#### Marine Environmental Depleted Uranium Survey Report Kirkcudbright Training Area – 2007

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## **Executive summary**

Depleted uranium (DU) ammunition has been test fired at the Kirkcudbright Training Area (KTA) since 1982. Routine environmental monitoring has been carried out at KTA since 1980 to assess the environmental impact of the firings on the terrestrial and marine environments. Results of these surveys are published in regular reports.

This report describes and interprets the results of the marine environmental survey undertaken to monitor DU levels in the environment at KTA in 2007. Samples of intertidal sediment, seaweed and seafood were collected from the shoreline in the local area. Underwater sediment samples were collected offshore from the KTA, and samples of locally fished, bottom dwelling crustaceans and molluscs were also obtained.

The results of the 2007 survey agree with those from previous years and do not indicate any health or environmental impact from the DU firings. There is no evidence to indicate that members of the public or site employees are, or have ever been, exposed to a radiological hazard from the marine environment as a result of test firing DU ammunition at the site.

## **Table of contents**

Exect	utive summary	1
List o	of figures	4
List o	of tables	5
1	Introduction	6
2	Background	6
3	Depleted uranium	7
4	Differentiating DU from natural uranium	8
5	Investigation/action levels	10
6	Sample collection	10
6.1	Sampling locations and dates of sampling	10
6.2	Intertidal sediment sampling methodology	11
6.3	Seaweed collection methodology	12
6.4	Biota collection methodology	12
6.5	Dose rate measurement methodology	12
6.6	Seafood purchase methodology	13
6.7	Underwater sediment collection methodology	13
7	Sample preparation, laboratory analysis and descriptors	15
7.1	Preparation and analysis	15
7.2	Sample descriptor code	16
8	Results and interpretation	17
8.1	Dose rates	18
8.2	Intertidal sediment results	18
8.3	Underwater sediment results	19
8.4	Intertidal seaweed results	19

8.5	Biota results						
8.6	Results summary 21						
9	Evaluation of potential exposure pathways 21						
9.1	External	radiation	21				
9.2	Inhalatio	on of re-suspended DU	21				
9.3	Ingestion	of DU contaminated foodstuffs	22				
10	Conclusi	ons	22				
11	1 List of references 23						
12	KTA 200	7 survey results (Pages 25 to 29)	24				
APPE	ENDIX A	Historical data	31				
ANNI	EX A	Issues to be considered when interpreting or comparing uranium data	34				
ANNI	EX B uranium	Change in the $^{238}$ U/ $^{234}$ U activity ratio of a medium containing natural with the addition of depleted uranium	37				
ANNI	ANNEX C Variability of uranium concentration and uranium isotopic ratios in marine environmental samples 38						
ANNI	EX D	Reference values for uranium in the Solway Firth and the UK	39				
ANNI	ANNEX E List of references to annexes 41						
Initia	Initial distribution 42						

## List of figures

Figure 1: Number of DU projectiles fired from KTA, from 1982 to 2007.	9
Figure 2: Approximate cumulative mass of DU projectiles fired from KTA, from 1982 to 2007.	9
Figure 3: KTA intertidal sampling locations, October 2007.	11
Figure 4: KTA underwater sampling locations, October 2007 (also showing line of fire for each battery).	14

## List of tables

Table 1: KTA intertidal sample station locations, October 2007.	11
Table 2: Summary of intertidal sample collection, October 2007.	13
Table 3: KTA underwater sampling locations, October 2007.	15
Table 4: Summary of results, all marine samples 2007 (values have been rounded to 1 decimplace for clarity).	nal 17
Table 5: Intertidal dose rate measurements, October 2007.	18
Table 6: Intertidal (shoreline) sediment samples, total uranium and isotopic ratio results –October 2007.	25
Table 7: Underwater sediment samples, total uranium and isotopic ratio results – October2007.	26
Table 8: Seaweed samples, total uranium and isotopic ratio results – October 2007.	28
Table 9: Marine biota samples, total uranium and isotopic ratio results – October 2007.	29
Table 10: Summary of marine results reported for 1996-2007.	32
Table 11: Typical uranium concentrations and isotopic ratios in marine samples.	38
Table 12: Reported <sup>238</sup> U concentrations in seaweed and marine biota in the UK.	39

## 1 Introduction

This report presents and interprets the results of the 2007 routine monitoring programme for depleted uranium (DU) in the marine environment bordering and offshore from the Kirkcudbright Training Area (KTA). Intertidal shoreline samples, underwater sediment and marine biota samples were collected, crustaceans were obtained, and environmental dose rate measurements were made in October 2007.

Routine marine environmental monitoring has been carried out annually since 1980 (i.e. before firing began) to measure uranium levels in the marine environment at the KTA and to identify the extent of any dispersion of DU that might result from operations at the site. The monitoring programme has consisted principally of the collection of seawater, marine sediment, seaweed and marine biota, which were subsequently analysed in the laboratory.

As may be expected over twenty-eight years, the monitoring programme has improved to reflect increasing knowledge of the local conditions and changes in best practice. This has resulted in differences such as sample type and reporting units. However, since 1996, sample type, analysis technique and reporting have been broadly consistent. The changes made to the sea water and underwater sediment sampling protocol over the years were explained in the Marine Environmental Depleted Uranium Survey Report Kirkcudbright Training Area – 2005 [1]. The rationale for sampling underwater sediment adopted in 2005 was followed during the 2007 survey.

The yearly sampling of sediment, seaweed and mussels, and the measurement of dose rates, from the intertidal shoreline of the Dumfries coast were added from 1994 onwards, as was the analysis of locally caught seafood. The total number of shore locations sampled in each survey is 11.

The monitoring for DU in the terrestrial environment undertaken at Kirkcudbright during 2007 is reported separately [2].

#### 2 Background

The KTA range is located on the coast of Dumfries and Galloway, near Castle Douglas. Since April 2006, the range has been part of the Defence Training Estate.

DU has been released into the environment at Kirkcudbright as a consequence of the test firing of DU ammunition during design and accuracy assessment trials. The trials, and current proof firings, involve firing DU projectiles through soft vertical targets into the Solway Firth.

Testing of projectiles historically has taken place at five locations on the Kirkcudbright site. Each battery location had a designated target and line of fire, and hence a predictable impact area ranging from several hundred metres to about 7 km offshore. Although a small fraction of the total number of penetrators malfunctioned and impacted on land, the

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vast majority of malfunctioning penetrators, and all the functioning penetrators, entered the Solway Firth. The number of DU rounds fired each year at Kirkcudbright from the five firing locations and the cumulative mass of DU fired to date, are presented in Figure 1 and Figure 2 respectively.

## **3** Depleted uranium

There are three broad types of uranium: natural, enriched and depleted. Uranium is a naturally occurring radioactive material that is found as a mixture of three isotopes: uranium-238 (<sup>238</sup>U), uranium-235 (<sup>235</sup>U) and uranium-234 (<sup>234</sup>U). It emits alpha and beta particles, gamma and X radiation. Uranium, in an 'enriched' form, is used as fuel in nuclear reactors. It is called enriched because, due to processing, it contains a higher concentration (> 0.72%) of <sup>235</sup>U than natural uranium. The by-product of the enrichment process is depleted uranium (DU), which has a reduced concentration of <sup>235</sup>U. Some <sup>234</sup>U is also removed in the depletion process. Because almost all (>99% by mass) of the uranium in natural and depleted uranium is <sup>238</sup>U, the <sup>238</sup>U concentration and therefore the <sup>238</sup>U activity remain almost the same whatever the depletion status<sup>1</sup>. The mass compositions of DU and of natural uranium are presented below.

	<sup>238</sup> U	<sup>235</sup> U	<sup>234</sup> U
Natural uranium	99.274%	0.72%	0.00554%
The DU used at Kirkcudbright	99.8%	0.20%	0.0008%

In natural uranium, the <sup>234</sup>U activity is normally in equilibrium with the <sup>238</sup>U activity, but as a result of depletion and the reduction in the concentration of <sup>234</sup>U, the <sup>238</sup>U/<sup>234</sup>U- activity ratio changes. The actual <sup>238</sup>U/<sup>234</sup>U activity ratio varies per batch of DU, according to the degree of depletion achieved during processing, but usually lies between 7:1 and 8:1. Consequently, DU is slightly less radioactive than natural uranium.

For the remainder of this report isotopic ratios will be stated as a single value representing the ratio of a number of Becquerels (Bq) of  $^{238}$ U to 1 Becquerel of  $^{234}$ U (i.e. a  $^{238}$ U/ $^{234}$ U ratio of 7 rather than 7:1).

When converting total uranium concentrations, reported in micro-grams of uranium per gram of sample ( $\mu g/g$ ) to activity, two specific activities have been used as conversion factors, depending on the provenance of the uranium. The specific alpha activity of uranium in the DU alloy used at Kirkcudbright is approximately 14 kBq/g, whereas the specific alpha activity of natural uranium is generally 25 kBq/g [3]. The chemical toxicity of DU is about the same as lead.

<sup>&</sup>lt;sup>1</sup> Changes in total uranium can be estimated with reasonable accuracy by measuring changes in the concentration or activity of 238U. (See ANNEX B.)

## 4 Differentiating DU from natural uranium

A fundamental requirement of the DU environmental monitoring programme is to quantify the impact of DU firing. Historically, this was achieved partly by measuring the amount of total uranium in environmental materials and using this total uranium as an upper bound of DU contamination levels. This is a very cautious approach as natural uranium is present in most natural materials. More sophisticated approaches are now used. These involve specific measurements of the activities or masses of the <sup>238</sup>U and <sup>234</sup>U isotopes as described below. Although isotope measurements are used in this work, references to total uranium measurements are included for consistency with previous reports. The limitations of using total uranium concentrations are discussed further in ANNEX A.

As noted in Section 3, a convenient fingerprint marker for DU is the  ${}^{238}U/{}^{234}U$  activity ratio. The DU fired at KTA has a  ${}^{238}U/{}^{234}U$  activity ratio of approximately 7, whereas natural uranium has an activity ratio close to 1. Environmental samples are therefore analysed for isotopes of  ${}^{238}U$  and  ${}^{234}U$  to determine activity ratios and hence identify the origin of the uranium.

Substantial deposition of DU, relative to an existing natural uranium inventory, is required before the  ${}^{238}$ U/ ${}^{234}$ U activity ratio in the environment diverges significantly from its natural ratio. (An illustration of the impact of increasing the DU mass in a sample on the sample uranium isotopic ratio is given in ANNEX B). For the ratio to approach 7 in an analytical sample, the mass of DU would have to be about one hundred times the mass of the uranium that is naturally present. Hence, the lower the natural uranium background, the lower the levels of DU contamination that may be detected by isotopic analysis.

Isotopic quantification is achieved by techniques such as Alpha Spectrometry (AS) or Mass Spectrometry (MS). AS can detect uranium to parts per billion, which is equivalent to mBq per kg, or to lower levels if count times are increased. MS is more sensitive than AS, but the lower levels detectable by MS are of no recognised health significance. Furthermore, because most MS measurements are not accredited by the UK Accreditation System (UKAS), the technique is not normally used in routine health and safety or environmental monitoring<sup>2</sup>.

Gamma Spectrometry measurements of environmental samples can also yield isotopic information, but the technique is not sensitive enough to reach the limits of detection required for environmental analysis unless relatively large quantities of uranium are present.

<sup>&</sup>lt;sup>2</sup> For academic studies, Inductively Coupled Plasma (ICP) MS has become the analytical technique of choice for the measurement of very low uranium content and /or the determination of the isotopic ratio in environmental and biological samples.



Figure 1: Number of DU projectiles fired from KTA, from 1982 to 2007.



Figure 2: Approximate cumulative mass of DU projectiles fired from KTA, from 1982 to 2007.

DSTL/CR31252 V1

## 5 Investigation/action levels

Whilst the Depleted Uranium Firing Environmental Review Committee (DUFERC), on which the MOD is represented, has set investigation or action levels for DU in terrestrial environmental samples, for marine environmental samples reference may be made to official guidance and legislation directly. For sediment, reference may be made to both the Radioactive Substances Act 1993 (RSA 93) or to the Generalised Derived Limit (GDL) for <sup>238</sup>U [4, 5]. For substances commonly used as foodstuff, the GDL is the appropriate reference quantity. For material such as seaweed, the reference level used is the European Council Food Intervention Level (CFIL) for other foodstuff of 1,250 mBq/g [6].

GDLs for uranium were last updated in 2000 by the National Radiological Protection Board, which is now part of the Health Protection Agency. GDL relate to an annual dose of 1 mSv for all pathways. This dose was also used as a benchmark by the Royal Society in their studies of the potential health effects of using DU munitions [7]. GDLs exist for marine crustaceans and molluscs. The GDLs for foodstuffs are for the edible fraction and are expressed for fresh mass, whilst the GDLs for marine sediment are for dry mass.

	Generalised Derived Limit for <sup>238</sup> U (mBq/g)
Marine crustaceans	1,000
Marine molluscs	1,000
Marine sediment	100,000

## 6 Sample collection

#### 6.1 Sampling locations and dates of sampling

In October 2007, for 10 locations, intertidal samples were collected at the annual survey pre-determined sampling locations within the restrictions imposed on the day of sampling by the height of the low tide. In contrast, sampling for Location 5 was moved by approximately 2 km northeast to Baccarry Bay because access to the pre-determined location had become overgrown and unsafe. Samples of intertidal sediment and, where available, seaweed and mussels were collected from the 11 sampling locations shown in Table 1 and Figure 3. Crabs, whelks and lobsters were purchased locally.

The provenance, types and numbers of samples procured from the intertidal sampling locations are listed in Table 2.

Samples of underwater sediment were collected from the sampling locations listed in Table 3 and illustrated in Figure 4, as described later in section 6.7.

#### 6.2 Intertidal sediment sampling methodology

The composition, abundance and availability of sediment vary with time and location. Because of this, each intertidal sediment sample was a composite from a number of patches within each of the general sampling areas shown on Figure 3. Sediment was collected as near to the Low Water Mark (LWM) as safely possible. Where several sediment types were present in the sampling area, the finest sediment was selected, and large shells and stones were discarded. At each sampling location, the uppermost layer of sediment was collected using a trowel, and combined with other sub-samples in a 0.8 litre watertight plastic container. This procedure was repeated at intervals along the shoreline until the container was full. Excess water was drained from the container prior to sealing.

	Sample Station	
Number	Name	Grid reference
1.	South Carse	NX 99456 59431
2.	Sandyhills Bay	NX 89213 55172
3.	Port o' Warren Bay	NX 87901 53438
4.	Port Donnel	NX 84776 53714
5.	Balcarry Bay	NX 82182 49544
6.	Abbey Burn Foot	NX 74200 44413
7.	Mullock Bay	NX 70996 43765
8.	Lower Nunton Bay	NX 66081 48424
9.	Brighouse Bay	NX 63508 45401
10.	Carrick Point	NX 57599 50722
11.	Mossyard Bay	NX 55207 52007

Table 1: KTA intertidal sample station locations, October 2007.



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Figure 3: KTA intertidal sampling locations, October 2007.

#### 6.3 Seaweed collection methodology

Seaweed was collected when found in sufficient abundance. There was no seaweed to collect at Location 1. The collected samples consisted of single species of seaweed, either *F. ceranoides* (Location 3 only), *F. vesiculosus* (Location 7, 9, 10, 11) or *F. spiralis* (other locations). Only the most recent growth of seaweed was collected, by cutting the end 5 cm of fronds. As with the sediment samples, each seaweed sample was a composite from a number of locations within a sampling area. Samples were collected at intervals along the shoreline until a 1.8 litre water tight plastic sample pot was full. The seaweed samples were washed in sea water at the time of collection, to remove loosely adherent sediment from the fronds.

#### 6.4 Biota collection methodology

Molluscs were only collected when found in abundance. Samples of mussel (*M. edulis*) were collected from 5 of the sampling areas shown on Figure 3. (See Table 2.) Mussels were collected at a number of points along the shoreline of a sampling location, and combined into a composite sample. The composite sample filled a 1.8 litre sampling pot for each sampling location. The mussel samples were washed at the time of collection to remove loosely adherent sediment from the shells. (Limpet or winkle samples are sometimes collected when mussels are not available. However during October 2007 neither mussels, winkles nor limpets could be found at six of the sampling areas.)

On the day of collection, the mussel samples were further rinsed in soft water to remove further sediment prior to cooking. They were then boiled in water for a few minutes, drained and prised from their shells taking care not to include sediment with the flesh. The shells and cooking water were discarded, as were any mussels that remained closed after boiling. The flesh was placed in an individual resealable plastic bag and frozen prior to transport to the laboratory.

#### 6.5 Dose rate measurement methodology

Gamma radiation measurements were made using three Mini Instruments Type 6-80/81 rate meters fitted with compensated Geiger Muller tubes. The instruments were deployed so that the centres of the Geiger Muller tubes were at one metre above ground, at the High Water Mark (HWM) and the low water mark (LWM<sup>3</sup>) at each sampling location (away from granite boulders and sea walls). Three 100 second counts were taken at the LWM at distance intervals of 1m, and one 100 second count was taken at the HWM. The average of the four counts was used to calculate the dose rate.

<sup>&</sup>lt;sup>3</sup> The gamma monitors were placed as near as was safely possible to the LWM.

Sample	Sample Type, and number of samples			Environmental Dose rates		
Area	Sediment	Seaweed	Mussel	LWM @ 1m	HWM @ 1m	
Number				height	height	
1	$\checkmark$	х	х	$\checkmark$	$\checkmark$	
2	$\checkmark$	$\checkmark$	х	$\checkmark$	$\checkmark$	
3	$\checkmark$	$\checkmark$	$\checkmark$	~	$\checkmark$	
4	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	
5	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	
6	$\checkmark$	$\checkmark$	х	$\checkmark$	$\checkmark$	
7	$\checkmark$	$\checkmark$	х	$\checkmark$	$\checkmark$	
8	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	
9	$\checkmark$	$\checkmark$	х	$\checkmark$	$\checkmark$	
10	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	
11	~	$\checkmark$	х	$\checkmark$	$\checkmark$	

Table 2: Summary of intertidal sample collection, October 2007.

Notes: LWM – Low Water Mark

HWM – High Water Mark × Sample not collected.

#### 6.6 Seafood purchase methodology

Four locally caught lobsters (*H.gammarus*), three crabs and a bag of locally farmed whelks (*B. undatum*) were purchased in Kirkcudbright and boiled within a day of purchase. These were then bagged and frozen pending transport to the Dstl laboratory. At the time of survey in October 2007, there were no locally fished queen scallops (*C. opercularis*) for sale in Kirkcudbright.

#### 6.7 Underwater sediment collection methodology

Underwater sediment samples (25) were collected during October 2007, from all 25 locations shown in Figure 4 and are listed in Table 3.

The dynamic nature of the sea makes the sampling of underwater sediment at precise locations difficult. A scooping device fitted with a rubberised bag was lowered in the water at each sampling location. This was dragged along the sea bed over a distance ranging between 50 and 100 m depending on tide and wind conditions. The location coordinates given in Table 3 should therefore be considered to be the approximate centre points of sampling areas of about 100 m radius.



Figure 4: Approximate KTA underwater sampling locations, October 2007 (also showing line of fire for each battery).

Sample Station Number	October 2007			
	Latitude			Longitude
		(North)		(West)
1	54°	45.078'	3°	59.782'
2	54°	45.600'	3°	59.808'
3	54°	44.527'	3°	59.746'
4	54°	45.569'	4°	2.577'
5	54°	45.032'	4°	2.541'
6	54°	44.479'	4°	2.522'
7	54°	45.211'	4°	1.174'
8	54°	45.083'	4°	1.243'
9	54°	44.827'	4°	1.391'
10	54°	44.684'	4°	1.477'
11	54°	44.991'	3°	58.807'
12	54°	44.856'	3°	58.758'
13	54°	45.484'	3°	57.969'
14	54°	45.375'	3°	57.744'
15	54°	45.673'	3°	57.125'
16	54°	45.136'	3°	57.108'
17	54°	44.575'	3°	57.081'
18	54°	42.874'	4°	1.311'
19	54°	42.747'	4°	1.347'
20	54°	43.235'	4°	2.493'
21	54°	42.732'	4°	2.476'
22	54°	42.225'	4°	2.451'
23	54°	43.292'	4°	0.243'
24	54°	42.785'	4°	0.226'
25	54°	42.258'	4°	0.192'

Table 3: KTA underwater sampling locations, October 2007.

## 7 Sample preparation, laboratory analysis and descriptors

#### 7.1 **Preparation and analysis**

Sample preparation was either carried out in the field at the time of collection, as described in sections 6.3, 6.4 and 6.6, or at the Dstl Environmental Sciences Department UKAS accredited radiochemistry laboratory, on receipt of the sample. The samples were analysed by AS in the laboratory. An outline of the approach is given below.

One mixed sample of mussel flesh from each of the five locations was submitted for analysis. Whelks were boiled and the meat was prised from each shell (the shells were

discarded). Cooked lobsters and crabs were opened up and the flesh removed for analysis, but the digestive tracts of the animals were not used. Both the white and the brown meat of the crabs were used for analysis. Each lobster and crab was analysed as a discrete sample, which gave 4 individual lobster samples (each sample consisting of the flesh of one lobster only), and 3 individual crab samples (each sample consisting of the flesh of one crab only). The whelk meat was divided, in a similar fashion, into 8 replicate samples of similar mass.

The sediment, seaweed and seafood samples thus obtained were weighed before drying to remove moisture, and then weighed again (with results being reported as dry weight). The samples were ashed to remove organic material, and homogenised by hand. During this process the quantity of uranium in each of the samples remains constant. The ashed samples were boiled in concentrated mineral acid (nitric acid and hydrochloric acid) to remove the 'loose' and leachable uranium from the sample. Recalcitrant matrices such as mineral grains are not broken down by the process and hence the natural uranium within them was not extracted. The samples were filtered to remove solids.

Uranium separation was carried out by extraction chromatography. Each eluted sample was electro-deposited onto a stainless steel planchette and the activity of each planchette was counted in a low background, silicon surface barrier, alpha spectrometer.

DU concentrations are reported in mBq/g (equivalent to Bq/kg) of dry weight.

#### 7.2 Sample descriptor code

Each sample was given a unique sample descriptor. For intertidal and underwater sediment, seaweed and mussels the descriptor comprised of a descriptor of sample type, followed by a location descriptor.

The sample type descriptors are as follows:

- I/T sed intertidal sediment;
- U/W sed underwater sediment;
- Seaweed F. vesiculosis/ F. ceranodies/ F. serratus/ F.spiralis
- Mussel *M. edulis* and
- Lobster, Crab, Whelks.

In the case of purchased sea food no location descriptor has been included because the precise location were the sample was collected is unknown. For replicate samples of whelk and for individual crabs and lobsters, letters A to H were added.

Five examples of the sample descriptors are given below:

I/T sed 3 – for intertidal sediment collected from intertidal location 3.

U/W sed 8 – underwater sediment collected at underwater location 8, (54°45.083'N and  $4^{\circ}1.243'W$ ).

Mussels 10 – Mussel sample from intertidal sampling location 10, Carrick Point.

Whelk C – Third replicate sample from the purchased composite sample of whelk.

Crab C – Third crab of the three crabs purchased.

## 8 **Results and interpretation**

A summary of the results for all marine samples collected in 2007 is given in Table 4. The detailed marine monitoring results<sup>4</sup> are given later in Table 5 to Table 9.

Historical monitoring results for the KTA marine environment are presented in Table 10 at APPENDIX A and provide an overview of the routine monitoring results through time, from 1996 to 2007 [1, 8, 9, 10, 11, 12, 13, 14, 15, 16].

	No. of		Total uranium concentration (mBq/g)			
Sample Type	Number of Samples	possibly containing detectable DU <sup>5</sup>	Mean	Standard deviation of the mean	Minimum	Maximum
Intertidal Sediment	11	0	16.3	6.0	6.9	27.6
Seaweed	10	0	15.5	4.5	10.7	25.6
Mussels	5	0	8.2	2.8	4.8	11.9
Lobsters	4	0	0.1	0.1	0.1	0.2
Whelks	8	0	0.7	0.3	0.4	1.2
Crabs	3	0	0.6	0.2	0.4	0.8
Underwater Sediment	25	4	26.6	4.8	19.5	42.3

Table 4: Summary of results, all marine samples 2007 (values have been rounded to 1 decimal place for clarity).

To identify whether the DU firing activities at KTA have had a detectable impact on uranium concentrations in the local marine environment, it is possible to compare the results for the routine monitoring programme with uranium activities and ratios reported for similar sample types across the UK. Two studies have reported ambient dose rates and uranium activity in marine sediment, seaweed and biota samples from a range of coastal locations in the UK [17, 18]. One of the studies [18] included samples from a location in the southwest of Scotland, named Sandyhills Bay (Location 2), which is sufficiently near

<sup>&</sup>lt;sup>4</sup> Tables 6 to 9 are presented on pages 25-29 of this report to aid readability.

<sup>&</sup>lt;sup>5</sup> A sediment sample may contain DU if it has a  ${}^{238}$ U/ ${}^{234}$ U isotopic ratio above a value of 0.8 (after subtracting the related uncertainty). However, ratios of about 1.0 were recorded at KTA even before DU firings began. (See Sections 8.2 and 8.3.)

to KTA to be geochemically similar and sufficiently far to be considered unaffected by activities at the range. (See ANNEX D.)

#### 8.1 Dose rates

Dose rate measurement results for 2007 are given in Table 5. The maximum recorded dose rate was 120 nGy/h. The results compare with four dose rate measurements averaging 72 nGy/h made by the Centre for Environment, Fisheries and Aquaculture Science (CEFAS) in 2005 over a salt marsh at Kirkcudbright [17] and with measurements made as part of the routine monitoring programme in previous years, and reflect background dose rates.

Station		Average Dose rate
Number	Location	(nGy/h)
1	South Carse	65
2	Sandyhills Bay	70
3	Port o' Warren Bay	110
4	Port Donnel	95
5	Balcarry Bay	120
6	Abbey Burn Foot	95
7	Mullock Bay	95
8	Lower Nunton Bay	65
9	Brighouse Bay	65
10	Carrick Point	65
11	Mossyard Bay	85

Table 5: Intertidal dose rate measurements, October 2007.

#### 8.2 Intertidal sediment results

No sample was radioactive within the meaning of the RSA 93, or exceeded 0.03% of the GDL for uranium in sediment [4, 5].

As mentioned in Section 4, total uranium concentrations have also been used historically to provide an upper bound on DU contamination levels. Previous monitoring reports [1, 14, 15] have compared sample results with the range of total uranium activity concentrations in UK coastal sediment<sup>6</sup>[27]. In 2007, the intertidal sediment samples all contained total U at activity concentrations (6.9 to 27.6 mBq/g of dry weight) that were within this range. (See Table 6.) The range of total uranium activity concentrations for the samples was also well within the range of results reported for intertidal sediment collected since 1996. (See APPENDIX A.)

<sup>&</sup>lt;sup>6</sup> Minerals containing uranium are widely distributed on the surface of the earth's crust and the concentration of natural uranium in the environment therefore varies between locations. Consequently there is no single, definitive, reference level for natural uranium in soils or the sediment that originate from these soils, but there is broad agreement in the range of values published in literature. Typical values in the UK range from 3.6 to 32.3 mBq/g. of dry weight but values of up to 100 times the typical range can be found locally.

For sediment obtained from Sandyhills Bay [18] in southwest Scotland (See ANNEX D.), only the <sup>238</sup>U activity concentration was reported ( $14 \pm 0.4 \text{ mBq/g}$  dry weight), as is common in environmental reports. (See ANNEX A.) It can be seen from Table 6, that all intertidal sediment samples collected at KTA in 2007 had a <sup>238</sup>U activity concentration at or below that value.

Within the range of the uncertainty for the analysis, the isotopic ratios for all the intertidal sediment samples collected are in agreement with a value of 0.8 reported in marine sediment in general. (See ANNEX C.)

#### 8.3 Underwater sediment results

No sample was radioactive within the meaning of the RSA 93 or exceeded 0.05 % of the GDL for uranium in sediment [4, 5].

The <sup>238</sup>U activity concentrations reported (in Table 7) for all the 25 underwater sediment samples collected (9.5 to 20.0 mBq/g of dry weight) were all within the UK coastal sediment range of 3.6 to 32.3 mBq/g of dry weight. (See ANNEX D.) The range of total uranium activity concentrations for all but one sample agreed with ranges of values reported in previous years. (Sample U/W sed 1 had a total uranium activity of natural origin that was approximately double that of the other 20 underwater sediment samples, but still less than 0.05% of the GDL for sediment. This variation is not deemed to be significant.)

The <sup>238</sup>U/<sup>234</sup>U isotopic ratios reported for 21 underwater sediment samples were, within the range of the uncertainty for the analysis, in agreement with values generally reported in marine sediment. (See ANNEX C.) Four samples (U/W sed 3, 5, 17, 19) had a uranium isotopic ratio (ranging from 0.9 to 1.0 after taking off the analytical uncertainty) greater than the typical isotopic ratio in marine sediment of 0.8. However, this is not unusual, and isotopic ratios of around 1.0 were recorded in sediments from KTA even before DU munitions testing began[16]. The finding is also radiologically insignificant as the total uranium activity concentrations (22.7-28.8 mBq/g) were very much less than 1% of the GDL for sediment of 100,000 mBq/g of dry weight.

#### 8.4 Intertidal seaweed results

No sample exceeded 2 % of the CFIL for uranium in other foodstuff [6].

The total uranium activity concentrations and the  $^{238}$ U activity concentrations for the seaweed samples obtained from the 10 locations sampled were low (10.7 to 25.6 mBq/g of dry weight for total uranium, 4.1 to 12.2 mBq/g of dry weight for  $^{238}$ U – see Table 8), and in agreement with the range of  $^{238}$ U activity concentrations reported for seaweed sampled in the UK as a whole (3.8 to 18.6 mBq/g). All samples had  $^{238}$ U activity concentrations that were slightly higher than values reported for seaweed obtained from Sandyhills Bay and reported elsewhere. The range of total uranium results is in agreement with values reported in previous years. (See APPENDIX A.)

There are no reported values in the literature of the  $^{238}U/^{234}U$  isotopic ratios in seaweed. Values reported here are in agreement with those reported for the same locations from previous years, and do not indicate the presence of depleted uranium.

#### 8.5 Biota results

The GDLs for foodstuffs are expressed for <sup>238</sup>U in fresh mass (or wet weight), as are activity concentrations for UK marine biota reported by CEFAS [17]. In contrast, the KTA 2007 survey results (for <sup>238</sup>U) reported here are per gram of dry weight. Consequently, activity results must be adjusted for the lost moisture to allow comparison with these two sources of data. From such comparison (detailed below), it is concluded that no sample exceeded 5 % of the GDL for uranium in molluscs and crustaceans [5].

The five mussel samples collected during October 2007 had total uranium activity concentrations from 4.8 to 11.9 mBq/g of dry weight, and <sup>238</sup>U activity concentrations of 2.2 to 5.3 mBq/g of dry weight. (See Table 9.) Since the drying step of the mussel sample preparation removed moisture equal to 75 % of the sample wet weight on average, the corresponding range of wet weight <sup>238</sup>U activity concentration for the mussel samples obtained in the survey would be 19.2 - 47.6 mBq/g. (Sample preparation is discussed in Section 6.3 and at ANNEX A). These values are two orders of magnitude below the GDL value of 1,000 mBq/g for <sup>238</sup>U activity concentrations in molluscs and crustaceans. (See Section 5.)

The <sup>238</sup>U activity concentrations of dry weight are slightly above the values reported for mussels sampled from Sandyhills Bay, but were at the lower end of the range of values reported for mussels sampled in the UK as a whole. The variation is not of any practical significance, at such low activity concentrations. The mean total uranium activity concentration is in agreement with mean values reported in previous years. (See APPENDIX A.)

No <sup>238</sup>U could be detected in any of the four lobsters, and in two of the three crab samples (Crab samples A and C). It was therefore not possible to calculate any isotopic ratio for these samples.

Since the preparation drying of crab sample B removed moisture equal to 88 % of the sample wet weight, the corresponding wet weight <sup>238</sup>U activity concentration for crab sample B obtained in the survey would be  $4.0 \pm 1.3$  mBq/g. (Sample preparation is discussed in Section 6.3 and at ANNEX A.) This value is two orders of magnitude higher than the mean <sup>238</sup>U activity concentrations for crabs obtained from the UK as a whole, but is very much less than the GDL of 1,000 mBq/g for <sup>238</sup>U activity concentrations in molluscs and crustaceans. (See Section 5.)

The dry weight total uranium activity concentrations for the duplicate whelk samples range from 0.4 to 1.2 mBq/g. and the <sup>238</sup>U activity concentrations detected in three of the whelk samples compare with <sup>238</sup>U activity concentrations for other molluscs from Sandyhills Bay. Their wet weight equivalent (based on 83% moisture loss) range from 2.35 to 7.1 mBq/g, and so are slightly elevated compared to values from the UK at large, but the variation is of no significance because the activity concentrations are well below

the GDL of 1,000 mBq/g for  $^{238}$ U activity concentrations in molluscs and crustaceans. (See Section 5.)

The mean total uranium activity concentrations for all biota samples are in agreement with mean values reported in previous years. (See APPENDIX A.)

#### 8.6 **Results summary**

These findings are in agreement with those from previous years and do not indicate any environmental impact from the DU firings.

#### 9 Evaluation of potential exposure pathways

The contamination of the marine environment with DU would result in four potential exposure pathways for humans. These are:

- external radiation exposure from contaminated seaweed or sediment;
- inhalation of DU contamination released into the air or re-suspended from sediment;
- ingestion of seaweed or food products contaminated with DU; and
- ingestion of DU contaminated sea water. Although ingestion of DU contaminated sea water is a theoretical possibility, seawater sampling was discontinued at the recommendation of SEPA in 2001. The technical assessment was that the immense dilution present in the Solway would never give rise to uranium levels detectable above background, or to any significant radiation dose from DU.

#### 9.1 External radiation

Radiation levels on the shore are indistinguishable from natural background levels and hazards due to external radiation exposure from DU are therefore considered to be insignificant.

#### 9.2 Inhalation of re-suspended DU

DU that has been deposited in sediment or in sediment attached to objects and plants such as fishermen's netting and seaweed may be re-suspended into the air and subsequently inhaled. Results reported here show that measured total uranium activity concentrations do not exceed background values for UK sediment and that there is no conclusive evidence of the presence of DU from munitions testing in the isotopic ratios. Consequently, it is concluded that there are no risks from inhalation of DU to any member of the public.

#### 9.3 Ingestion of DU contaminated foodstuffs

Total uranium concentrations in the marine foodstuffs were in agreement with background levels for such sample types. There was no evidence of depletion in the isotopic ratio. It is concluded that there are no risks from ingestion of DU to any member of the public.

## 10 Conclusions

It is concluded that:

- the survey results show no conclusive evidence of DU from munitions testing being present in any marine environmental sample collected in the year 2007;
- uranium activity concentrations are far below levels (GDLs and CFILs) set by government and regulatory authorities to ensure protection of the public; and,
- there is no evidence to suggest that members of the public are or have been subjected to any radiation hazard from the marine environment due to the firing of DU ammunition at Kirkcudbright.

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12 KTA 2007 survey results (Pages 25 to 29)

Sample Wet Dry Ashed Measured Activity of D									Dry S	Sam	ple (n	nBq/g	)		Ratio of			
Descriptor	weight (g)	weight (g)	weight (g)		<sup>238</sup> U	J	2	<sup>235</sup> U	J	2	<sup>234</sup> U	[	Тс	otal	U	238	$U/^2$	<sup>34</sup> U
IT sed 1	508.9	408.4	405.0	3.5	±	0.9		<	0.6	3.3	±	0.9	6.9	±	1.3	1.1	±	0.4
IT sed 2	476.2	377.6	373.1	5.2	±	1.2		<	0.7	5.2	±	1.2	10.5	±	1.8	1.0	±	0.3
IT sed 3	484.3	381.9	374.2	8.5	±	1.7		<	0.6	8.2	H	1.6	17.0	±	2.3	1.0	±	0.3
IT sed 4	451.2	349.9	343.7	1.7	±	8.4		<	0.6	9.0	H	1.7	17.6	Ħ	2.4	0.9	H	0.3
IT sed 5	495.1	326.0	307.1	2.4	±	13.5		<	0.7	13.6	H	2.4	27.6	±	3.4	1.0	±	0.3
IT sed 6	447.3	296.4	287.6	1.6	±	7.4		<	0.7	7.9	H	1.7	15.8	±	2.3	0.9	±	0.3
IT sed 7	495.0	325.1	306.4	2.3	±	13.7		<	0.6	12.1	H	2.1	26.2	Ħ	3.1	1.1	H	0.3
IT sed 8	481.9	376.4	366.2	1.5	±	7.8		<	0.6	6.9	H	1.4	15.0	±	2.1	1.1	±	0.3
IT sed 9	519.3	399.4	386.7	1.4	±	7.0		<	0.6	7.6	H	1.5	14.8	Ħ	2.0	0.9	H	0.3
IT sed 10	449.6	331.4	320.6	1.4	±	6.6		<	0.6	6.4	ŧ	1.4	13.3	±	2.0	1.0	Ŧ	0.3
IT sed 11	520.7	403.6	396.7	1.6	±	7.7		<	0.7	7.0	±	1.5	15.1	±	2.2	1.1	±	0.3

Table 6: Intertidal (shoreline) sediment samples, total uranium and isotopic ratio results - October 2007.

**Note:** Activity results have been rounded to 1 decimal place. A specific activity for DU of 14.0 MBq/kg has been used. All uncertainties are stated at a 95% confidence level. Limits of Detection (LOD) are calculated by a 'modified Currie' formula<sup>7</sup> at 95%. The total activity is calculated from the sum of the actual activities for each isotope, regardless of the LOD quoted for that isotope, hence in Table 6, where activities are reported as less than LOD for any of the uranium isotopes, the total uranium value may not be equal to the sum of the individual isotopic values.

<sup>&</sup>lt;sup>7</sup> Revisiting Currie - how low can you go? by Hurtgen C, Jerome S, Woods M. Applied Radiation and Isotopes 53 pp 45-50 (2000) Dstl/CR31252 V1

Sample	Wet	Dry	Ashed		Measured Activity of Dry Sample (mBq/g)								Ratio of					
descriptor	weight	weight	weight		<sup>238</sup> U			<sup>235</sup> U			<sup>234</sup> U		Т	otal I	IJ	238	$^{3}U/^{23}$	$^{4}$ U
	(g)	(g)	(g)										-	otur	e -			
U/W sed 1	493.0	351.2	336.0	20.0	±	3.3	1.0	±	0.5	21.3	±	3.5	42.3	±	4.8	0.9	±	0.2
U/W sed 2	512.7	354.2	334.6	9.5	±	1.6		<	0.5	9.6	±	1.6	19.5	±	2.3	1.0	±	0.2
U/W sed 3	537.0	387.0	370.7	13.5	±	2.3		<	0.6	11.1	±	2.0	24.7	±	3.0	1.2	±	0.3
U/W sed 4	489.9	322.3	303.1	14.6	±	2.3	0.7	Ħ	0.3	14.1	Ŧ	2.2	29.4	±	3.2	1.0	±	0.2
U/W sed 5	519.4	362.9	336.8	15.6	±	2.5		<	0.6	11.6	±	2.0	27.7	±	3.2	1.3	±	0.3
U/W sed 6	530.3	367.6	351.5	13.6	±	2.3	0.8	±	0.4	14.2	±	2.4	28.6	±	3.3	1.0	±	0.2
U/W sed 7	597.0	390.9	372.5	11.6	±	2.1		<	0.7	11.3	±	2.0	23.3	±	2.9	1.0	±	0.3
U/W sed 8	578.4	386.0	368.1	13.3	±	2.2	0.6	±	0.4	13.0	±	2.2	26.9	±	3.1	1.0	±	0.2
U/W sed 9	548.8	384.4	365.8	11.9	±	2.2		<	0.8	14.2	±	2.5	26.8	±	3.4	0.8	±	0.2
U/W sed 10	556.7	395.1	377.7	12.4	±	2.2	0.8	Ħ	0.4	12.4	Ŧ	2.2	25.5	±	3.1	1.0	±	0.3
U/W sed 11	546.9	390.6	374.9	11.6	±	2.1		<	0.7	12.3	±	2.2	24.5	±	3.1	0.9	±	0.2
U/W sed 12	523.4	371.3	354.6	12.0	±	2.2		<	0.8	12.9	±	2.3	25.3	±	3.2	0.9	±	0.2
U/W sed 13	509.5	370.4	354.4	12.6	±	2.2	0.8	±	0.4	11.6	±	2.1	25.0	±	3.1	1.1	±	0.3
U/W sed 14	498.5	350.6	337.0	10.5	±	1.9	0.6	±	0.3	10.3	±	1.8	21.4	±	2.6	1.0	±	0.3
U/W sed 15	540.7	371.1	356.4	9.8	±	1.9		<	0.7	10.1	±	2.0	20.1	±	2.7	1.0	±	0.3
U/W sed 16	567.4	378.8	364.3	10.3	±	1.9		<	0.7	10.4	±	1.9	20.8	±	2.7	1.0	±	0.3
U/W sed 17	460.9	327.1	313.6	12.0	±	2.2		<	0.8	10.2	±	2.0	22.7	±	3.0	1.2	±	0.3
U/W sed 18	482.6	330.6	315.3	13.7	±	2.3		<	0.7	13.4	±	2.3	27.4	±	3.3	1.0	±	0.2
U/W sed 19	486.0	336.3	325.5	15.0	±	2.5	1.0	±	0.5	12.8	±	2.2	28.8	±	3.4	1.2	±	0.3
U/W sed 20	480.4	324.9	308.4	13.1	±	2.4		<	0.8	13.6	±	2.4	27.3	±	3.4	1.0	±	0.2
U/W sed 21	469.8	315.8	298.4	17.3	±	2.9		<	0.7	15.9	±	2.7	33.8	±	4.0	1.1	±	0.3
U/W sed 22	546.0	295.4	280.8	14.5	±	2.4	0.8	±	0.4	14.4	±	2.4	29.7	±	3.4	1.0	±	0.2
U/W sed 23	514.0	306.0	291.4	14.1	±	2.4		<	0.6	12.9	±	2.3	27.5	±	3.3	1.1	±	0.3
U/W sed 24	544.2	283.8	268.1	12.9	±	2.3		<	0.7	12.5	±	2.3	25.5	±	3.3	1.0	±	0.3
U/W sed 25	502.5	306.7	291.9	15.9	±	2.6	0.8	±	0.4	14.7	±	2.4	31.4	±	3.6	1.1	±	0.3

Table 7: Underwater sediment samples, total uranium and isotopic ratio results - October 2007.

Note: Activity results have been rounded to 1 decimal place. A specific activity for DU of 14.0 MBq/kg has been used. All uncertainties are stated at a 95% confidence level. Limits of Detection (LOD) are calculated by a 'modified Currie' formula<sup>8</sup> at 95%. The total activity is calculated from the sum of the actual activities for each isotope, regardless of the LOD quoted for that isotope, hence in Table 7, where activities are reported as less than LOD for any of the uranium isotopes, the total uranium value may not be equal to the sum of the individual isotopic values.

<sup>&</sup>lt;sup>8</sup> Revisiting Currie - how low can you go? by Hurtgen C, Jerome S, Woods M. Applied Radiation and Isotopes 53 pp 45-50 (2000) Dstl/CR31252 V1

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Sample Descriptor	Wet weight	Dry weight	Ashed weight		Measured Activity of Dry Sample (mBq/g)									Ratio of $238_{\rm LL}/234_{\rm LL}$				
	(g)	(g)	(g)		<sup>238</sup> U			<sup>235</sup> U			<sup>234</sup> U		]	Fotal U	J		U	
Seaweed 2	202.1	60.0	18.3	6.3	±	2.1		<	1.4	8.0	±	2.4	14.6	Ŧ	3.2	0.8	±	0.4
Seaweed 3	200.9	43.5	11.6	5.4	±	1.5		<	0.9	5.1	±	1.5	10.7	Ħ	2.1	1.1	±	0.4
Seaweed 4	195.6	47.9	12.6	9.6	±	2.2		<	0.9	9.4	±	2.2	19.2	Ħ	3.1	1.0	±	0.3
Seaweed 5	253.8	52.4	13.0	7.8	±	1.6		<	0.6	8.3	±	1.7	16.5	Ħ	2.4	0.9	±	0.3
Seaweed 6	208.9	41.8	11.8	5.6	±	1.6		<	0.9	6.4	±	1.7	12.2	Ħ	2.3	0.9	±	0.3
Seaweed 7	187.6	57.5	11.7	12.2	±	2.0		<	0.4	13.0	±	2.1	25.6	Ħ	2.9	0.9	±	0.2
Seaweed 8	192.6	55.6	12.4	4.1	±	1.9		<	1.7	8.7	±	3.0	13.1	Ħ	3.6	0.5	±	0.3
Seaweed 9	202.2	48.7	9.7	6.7	±	2.0		<	1.0	7.3	±	2.2	14.2	Ħ	3.0	0.9	±	0.4
Seaweed10	177.4	56.1	10.4	7.9	±	1.2	0.4	±	0.2	9.4	±	1.4	17.6	±	1.8	0.9	±	0.2
Seaweed11	178.4	51.0	11.1	5.0	±	1.0		<	0.3	6.4	±	1.2	11.5	±	1.5	0.8	±	0.2

Table 8: Seaweed samples, total uranium and isotopic ratio results – October 2007.

Note: Activity results have been rounded to 1 decimal place. A specific activity for DU of 14.0 MBq/kg has been used. All uncertainties are stated at a 95% confidence level. Limits of Detection (LOD) are calculated by a 'modified Currie' formula<sup>9</sup> at 95%. The total activity is calculated from the sum of the actual activities for each isotope, regardless of the LOD quoted for that isotope, hence in Table 8, where activities are reported as less than LOD for any of the uranium isotopes, the total uranium value may not be equal to the sum of the individual isotopic values.

<sup>&</sup>lt;sup>9</sup> Revisiting Currie - how low can you go? by Hurtgen C, Jerome S, Woods M. Applied Radiation and Isotopes 53 pp 45-50 (2000) Page 28 of 44

Sample Descriptor	Wet weight	Dry weight	Ashed weight		Measured Activity of Dry Sample (mBq/g)									Ratio of				
	(g)	(g)	(g)		<sup>238</sup> U			<sup>235</sup> U		<sup>234</sup> U		Total U		U		07	U	
Mussel 3	79.1	22.0	2.3	2.2	±	0.4		<	0.1	2.5	±	0.4	4.8	±	0.5	0.9	±	0.2
Mussel 4	97.3	25.2	2.4	4.0	±	0.6	0.2	±	0.1	4.8	±	0.7	8.9	±	0.9	0.8	±	0.2
Mussel 5	98.0	20.9	3.1	5.3	±	0.8	0.3	±	0.1	6.3	±	0.9	11.9	±	1.2	0.9	±	0.2
Mussel 8	151.0	40.8	5.9	2.7	±	0.4	0.1	±	0.1	3.3	±	0.5	6.1	±	0.6	0.8	±	0.2
Mussel 10	114.5	28.3	2.3	4.4	±	0.6	0.3	±	0.1	4.8	±	0.6	9.4	±	0.9	0.9	±	0.2
Whelk A	122.8	13.9	1.0	0.4	±	0.1		<	0.1	0.6	±	0.2	1.0	±	0.2	0.6	±	0.3
Whelk B	123.5	9.6	1.0		<	0.4		<	0.2	0.4	±	0.2	0.8	±	0.3		N/A	
Whelk C	117.3	14.2	1.0		$\vee$	0.3		<	0.2	0.3	±	0.1	0.5	±	0.2		N/A	
Whelk D	120.4	12.4	0.8		$\vee$	0.6		$\langle$	0.3		<	0.4	0.4	±	0.2		N/A	
Whelk E	91.7	20.6	1.3		$\langle$	0.8		<	0.4		<	0.4	0.5	±	0.3		N/A	
Whelk F	98.7	21.9	1.5	0.4	±	0.1		<	0.1	0.5	±	0.1	0.9	±	0.2	0.9	±	0.4
Whelk G	77.1	13.5	1.3	0.6	±	0.2		<	0.2	0.6	±	0.2	1.2	±	0.3	1.1	±	0.6
Whelk H	73.7	17.3	1.2		<	0.5		<	0.3	0.4	±	0.2	0.7	±	0.3		N/A	
Crab A	148.2	16.4	1.7		<	0.8		<	0.4		<	0.5	0.4	±	0.3		N/A	
Crab B	150.9	19.4	2.0	0.5	±	0.2		<	0.1	0.3	±	0.1	0.8	±	0.2	1.6	±	0.8
Crab C	148.0	18.7	2.0		<	0.7		<	0.4		<	0.4	0.5	±	0.3		N/A	
Lobster A	96.8	12.5	0.9		$\vee$	0.2		<	0.1		<	0.1	0.1	±	0.1		N/A	
Lobster B	96.1	11.0	0.7		$\vee$	0.2		<	0.1		<	0.1	0.1	±	0.1		N/A	
Lobster C	93.6	11.1	0.7		<	0.3		<	0.2		<	0.2	0.1	±	0.1		N/A	
Lobster D	97.2	11.1	0.8		<	0.9		<	0.4		<	0.5	0.2	±	0.2		N/A	

Table 9: Marine biota samples, total uranium and isotopic ratio results - October 2007.

Note: Activity results have been rounded to 1 decimal place. A specific activity for DU of 14.0 MBq/kg has been used. All uncertainties are stated at a 95% confidence level. Limits of Detection (LOD) are calculated by a 'modified Currie' formula<sup>10</sup> at 95%. The total activity is calculated from the sum of the actual activities for each isotope, regardless of the LOD quoted for that isotope, hence in Table 9, where activities are reported as less than LOD for any of the uranium isotopes, the total uranium value may not be equal to the sum of the individual isotopic values.

<sup>&</sup>lt;sup>10</sup> Revisiting Currie - how low can you go? by Hurtgen C, Jerome S, Woods M. Applied Radiation and Isotopes 53 pp 45-50 (2000) Dstl/CR31252 V1

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Appendix A Historical data

Table 10: Summary of marine results reported for 1996-2007.

Sample Type	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007
Shore sediment	11.9-38.1 ( <b>10</b> ) (0.9-1.1)	14.3-19.0 ( <b>10</b> ) (0.8-1.2)	12.2-31.8 (10) (0.6-1.2)	11.0-32.6 ( <b>10</b> ) (0.9-1.1)	2.2-44.7 (10) (0.8-1.2)	$\begin{array}{c} 10.7-31.1 \\ (9) \\ (0.9-1.4) \end{array}$	12.0-31.9 (10) (0.9-1.2)	9.8-714.4 ( <b>10</b> ) (0.8-1.3)	$\begin{array}{c} 4.2-35.0 \\ (11) \\ (0.9-1.3) \end{array}$	9.8-24.5 (11) (0.8-1.3)	7.5-29.9 (11) (0.8-1.3)	6.9-27.6 (11) (0.9-1.1)
Seaweed	(0.7-0.9)	$\begin{array}{c} (0.0 \ 1.2) \\ 9.8-22.7 \\ (9) \\ (0.8-1.1) \end{array}$	$\begin{array}{c} (0.0 \ 1.2) \\ \hline 5.0-19.6 \\ (9) \\ (0.8-1.2) \end{array}$	$\begin{array}{c} (0.3 \ 1.1) \\ \hline 7.8-14.1 \\ (9) \\ (0.8-1.3) \end{array}$	$\begin{array}{c} (0.0 \ 1.2) \\ 1.3 \ -5.1 \\ (9) \\ (0.9 \ -1.1) \end{array}$	$\begin{array}{c} (0.9 \ 1.1) \\ 6.8 - 12.1 \\ (7) \\ (0.8 - 1.3) \end{array}$	$\begin{array}{c} (0.7 - 1.2) \\ \hline 0.2 - 16.7 \\ (9) \\ (0.7 - 1.2) \end{array}$	$\begin{array}{c} (0.0 \ 1.0) \\ 7.2 - 34.2 \\ (9) \\ (0.7 - 0.9) \end{array}$	9.2-22.6 (9) (0.8-1.0)	4.9-18.2 (10) (0.8-1.2)	6.9-16.0 (9) (0.8-1.0)	$\begin{array}{c} (0.5 \ 1.1) \\ \hline 10.7 \ 25.6 \\ (10) \\ (0.5 \ 1.1) \end{array}$
Mussels	6.8-14.9 (8) (0.8-1.1)	(0.0 1.1) 4.3-11.4 (8) (0.8-1.1)	(0.0 1.2) 7.6-12.2 (5) (0.8-1.1)	6.6-11.1 (5) (0.8-1.1)	(0.9 - 1.1) 3.6 - 7.2 (5) (0.8-0.9)	$\begin{array}{c} (0.0 \ 1.0) \\ 2.9-11.6 \\ (7) \\ (0.3-0.9) \end{array}$	6.4-10.5 (5) (0.9-1.0)	(0.7-0.9) 5.6-7.4 (5) (0.7-0.9)	$\begin{array}{c} (0.00 \ 1.00) \\ 4.0-7.8 \\ (4) \\ (0.8-0.9) \end{array}$	$\begin{array}{c} (0.0 \ 1.2) \\ 5.1 - 10.7 \\ (4) \\ (0.9 - 1.0) \end{array}$	2.6-12.6 (7) (0.9-1.1)	(0.6-0.9)
Whelks	×	×	2.5-3.0 (3) (0.8-1.0)	0.2-0.4 ( <b>4</b> ) (0.3-1.3)	0.6-1.3 (5)	2.0-3.7 (8) (0.5-1.4)	$ \begin{array}{r} 1.1-39.4 \\ (8) \\ (0.9 \pm \\ 0.5^{a}) \end{array} $	×	×	0.7-2.9 (6) (0.8-1.6 <sup>°</sup> )	0.1-0.1 (4) ( <i>n/a</i> )	$\begin{array}{c} 0.4-1.2 \\ (8) \\ (0.9-1.1^b) \end{array}$
Scallops	×	2.8-3.5 (4) (0.8-1.0)	2.8-3.2 (3) (0.8-1.1)	0.7-1.1 ( <b>4</b> ) (1.3-1.5)	(0.3-1.6)	2.8-13.3 (8) (0.7-1.6)	×	1.5-33.0 (6) ( <i>n/a</i> )	2.8-3.1 (2) (0.9-9.9)	2.8-13.3 (3) (n/a)	0.1-0.2 (4) ( <i>n/a</i> )	×
Crabs	×	$0.6 \pm 0.3 \\ (1) \\ (0.9 \pm 0.5)$	×	0.3-0.4 (2) (1.5-1.7)	0.2-1.4	1.2-2.3 (2) (0.9-1.0)	$0.8-23.8 \\ (4) \\ (0.9 \pm 0.5^{a})$	0.1-2.3 (3) ( <i>n/a</i> )	×	0.4-1.6 (6) ( <i>n/a</i> )	0.1-0.6 (5) (n/a)	$0.4-0.8 \\ (3) \\ (1.6 \pm 0.8^{a})$
Lobsters	×	×	$0.2 \pm 0.1$ (1) ( <i>n/a</i> )	0.3-0.3 (2) (0.1-0.5)	(0.1-3.7)	2.4-3.2 (4) (0.7-1.6)	1.2-1.8 (2) (n/a)	0.3-3.4 (3) ( <i>n/a</i> )	$0.4 \pm 0.1$ (3) $(1.0 \pm 0.4^{a})$	0.4-0.9 (4) ( <i>n/a</i> )	0.1-0.1 (2) (n/a)	0.1-0.2 (4) ( <i>n/a</i> )
Underwater sediment	22.3-30.0 (6) (0.9-1.2)	22.3-27.2 (6) (0.9-1.1)	21.1-28.0 (6) (0.9-1.1)	15.4-32.1 (6) (0.5-1.1)	0.2-12.8 (6) (0.8-1.3)	19.3-31.0 ( <b>34</b> ) (0.8-1.3)	15.2-30.4 ( <b>33</b> ) (0.9-1.2)	20.2-30.9 (36) (0.9-1.2)	17.0-33.2 (64) (0.8-1.2)	16.3-24.3 (18) (0.9-1.3)	16.6-26.6 (25) (0.8-1.3)	19.5-42.3 (25) (0.8-1.3)

**Notes**: For simplicity, results for both the total uranium activity and the isotopic  ${}^{238}U/{}^{234}U$  ratio are reported as follows:



b: values reported for 3 samples only

- When there are multiple results for a sample type, the range (min max) is reported. The analytical error for these results which are reported in the yearly survey reports varies per sample type. It is typically below 20% for sediment, but can be as high as 95% for biological samples. The number of samples is indicated in bold in brackets.
- Where the isotopic activities are below the Limit of Detection (LOD) for more than one isotope, the isotopic ratios could not be calculated and are reported as n/a.

# ANNEX A Issues to be considered when interpreting or comparing uranium data

A number of issues may give rise to uncertainties when interpreting or comparing uranium data:

- analytical technique and reporting conventions;
- statistical variation;
- spatial variability;
- temporal variability; and
- species variation (for plant and animal samples).

#### Analytical approaches and reporting conventions

Sediment sample results may be reported as either dry weight or wet weight depending on whether the masses of the samples were obtained prior to or after drying. This will have implications for comparison of results between the surveys at Kirkcudbright, which are reported as dry weight and other UK uranium in sediment data, which may be reported as wet weight. Sediment samples reported as dry weight will appear to have concentrations of uranium at least 20% higher than those reported as wet weight (although this will depend on the moisture content).

For analysis techniques such as ICP-MS or AS, uranium may be extracted from samples and into solution by either leaching the sediment samples or totally dissolving them. Total dissolution will give rise to higher uranium results, because the analysis will include all uranium, including that which is contained within the mineral grains, whereas leached samples will only contain uranium that is either easily dissolved or is sorbed to the surfaces of mineral grains. Total sample analysis techniques such as gamma spectrometry will give results similar to those for total dissolution. Given the differences between the results for total analysis and leached analysis, care should be taken when comparing sets of data to ensure that either the same approach has been used or that differences are appropriately discussed.

Uranium concentrations in seaweed may be affected by contamination of surfaces with sediment particles. Preparation of seaweed for analysis may or may not involve a washing stage. Hence, it is important to be aware of the preparation approaches that have been applied when comparing the results of seaweed analyses.

Seaweed and marine biota sample results may be reported as either dry weight or wet weight depending on whether the masses of the samples were obtained prior to or after drying. For marine biota, samples reported as dry weight will appear to have concentrations of uranium higher than those reported as wet weight. The relative increase in uranium concentration will depend on the moisture content, and can be as much as 300% for molluscs (whelks, scallops) [A19].

Dstl/CR31252 V1

In addition, uranium activity (or concentration) results are expressed either as total uranium activity (or concentration), or as a breakdown of the <sup>238</sup>U, <sup>235</sup>U, and <sup>234</sup>U isotopic activities, or are expressed in terms of the  $^{238}$ U activity (or concentration) only<sup>11</sup>.

#### **Statistical variations**

There will be minor variations between the true uranium content of a sample and that reported by an analytical technique. This variation is highlighted in the counting statistics for the technique. The statistical uncertainties of laboratory results are likely to be small in comparison with the true variation in activity between samples.

#### **Spatial variations**

Any DU contamination will not be uniformly distributed within a sampling area, but will depend on the surface water flow, and the movement of tides and sediment. Hence, the repeat sampling and analysis of sediment from within an area may give rise to a significant degree of variation.

In addition to DU contamination due to firing at the KTA, there may be variations in uranium concentrations due to local anthropogenic or natural discharges. For example, natural uranium concentrations may be enhanced by the local application of phosphate based fertilisers to agricultural land. Veins rich in uranium minerals occur naturally along the coast of the Solway Firth, such as uraninite found at Needle's Eye, approximately 24 km away on the north coast of the estuary. These features are thought to be present on a more regional basis, although this has not been studied [A20, A21].

#### **Temporal variations**

There will be natural temporal variations in the uranium concentration and the abundance of the various isotopes in the seawater due to seasonal variations in rainfall. Rainfall can impact on the dissolution of uranium, and its migration in surface waters, which eventually enter the sea in coastal regions.

The activities of samples from any particular sampling site may vary from year to year. This may relate to temporal changes in uranium concentration, but will also be affected by spatial variation. (See above.)

Marine plant uptake of radionuclides is affected by the period in the plant growing cycle. This is also mirrored in the animal uptake of radionuclides within their life cycle.

#### **Species variations**

Plant uptake of radionuclides is affected by the substrate characteristics (uranium concentration and speciation as well as other physico-chemical characteristics) and varies with plant species.

<sup>&</sup>lt;sup>11</sup> As seen in Section 3, there are three broad types of uranium (natural, enriched and depleted). An increase or decrease in <sup>238</sup>U value (either concentration or activity) reflects a corresponding increase or decrease in the total uranium value. Within each type of uranium (i.e. for samples of identical uranium isotopic mix), the change in <sup>238</sup>U is proportional to the change in total U. Across all types however, the change in <sup>238</sup>U is not proportional to the change in total U, because each type has a different isotopic concentration and activity breakdown. Instead such a change is merely a qualitative indicator of a change in total U. Dstl/CR31252 V1

Animal uptake of uranium is affected by their life habits, feeding patterns, physiology and the uranium concentration in their foodstuffs and the environment. The marine biota that have been sampled were chosen for their importance in the human food chain. Biota that live in intertidal fringes have been selected to provide samples representative of a diet of sea food gathered by consumers. In addition, biota that live on the sea bed were selected for their relevance to a diet of local sea food purchased by consumers in the local Kirkcudbright area.

Mass of depleted	Ac	tivity <sup>13</sup> (mBq	/kg)	Ratio of Total Activity Natural	<sup>238</sup> U/ <sup>234</sup> U Activity Ratio		
uranium added <sup>12</sup>	<sup>238</sup> U	<sup>235</sup> U	<sup>234</sup> U	Activity	Activity Ratio		
0	3.7 x10 <sup>4</sup>	1.7 x10 <sup>3</sup>	3.8 x10 <sup>4</sup>	1.0	1.0		
1	7.4 x10 <sup>4</sup>	2.2 x10 <sup>3</sup>	4.3 x10 <sup>4</sup>	1.6	1.7		
2	1.1 x10 <sup>5</sup>	2.7 x10 <sup>3</sup>	4.8 x10 <sup>4</sup>	2.1	2.3		
3	1.5 x10 <sup>5</sup>	3.2 x10 <sup>3</sup>	5.3 x10 <sup>4</sup>	2.6	2.8		
4	1.9 x10 <sup>5</sup>	3.7 x10 <sup>3</sup>	5.8 x10 <sup>4</sup>	3.2	3.2		
5	2.2 x10 <sup>5</sup>	4.1 x10 <sup>3</sup>	6.3 x10 <sup>4</sup>	3.8	3.6		
6	2.6 x10 <sup>5</sup>	4.6 x10 <sup>3</sup>	6.8 x10 <sup>4</sup>	4.3	3.9		
7	3.0 x10 <sup>5</sup>	5.1 x10 <sup>3</sup>	7.2 x10 <sup>4</sup>	4.9	4.1		
8	3.4 x10 <sup>5</sup>	5.6 x10 <sup>3</sup>	7.7 x10 <sup>4</sup>	5.4	4.3		
9	3.7 x10 <sup>5</sup>	6.0 x10 <sup>3</sup>	8.2 x10 <sup>4</sup>	6.0	4.5		
10	4.1 x10 <sup>5</sup>	6.5 x10 <sup>3</sup>	8.7 x10 <sup>4</sup>	6.5	4.7		
20	7.8 x10 <sup>5</sup>	1.1 x10 <sup>4</sup>	1.4 x10 <sup>5</sup>	12.0	5.8		
60	2.3 x10 <sup>6</sup>	3.1 x10 <sup>4</sup>	3.3 x10⁵	34.1	6.9		
80	3.0 x10 <sup>6</sup>	4.0 x10 <sup>4</sup>	4.3 x10 <sup>5</sup>	45.1	7.1		
100	3.8 x10 <sup>6</sup>	5.0 x10 <sup>4</sup>	5.2 x10 <sup>5</sup>	56.1	7.2		
200	7.5 x10 <sup>6</sup>	9.8 x10 <sup>4</sup>	1.0 x10 <sup>6</sup>	111.0	7.4		
600	2.2 x10 <sup>7</sup>	2.9 x10 <sup>5</sup>	3.0 x10 <sup>6</sup>	332.0	7.6		
800	3.0 x10 <sup>7</sup>	39 x10 <sup>5</sup>	3.9 x10 <sup>6</sup>	442.0	7.6		
1000	3.73 10 <sup>7</sup>	4.8 x10 <sup>5</sup>	4.9 x10 <sup>6</sup>	552.0	7.6		

## ANNEX B Change in the <sup>238</sup>U/<sup>234</sup>U activity ratio of a medium containing natural uranium with the addition of depleted uranium

Table from Volume 2 - Appendices, WS Atkins Environmental Assessment on DU Firings.

<sup>&</sup>lt;sup>12</sup> The value represents the additional mass of depleted uranium added (all radionuclides) relative to the original mass of natural uranium present (3 mg U/kg soil)

<sup>&</sup>lt;sup>13</sup> Table assumes 3 mg/kg of natural uranium in a medium, prior to addition of DU, and present in the following proportion: <sup>238</sup>U (2.978 mg U/kg); <sup>235</sup>U (0.022 mg U/kg); <sup>234</sup>U (2e-04 mg U/ kg). Dstl/CR31252 V1

# ANNEX C Variability of uranium concentration and uranium isotopic ratios in marine environmental samples

There are few specific examples that demonstrate the variability of uranium concentrations and isotopic ratios within marine environmental samples. To understand these, it is useful to consider the flux of uranium between the various components of the marine environment.

Uranium occurs naturally in seawater and its concentration generally varies in proportion to salinity. It is present in open seawater at an average concentration of  $82.5 \pm 5$  Bq/m<sup>3</sup>, with a  $^{238}$ U/ $^{234}$ U activity ratio of  $0.88 \pm 0.03$  at a salinity of 35 ‰. The isotopic ratio of seawater is below unity, as  $^{234}$ U is preferentially mobilised from the soil during chemical weathering, thus enhancing its presence in seawater [A24, A22]. The physical mixing of low U- river water <sup>14</sup> and high U- sea water in estuary environments generally results in a dilution of uranium in the estuarine waters. As a result uranium concentrations in estuarine water are mostly lower than open ocean values, and increase linearly with salinity [A24].

Concentrations of uranium in marine sediment are variable (from 32.5 to 1,625 mBq/g dry weight) and vary primarily as a function of the geology of the area.

Sample type	Uranium concentration	Typical <sup>238</sup> U/ <sup>234</sup> U ratio
Ocean water	82.5 Bq/m <sup>3</sup>	0.88
Estuarine water	<82.5 Bq/m <sup>3</sup>	<0.88
Marine sediment ( <sup>238</sup> U only)	32.5-1,625 mBq/g	0.81

Table 11: Typical uranium concentrations and isotopic ratios in marine samples [A23, A24, A25].

Levels of uranium also vary depending on the precise location and depth of the sediment sample, as these directly affect its chemical characteristics, and hence its potential for concentrating uranium from the marine waters [A24, A25]. Both low oxygenation<sup>15</sup> and low salinity<sup>16</sup> provide favourable conditions for uranium scavenging from the water column by a variety of processes. These include the precipitation of U-rich colloids into the low salinity zone and the reduction of uranium into insoluble forms [A25, A26].

<sup>&</sup>lt;sup>14</sup> Concentrations of uranium in rivers vary considerably, with carbonate and dissolved solids concentrations, with an average of 7.5-15 Bq/m<sup>3</sup>, and an isotopic <sup>238</sup>U/<sup>234</sup>U activity ratio of 0.77-0.83 as <sup>234</sup>U is preferentially mobilised during chemical weathering.

<sup>&</sup>lt;sup>15</sup> Low oxygenation is found with increasing depth and increasing organic content

<sup>&</sup>lt;sup>16</sup> Low salinity is dependent on river rate of flow, proximity to river outflow and depth of water. Page 38 of 44

# ANNEX D Reference values for uranium in the Solway Firth and the UK

For UK coastal sediment, <sup>238</sup>U concentrations generally range from 3.6 and 32.3 mBq/g of dry weight [A26]. Values reported for shore sediment uranium concentrations in the Solway Firth vary with location and the specific uranium rich geology of the area:

Values for <sup>238</sup>U, <sup>235</sup>U and <sup>234</sup>U for sediment collected at Rascarrel Bay in 2005 were reported by CEFAS as 4.7, 0.39 and 5.4 mBq/g of dry weight respectively.

Values reported for <sup>238</sup>U concentrations at Sandyhills Bay, in southwest Scotland, were lower than worldwide average values for seawater and sediment reported in ANNEX C. Seawater and sediment from Sandyhills Bay had <sup>238</sup>U concentrations of  $40 \pm 3$  Bq/m<sup>3</sup> and  $14 \pm 0.4$  mBq/g dry weight<sup>17</sup>, respectively [A27]. No other uranium isotope was measured, so no indication could be provided for the isotopic ratio.

However, semi-quantitative analysis of shore sediment samples obtained from the mudflats offshore from the uraninite vein, located approximately 24 km east from KTA at Needle's Eye, yielded uranium values of up to several hundred parts per million. These activities were measured both around open, oxygenated root channels and in near surface peat material. This analysis has suggested that uranium disperses seawards from the vein and ground waters, and is retarded in this by organic material in the sediment [A21].

Sample	Location	Activity per wet weight (mBq/g)	Activity ( <sup>238</sup> U)per dry weight (mBq/g)
Seaweed	[A27] Sandyhills Bay		$3.8 \pm 0.1$
	[A27] UK		3.8 - 18.6
Mussel	[A27] Sandyhills Bay		$1.1 \pm 0.1$
	[A27] UK		1.01 – 37.1
	[A27] Sandyhills Bay		$2.72 \pm 0.01$
	(winkle)		1.36 - 18.9
Mollusc	[A27] UK [17] UK (molluse & winkle)	0.89	
	[17] Parton (winkle)	1.5	
Crab	[17] UK	0.046	
	[17] Parton	0.063	
Lobster	[17] UK	0.035	
	[17] Parton	0.034	

Table 12: Reported <sup>238</sup>U concentrations in seaweed and marine biota in the UK.

 <sup>&</sup>lt;sup>17</sup> Sample preparation for the sediment sample included drying of the wet sample, grinding, homogenising, followed by acid leaching, electroplating and ICP-MS analysis for U.
 Dstl/CR31252 V1
 Page 39 of 44

Literature values reported for <sup>238</sup>U in seaweed and marine biota samples are shown in Table 12 for the Solway Firth area; also reported are estimated values of <sup>238</sup>U from natural sources in aquatic foodstuff for the UK given by CEFAS [17, A27].

Uranium-238 values reported for seaweed and marine biota at Sandyhills Bay were: <sup>238</sup>U concentrations of  $3.8 \pm 0.1$  mBq/g dry weight for seaweed (*F. vesiculosus*) and  $1.1 \pm 0.1$  mBq/g dry weight for mussel (Mussels) [A27]. In 2005, <sup>238</sup>U concentrations for marine samples procured in Parton, near Whitehaven, ranged from 0.034 mBq/g wet weight for lobster, to 0.063 mBq/g wet weight for crab [A17]. However, it should be noted that the uranium inventory at Parton is dominated by historical anthropogenic input of natural uranium from a local phosphate processing plant.

#### **ANNEX E** List of references to annexes

- [A19] Correspondence from Mr G Hunt (CEFAS) to Mr Phillips (MoD Directorate of Safety, Environment and Fire Policy), 23 February 2001.
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10. Date of Issue	11. Paginatio	n	12. No. of References					
14 <sup>th</sup> November 2008	44		27					
13. Abstract (A brief (approximately 1	50 words) fact	ual summary of the report	:)					
This report describes and interprets the results of the marine environmental survey undertaken to monitor DU levels in the environment at KTA in 2007. Samples of intertidal sediment, seaweed and seafood were collected from the shoreline in the local area. Underwater sediment samples were collected offshore from the KTA, and samples of locally fished, bottom dwelling crustaceans and molluscs were also obtained. The results of the 2007 survey agree with those from previous years and do not indicate any health or environmental impact from the DU firings. There is no evidence to indicate that members of the public or site employees are, or have ever been, exposed to a radiological hazard from the marine environment as a result of test firing DU ammunition at the site.								
14. Abstract Protective Marking inclue	ding any Cave	ats						
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Kirkcudbright Training Area, Environr	nental Survey,	Depleted Uranium, DU, N	larine Environment. 2007					

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