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Radionuclide observables of underwater nuclear explosive tests

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A B S T R A C T

There remain technical challenges for an On-site Inspection (OSI) in the high seas environment, which gathers evidence of a violation of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). For terrestrial nuclear explosions, the radionuclide observables are well defined and States Parties have chosen 17 particulate radionuclides that allow discrimination from other nuclear events. However, an underwater nuclear explosion generates induced radionuclides from the neutron activation of seawater, which has the potential to interfere with the measurement of the radionuclide observables using gamma-spectrometry techniques. To understand these effects the inventory of OSI relevant (6.0×10^{16} Bq) and activation (1.6×10^{19} Bq) radionuclides has been calculated for a 1 kT underwater nuclear explosion. The activation products consist predominantly of ^{38}Cl and ^{24}Na, which decay to 5.56% and 0.0007% of their initial activity within 1 and 14 days. Monte Carlo techniques have been used to assess spectral interferences within this timeframe. It is demonstrated that during this period they do not interfere with the measurement of the existing radionuclide observables. Additionally, ^{24}Na has been identified as useful for inspection purposes.

1. Introduction

1.1. On-site inspection

An On-site Inspection (OSI) is a key aspect of the verification regime of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), which bans all nuclear explosions, whether for military, civil or any other purposes (UN, 1996). As conditions warrant, an OSI may be conducted in underground, underwater, sub-seafloor and atmospheric inspection environments (CTBTO, 2013), with the object of verifying whether a nuclear explosion has occurred in violation of the Treaty and to gather facts which might assist in identifying any possible violator. While the Treaty has not yet entered into force, and remains in provisional operations, primary emphasis has been on developing the OSI capability for an underground nuclear explosion (UNE) scenario (Burnett and Davies, 2015; Burnett and Milbrath, 2016; Burnett et al., 2016, 2017; Zucca, 2014). There remain technical challenges to undertaking an OSI in the underwater, or high seas environment. Among these, and the focus of this research, are the radionuclide observables suitable for OSI measurement that provide discrimination of an underwater nuclear explosion (often referred to as an UNDEX or underwater explosion) from other radiation sources, such as reactor incidents or releases, nuclear waste and medical isotopes. These have been defined for the UNE terrestrial scenario, as 17 particulate radionuclides (^{95}Zr, ^{95}Nb, ^{99}Mo, ^{99m}Tc, ^{103}Ru, ^{106}Rh, ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{144}Pr and ^{147}Nd) and four gaseous radionuclides (^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{37}Ar). These radionuclides are predominantly fission products, with the exception of ^{134}Cs and ^{37}Ar produced by neutron capture of ^{133}Cs and ^{40}Ca. Neutron activation becomes more important in the underwater environment, as seawater contains dissolved amounts of the majority of the 92 naturally occurring elements (Wright and Colling, 1995). This creates radioactive species that have potential to interfere with the measurement of the radionuclide observables.

1.2. Underwater nuclear explosions

Of the 2057 nuclear explosive tests undertaken during 1945–2017, only 8 were conducted underwater during 1946–1962 (Table 1). This included 5 by the United States in the Pacific Ocean and 3 by the former Soviet Union in the Arctic Ocean (close to Novaya Zemlya). There are characteristic phenomena associated with an underwater nuclear explosion that depend on yield, device detonation and water depth, and the area of the water body. When a nuclear weapon explodes underwater, energy is transferred to the surrounding water mass by radiation. This causes superheating of the seawater, resulting in expansion and the formation of a shockwave. The shockwave propagates outward transferring approximately half of the explosion energy and leaving behind a bubble of steam at high pressure and temperature (Pritchett, 1971b). The bubble rises due to buoyancy, and expands until its pressure is below the ambient pressure (over expansion), and then contracts,

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Table 1
Historic underwater nuclear explosions.

Test name	Country	Date	Bomb depth (m)	Yield (kT)
Baker	USA	25-Jul-46	27	20
Wigwam	USA	14-May-55	610	30
22 (Joe 17)	USSR	21-Sep-55	10	3.5
48	USSR	10-Oct-57	30	10
Wahoo	USA	16-May-58	150	9
Umbrella	USA	8-Jun-58	50	9
122 (Korall-1)	USSR	23-Oct-61	20	4.8
Swordfish	USA	11-May-62	198	< 20

thereby increasing its pressure and causing condensation. This process continues at a decreasing rate until the bubble erupts from the surface, releasing mostly gaseous fission products (of which iodine is the most abundant), and hurtling large masses of water aloft as the plume (Pritchett, 1971a). When the plume water falls back to the surface a radioactive cloud called the base surge is produced (Dolan, 1972). This travels outwards and is typically highly radioactive (Weary et al., 1981). This is followed by late-time explosion effects, such as residual upwelling along the explosion axis and turbulent diffusion of the radioactive surface pool. The explosive process typically results in three sources of radioactive contamination: airborne activity, residual fallout and water contamination. Notably, during the Wigwam nuclear explosion only 1% of the radioactivity was present in the airborne fraction with the potential to become fallout (Weary et al., 1981). This value is in agreement with observations for other underwater nuclear explosions (Lee, 1979).

2. Methodology

2.1. OSI relevant radionuclides

The OSI relevant radionuclides are produced from a nuclear explosion either as fission or activation products (Table 2). They can provide discrimination from different nuclear sources, especially when used in combination, and in the instance of $^{137}Cs/^{134}Cs$ for reactor accidents. This is because ^{134}Cs is produced predominantly by neutron capture of ^{133}Cs ($^{133}Cs(n, \gamma)^{134}Cs$) due to a low fission yield (0.0000279% per fission). Thus nuclear explosion debris samples would be expected to have a low $^{137}Cs/^{134}Cs$ ratio. Other ratios are also useful (e.g. $^{95}Zr/^{95}Nb$, $^{99}Mo/^{99m}Tc$) and can provide timing information relating to the explosion (Harms et al., 2009; Nir-El, 2006; Yamba et al., 2016). The list of radionuclides was chosen so as to encompass an OSI timeframe from one week to two years after an alleged nuclear explosion, and be measurable in field conditions with no common or otherwise expected interferences that could allow misidentification. It is the latter requirement that is problematic in the underwater environment, where significant quantities of induced radionuclides may also be produced by neutron activation of the seawater. Whilst many of these elements also occur in terrestrial soils and rocks, their concentration (salinity) is typically an order of magnitude less than seawater, such that fewer activation products are generated.

The initial inventory of OSI relevant radionuclides can be calculated using fission product yield data (England and Rider, 1994) in the manner described by the authors' previous research (Burnett et al., 2018). This depends on the yield, which for a fission-based explosion, is the amount of explosive energy (in trinitrotoluene equivalents) discharged from the detonation of fissionable nuclear material (e.g. ^{235}U, ^{239}Pu). As one kT is equivalent to 1.45×10^{23} fissions, and if the fission probability of a particular fission product is f, and its decay constant is $\lambda\ s^{-1}$, the activity following a 1 kT nuclear explosion is $1.45 \times 10^{23}\ f\lambda$ Bq (Chamberlain, 2004). As the relevant radionuclides are produced directly by fission and indirectly by the decay of other fission products, a computer program called RadICalc (Radiation Intensity Calculator) was used for inventory calculations (Robinson et al., 2015). This computes initial radionuclide activities for all fission products (using independent fission yield) and undertakes complex decay and in-growth calculations using the Bateman's Laplace transform-based solution to the differential equations for radioactive decay (Bateman, 1910). RadICalc utilises nuclear data (version B-VII.1 for ^{239}Pu pooled fast neutron fission decay) from the evaluated nuclear data file (ENDF) hosted by Brookhaven National Laboratory (NNDC, 2018). The exception to this approach, was the activation produced ^{134}Cs, which can be derived by direct linear scaling of reported nuclear explosive test values (Battelle, 1972; IAEA, 2008). Also consistent with previous research (Burnett and Davies, 2015; Burnett and Milbrath, 2016; Burnett et al., 2016, 2017), a ^{239}Pu fission-based nuclear explosion with explosive yield of 1 kT was modelled.

2.2. Seawater activation

Prompt neutrons and gamma-rays produced by an underwater nuclear explosion, are rapidly attenuated by the surrounding seawater and typically travel < 1 m (Weary et al., 1981). Water molecules (H_2O) do not activate, as neutron capture of hydrogen (^1H) and oxygen (^{16}O) produces stable isotopes of deuterium (^2H) and ^{17}O. On the short-time scales of an underwater detonation, there is relatively little double capture to form radioactive tritium (^3H) commonly associated with

Table 2
The OSI relevant radionuclides. Each radionuclide is categorized as volatile (V), refractory (R) or intermediate (V-R). The fission product yields are given per 100 fissions for ^{239}Pu pooled fast neutron fission decay.

Nuclide	Half-life	E (keV)	I (%)	Ind. Yield	Cum. Yield	V or R	Activity
^{95}Zr	64.03 d	756.7	54.4	9.30×10^{-2}	4.67	R	1.8×10^{13}
^{95}Nb	34.99 d	765.8	99.8	1.92×10^{-4}	4.67	R	6.4×10^{10}
^{99}Mo	2.75 d	739.5	12.1	1.31×10^{-2}	5.98	R	5.6×10^{13}
^{99m}Tc	6.0 h	140.5	89.1	1.49×10^{-6}	5.26	R	6.9×10^{10}
^{103}Ru	39.26 d	497.1	91.0	1.29×10^{-3}	6.83	V	3.8×10^{11}
^{106}Rh	373.61 d	621.9	9.9	3.85×10^{-1}	4.36	V	3.7×10^{16}
^{131}I	8.02 d	364.5	81.7	2.01×10^{-2}	3.88	V	2.9×10^{13}
^{132}Te	3.20 d	228.2	88.0	3.00	5.15	V	1.1×10^{16}
^{132}I	2.30 h	667.7	98.7	1.74×10^{-1}	5.33	V	1.1×10^{16}
^{134}Cs	2.07 y	604.7	97.6	Activation	Activation	V	6.1×10^{11}
^{137}Cs	30.08 y	661.7	85.1	1.00	6.58	V	1.1×10^{12}
^{140}Ba	12.75 d	537.3	24.4	9.17×10^{-1}	5.32	V-R	1.1×10^{15}
^{140}La	1.68 d	1596.2	95.4	9.87×10^{-3}	5.33	V-R	6.9×10^{13}
^{141}Ce	32.51 d	145.4	48.3	2.46×10^{-4}	5.15	V-R	8.8×10^{10}
^{144}Ce	284.95 d	133.5	11.1	1.64×10^{-1}	3.67	R	7.3×10^{12}
^{144}Pr	17.28 m	696.5	1.3	5.10×10^{-5}	3.67	R	4.9×10^{13}
^{147}Nd	10.98 d	91.1	27.9	1.72×10^{-3}	1.99	R	1.8×10^{12}