text. The times and doses delivered during ^{131}I therapy sessions at the hospital are also indicated.

The detected activities for ^{131}I are correlated to the application of the radioisotope at the hospital in both, time and magnitude. On the day before the first treatment no activity was measured. A delay of about 2 days follows. Likely due to the combination of biological half-life and sewage transport to and through the wastewater treatment plant, before the detected activity peaks. Discharge into the river system and attrition due to radioactive decay then reduces the detected activity over about three days to zero. A second increase in detected activity, with the same observed time-delay, occurs following the next application of the radioisotope at the hospital.

The observed correlation between measurement and therapy at the hospital identifies the latter as the source of the ^{131}I in the outflow water. The well-separated spikes of ^{131}I activity in Canberra sewage should permit detailed studies of the pathway of the isotope through wastewater treatment. Furthermore, the isotope may be employed as tracer of sewage transport in Canberra and indicate the efficacy of sewage dilution in the river system on discharge from the treatment plant.

A gamma-ray spectra for the entrails of one of the carp samples are shown in Fig. 5(a). The detected activity associated with ^{131}I has been found to be elevated above those measured for the samples of the outflow water when normalised by weight. The detected levels are (9 ± 1) Bqkg^{-1} and (11 ± 2) Bqkg^{-1}, respectively. This is almost an order of magnitude larger than the maximum detected specific activity in the outflow water. This is consistent with other studies that concludes that ^{131}I is bio-concentrated by carp with a very high assimilation rate [8].

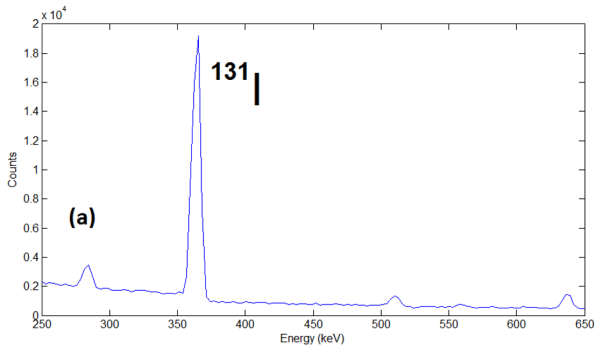

Fig. 5: Gamma-ray spectroscopy of one of the fish samples from sampling point ①. The detected number of counts are plotted versus γ-ray energy. Note that the higher ^{131}I levels allow the 284.3 and 637.0 keV lines to be identified in addition to the 364.5 keV photo peak.

.5 Conclusions

The radioecology of the therapeutic isotope ^{131}I is important, since it is applied in nuclear medicine at very high activities. In line with international radiation protection considerations [2,9], this radioactivity is generally being discharged with normal sewage. In Canberra, Australia, the discharge and transport of the isotope through the sewage system has never been studied before. Only a small number of studies from other urban environments exist. Importantly, the particular sewage treatment procedures in Canberra allow for a relatively fast transit of water through the plant.

This work has shown that levels of ^{131}I in Canberra sewage can be detected and quantified at the outflow of the sewage treatment plant. The data are highly suggestive of a strong correlation in time and magnitude between the detection and the application at the nuclear medicine unit of the hospital identifies the therapeutical use as the source of the radioactivity. This is likely the only source of the isotope.

Due to the infrequent application in nuclear medicine and the relatively short radioactive half-life of the isotope, the ^{131}I activity in Canberra sewage rises and falls sharply. These spikes of activity may successfully be used in tracer experiments that may indicate the transport and dilution efficacy of the sewage treatment procedures applied. Results might inform sewage treatment solutions in other locations.

Enhanced activity levels of ^{131}I have been measured for fish samples. This is in agreement with other work and may have relevance, when the exposure of the biosphere due artificial ionising radiation is considered. Further work is underway that will include the determination of absolute sample activities and detailed studies of uptake by wildlife and plants. The dilution of sewage will be studied by conducting measurements at short distance intervals along the outflow stream from the treatment plant and along the Molonglo river.

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