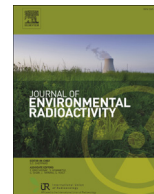




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Robustness testing of an in-situ caesium extraction unit

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ABSTRACT

Following on from the successful development and validation of a prototype automated unit for the extraction of radiocaesium from seawater, the unit was redesigned to be smaller, easier to transport and better suited for field work. An extension of the validation process was undertaken to include robustness testing. Robustness testing evaluates the behaviour of a procedure when deliberate small changes are made. This requires the identification and testing of experimental factors which could have an impact on the results. The Plackett-Burman approach was adopted to minimise the number of experiments needed for testing. The study showed that the analytical method was not sensitive to any of the factors tested, indicating that the method is robust.

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1. Introduction

Anthropogenic radionuclides, including radiocaesium (^{134}Cs and ^{137}Cs) can enter the aquatic environment following atmospheric nuclear weapons detonations or as a result of accidents in nuclear power plants or nuclear powered vessels. Following the accident at the Fukushima Dai-ichi nuclear power plants in 2011, a large amount of radiocaesium was released to nearby oceans. This was via direct releases of radiocaesium to coastal waters and atmospheric releases and subsequent deposition to the surface of ocean waters. Ocean modelling predicted that following their release, these radionuclides would reach northern Australian waters sometime between 2016 and 2021 in substantially diluted levels (Povinec et al., 2013). In addition, as part of preparedness planning for nuclear powered warship visits, Australian authorities have a requirement to measure radionuclides in ports which are visited by these vessels (Department of Defence, 2003). Periodic monitoring of coastal waters is therefore necessary to understand background levels and to reassure the public that there are no impacts on the health of the public and the environment as a result of nuclear powered warship visits. This required the development of an accurate method for determining radiocaesium levels in seawater, especially at very low levels.

Current radiocaesium background levels in Australian waters are approximately 1 mBq/L (Bokor et al., 2016) and the quantitative determination of radiocaesium at these low levels requires substantial pre-concentration of the sample prior to measurement. The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) successfully developed an automated Caesium Extraction Unit (CEU) for the in-situ extraction of radiocaesium, using chemically coated filter cartridges, from 1000 L of seawater. The cartridges are then treated in the laboratory to enable quantification of the radiocaesium.

The coating material on the filter cartridges is potassium copper hexacyanoferrate, which is prepared by mixing 5 g of potassium hexacyanoferrate ($\text{K}_4\text{Fe}(\text{CN})_6$) and 100 mL of copper nitrate solution (50 g/L $\text{Cu}(\text{NO}_3)_2$) in 5 L of water. Two of these cartridges are placed in series after a 10 μm and a 1 μm pre-filter. Following the extraction, the coated cartridges are dried and ashed. The ash is placed into small petri dishes for counting by high resolution gamma spectrometry. The procedure that was developed and allowed the accurate quantification of radiocaesium with a detection limit of 0.2 mBq/L is detailed in Bokor et al., (2016).

Placing the two coated filter cartridges in series allows the cartridge extraction efficiency (E) to be determined (Equation (1)), which is then used to calculate the activity concentration (C) of the sample (Equation (2)). Calculation of the efficiency is based on the assumption that both cartridges have the same extraction capacity.

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$$E(\%) = \frac{A_1 - A_2}{A_1} \times 100 \quad (1)$$

where:

E = cartridge extraction efficiency
 A_1 = activity in the 1st coated cartridge (Bq)
 A_2 = activity in the 2nd coated cartridge (Bq)

The activity concentration of radiocaesium in the seawater (Bq/L) can be calculated using the following formula:

$$C(\text{mBq/L}) = \frac{A_1}{V \times (E \times 0.01)} \times 1000 \quad (2)$$

where:

V = sample volume (L)

While validation characteristics, including selectivity, precision, trueness, linearity, limit of detection and traceability were previously investigated and reported (Bokor et al., 2016) and indicated that the method was reliable and fit for purpose, the original unit required redesign to make it smaller, lighter and more suitable for fieldwork (Fig. 1 and Fig. 2). Following the redesign of the CEU, a test for robustness, which would provide additional information and complete the validation process, was undertaken and is reported in this study. Robustness testing, as defined by Burns et al., (2009), examines the performance of the method when small deliberate changes are made to the parameters/factors. This testing is sometimes referred to as “ruggedness testing” (Magnusson and Ornamark, 2014).

The first step in robustness testing is to identify which factors may have an effect on the result. To test the variation in every factor individually, would require many experiments. An alternative is to use a multifactorial approach as described by Plackett and Burman, (1946). In this study seven factors were investigated at two levels e.g. high (+) and low (–), which according to the Plackett and Burman design requires eight experiments (AMCTB, 2013). Each factor will have equal number of high (+) and low (–) levels within one experiment, such that the overall effect of the factors can be calculated (Plackett and Burman, 1946; Paule et al., 1986).



Fig. 2. Size comparison of the original unit (left) with the redesigned one (right).

2. Experimental

The factors that may have an influence on the experimental results were identified and are listed in Table 1 along with their nominal values. The factors investigated were; the concentration of the solutions for preparing the coated cartridges (F1 and F2), environmental factors: salinity (F3) and temperature (F4), instrumental factor: flow rate (F5), ashing temperature (F6) and packing density (F7).

Knowledge of the expected variation in the factors was used to determine the low and high deviations from the nominal values. The variability of the environmental factors; salinity and temperature, were based on the expected range in Australian coastal waters. For the robustness experiments, the nominal values were varied by a set amount as shown in Table 1. Careful consideration was given to the experimental design to ensure that the adjustments to the factors could be achieved.

For this study 100 L of filtered seawater, collected at Newport in Port Phillip Bay, was used for each experiment. This was spiked with ^{137}Cs , to an activity concentration of 0.50 ± 0.01 Bq/L, to minimise the counting uncertainties arising from gamma measurements.

3. Results and discussion

3.1. Calculation and interpretation of effects

The design, results and calculated effects of the experiments are shown in Table 2, with the + and – denoting the higher and lower deviations from the nominal values.

The effect, E_x is calculated using Equation (3).

$$E_x = \frac{\sum Y(+)}{N/2} - \frac{\sum Y(-)}{N/2} \quad (3)$$

where:

E_x = Effect for factor (x) on the responses Y(+ or –)
 $\sum Y(+)$ and $\sum Y(-)$ are the sums of the responses Y
 N = number of total experiments, N = 8.

To identify important factors, the half-normal probability plot (Daniel, 1959) was used, where Absolute Effects ($|E_x|$) are plotted in an increasing order on the y axis with values of Rankit on the x axis. Rankit values are calculated using a statistical tool, based on the



Fig. 1. The redesigned unit showing the configuration of the filter housings.

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