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Environmental radionuclide monitoring of Canadian harbours: a decade of analyses in support of due diligence activities by the Royal Canadian Navy

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ABSTRACT

The Royal Canadian Navy has conducted a comprehensive programme of safety, security and environmental monitoring since the first visits of nuclear powered and nuclear capable vessels (NPV/NCVs) to Canadian harbours in the late 1960s. The outcomes of baseline monitoring and vessel visit sampling for the period 2003–2012 are described for vessel visits to Halifax (NS), Esquimalt (BC) and Nanoose (BC). Data were obtained by gamma-ray spectroscopy using high purity germanium detectors. No evidence was found for the release of radioactive fission or activation products by NCV/NPVs during the study period, although anthropogenically produced radionuclides were observed as part of the study's baseline monitoring. Background activities of Cs-137 can be observed in sediments from all three locations which are derived from well-documented radioactivity releases. The detection of I-131 in aquatic plants is consistently observed in Halifax at activities as high as 15,000 Bq kg^{-1} dry weight. These data are tentatively assigned to the release of medical I-131, followed by bioaccumulation from seawater. I-131 was also observed in aquatic plants samples from Esquimalt (33 Bq kg^{-1}) and Nanoose (20 Bq kg^{-1}) for a single sampling following the Fukushima Daiichi accident.

1. Introduction

Following a process of safety evaluation and review, the first nuclear powered/nuclear capable vessels (NCV/NPV), from the United States and Britain, entered Canadian ports in 1967. In subsequent years, several further environmental evaluations and technical safety assessments have been conducted. Initial and subsequent assessments have concluded that the environmental risk associated with NCV/NPV visits is low. A comprehensive baseline study of environmental matrices and foodstuffs was performed in 1996 (Waller and Cole, 1999). The Royal Canadian Navy (RCN) continues to develop and refine a Nuclear Vessel Visit Safety Programme (NVVSP). The latter document, which is updated annually, is based on unclassified information and open discussion, although authorisation for external distribution resides with the RCN Nuclear Safety Officer. Central to the NVVSP are two Nuclear Emergency Response (NER) teams based in Halifax, Nova Scotia and in Esquimalt, British Columbia; the latter covering both CFB Esquimalt and Canadian Forces Maritime Experimental and Test Range (CFMETR) in Nanoose, British Columbia. NER teams are trained to respond to emergencies, but also conduct routine radiation monitoring during NCV/NPV visits. These teams take environmental samples during NCV/NPV vessel visits and obtain baseline environmental samples according to a schedule defined in the NVVSP. Seawater samples are taken during NPV/NCV visits as part of a Visit Specific Strategy (VSS). Baseline sampling consists of seawater, sediment, aquatic plants and sea-life to form a Continuous Sampling Strategy (CSS) programme. The collection, analysis and reporting of these samples form the RCN Environmental Radionuclide Monitoring Programme (ERMP). All ERMP samples are analysed by gamma-ray spectroscopy using high purity germanium (HPGe) detectors at the Analytical Sciences Group (ASG) at the Royal Military College of Canada (RMCC) (Kingston, ON). To provide analytical resources on each coast, seawater samples are also analysed using identical instrumentation at the Department of Occupational Health, Safety and Environment, University of Victoria (Victoria, BC) and the Dockyard Laboratories of Defense Research and Development Canada (Halifax, NS). All three laboratories are accredited to the ISO 17025 standard by the Canadian Association for Laboratory Accreditation (CALA) for ERMP

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analyses. The purpose, structure and operation of the ERMP, as well as the results of samples obtained during 2002 and 2003, have been previously reported (Nielsen et al., 2007). The present work will briefly describe and update ERMP structure and operation, whilst focussing primarily on VSS and CSS data obtained during the period 2003–2012.

The safety record of NPV/NCVs is extremely high. Releases of radioactivity from NATO vessels are almost unknown. Even following the sinking of the Russian nuclear-powered submarine, Kursk, in 2000, radioactivity measurements of the submerged vessel in 2002 and during recovery in 2003 showed no significant release of radioactivity (Amundsen et al., 2002; Baranova et al., 2003). However, public concern is demonstrated by the media attention given to a leak from the nuclear powered submarine, USS Houston, in 2006–2008. The total loss of radioactive material from this vessel over a two year period was described as being comparable with the activity of a domestic smoke alarm. However, the vessel had visited a number of ports in both the US and Japan, and the disclosure stimulated public concern (CNN, 2008). In contrast, medical radioisotopes do not seem to be of equivalent concern to the general public. A number of studies have identified medical radionuclides in sewage sludge, wastewater and surface water. Recent studies in the US (Hay et al., 2011; Rose et al., 2012, 2013), Europe (Fischer et al., 2009; Krawczyk et al., 2013) and Australia (Veliscek Carolan et al. 2011), all quantify medical I-131 in the environment, whilst some of these studies also identify the presence of other medical isotopes, such as Ga-67, Tc-99m, In-111 or impurities such as Mo-99, Ru-103 and Te-123m. The activities of I-131 measured in effluent routinely exceed 1 Bq dm^{-3}. The short-half-lives and low environmental activities of these radioisotopes present little risk and cause no public concern. However, it is essential that appropriate baseline and monitoring studies are made to distinguish such medical radioisotopes from potential releases associated with NPV/NPV visits, since the misinterpretation of such data could stimulate unfounded public concern. The ERMP represents such a Canadian activity.

2. Experimental

Sampling has previously been described in detail (Nielsen et al., 2007). The CSS sampling strategy comprises the analysis of seawater, sediment, plant and sea-life samples from NPV/NCV visit locations in Halifax, Esquimalt and Nanoose (Table S1 and Figures S1–S3). The numbers of samples per annum are; seawater (12), sediment (4), plant (4) and sea-life (2) with the specific sampling frequencies per location being described (Table S2). Sampling locations are defined by the size and tidal characteristics of the harbours and vary in number between three and eight. The sampling programme cycles through these locations. All samples are grab samples with sediment, plants and sea-life being obtained by RCN divers. Plant and sea-life samples are representative of local flora and fauna, but not restricted to specific species. Typical inter-tidal seaweeds, such as *Ascophyllum nodosum*, *Saccharina latissima* and *Fucus garneri* are obtained. The majority of sea-life samples consist of readily obtained crab and starfish species. VSS seawater samples are obtained immediately prior to NPV/NCV visits, then daily at high tide, and finally on departure. Samples are collected in 1 dm^3 polyethylene bottles and are transferred to ASG, RMCC for analysis under chain of custody. Duplicate VSS and CSS seawater samples are also transferred to their respective coastal laboratories for analysis. Quality control procedures require that at least 10% of samples are obtained in duplicate for each laboratory. Samples are stored at 4 °C during shipping and prior to analysis. Analytical procedures conform to ISO 17025. Seawater is analysed as received using a 0.45 dm^3 sample. Other matrices are homogenised before the analysis of a 0.20 dm^3 sample. A separate sub-sample is prepared to provide maximum surface area and analysed for moisture content by drying to constant mass in a stream of dry air at ambient temperature (sea-life and plants) or at 110 °C (sediment). All data are reported in Bq kg^{-1} as time of sampling activity. All data, with the exception of seawater, are presented as dry weight values.

Analyses were conducted until 2009 using HPGe detectors of 20% efficiency (Ortec GMX, Ortec, Oakridge, TN) with in-house constructed lead shielding (15.3 cm thickness) lined with 0.64 cm thickness oxygen-free high conductivity copper and 0.32 cm thickness plexiglass. Distances from the centre of the germanium crystal to shield were 17.8–19.1 cm. These systems were superseded in 2009 by 40% efficiency GMX40P4-70-S systems from the same vendor using commercial lead shielding with a tin/copper liner (Ortec, HPLBS1). Species-specific analyses of I-131 in plants were conducted using a 60% efficiency detector (GEM60P4, Ortec). Calibration standards were obtained from AEA (Nielsen et al., 2007), and subsequently from 2006 onwards from Eckert and Ziegler (Atlanta, GA). Control standards were obtained from Isotope Products (Valencia, CA) and latterly from the same laboratory as part of Eckert and Ziegler. Until 2006 nine radionuclides (Co-57, Co-60, Sr-85, Y-88, Cd-109, Sn-113, Cs-137, Ce-139, Hg-203) were used for calibration and control, after which Pb-210 and Am-241 were added to improved efficiency calibration for low energy gamma-rays. Quality control procedures require the analysis of laboratory blanks, typically tap water, and at least 10% of field samples are analyses in duplicate by the laboratories. Analyses are accredited to the ISO 17025 standard for 26 activation and fission products, although data are examined using extensive libraries of fission, activation and medical radionuclides. Typically, data for Co-60, Cs-137 and I-131 are reported, but all data above detection limit would be reported to the RCN. Such data form part of annual reports which are available to the public.

3. Results and discussion

The combination of CSS and VSS sampling protocols results in the analysis of more than one hundred samples per annum, along with associated blank, control and duplicate data. For the 26 accredited radionuclides considered (Table 1) over the ten-year reporting period, the data matrix becomes excessively large. Moreover, given the due diligence nature of the programme, the reporting of such data are not useful. Thus, it can be stated that during the period of interest (2003–2012) no radionuclides have been observed above detection limits for any seawater samples analysed as part of the CSS programme. Detection limits are subject

Table 1
Radionuclides studied in the present work.

Element	Isotopes
Barium	Ba-140
Cerium	Ce-144
Caesium	Cs-134, Cs-136, Cs-137, Cs-138
Cobalt	Co-60
Iodine	I-131, I-132, I-133, I-134, I-135
Lanthanum	La-140
Molybdenum	Mo-99
Niobium	Nb-95
Rubidium	Rb-86, Rb-88
Ruthenium	Ru-103, Ru-106
Strontium	Sr-91
Tellurium	Te-129m, Te131m, Te-132
Yttrium	Y-90m, Y-91m
Zirconium	Zr-95