

Evidence

Survey of gamma dose rates in air around the Esk Estuary related to radioactivity levels in sediments

Report: SC060063/R3

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Miranda Kavanagh

Director of Evidence

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Executive summary

Over the last 30 years the Sellafield site has reduced the amount of radioactivity it discharges in liquid effluent, and radionuclide activity concentrations in the environment around Sellafield have declined. Monitoring results from the nearby Esk Estuary show a less clear trend, with activity concentrations of some radionuclides fluctuating from year to year, and fluctuations in the total gamma dose rates measured. This is thought to be due to the dynamic nature of sediment distribution in the estuary: sediment is redistributed under the influence of the prevailing tidal and freshwater conditions, and older, deeper, more contaminated sediments are exposed or buried.

The objectives of the study were to:

- assess the current external gamma radiation dose rates in the Esk estuary; and
- assess changes in the measured dose rates relative to a survey of the estuary that was undertaken in 1989.

Scientists from the University of Liverpool's Institute for Sustainable Water Integrated Management and Ecosystem Research (SWIMMER) completed the fieldwork over a 14-day period between July and August 2007. The spatial coverage was comparable with that of the 1989 survey and a total of 576 gamma dose rate measurements were made around the estuary. Samples of surface soil were collected for particle size analysis (PSA) at 60 of the locations where the dose rate was measured. Nine sediment cores were collected from three locations close to Muncaster Bridge, to allow the radionuclide depth distribution to be determined and the relative contribution of natural and anthropogenic radionuclides to be calculated.

The study found that external gamma dose rates within the Esk Estuary had decreased significantly between 1989 and 2007. The total measured dose rate range in 1989 was $0.070 - 0.608 \,\mu\text{Gy}\,\,\text{h}^{-1}$ with a mean of $0.232 \,\mu\text{Gy}\,\,\text{h}^{-1}$; in 2007 this had reduced to $0.064 - 0.235 \,\mu\text{Gy}\,\,\text{h}^{-1}$ with a mean of $0.123 \,\mu\text{Gy}\,\,\text{h}^{-1}$.

The depth at which the highest concentration of anthropogenic radionuclides was found, and the lack of correlation between measured dose rates and surface sediment particle size, provides evidence suggesting that part of the reason for the reduction in measured dose rates is the burial of contaminated sediment deposits by uncontaminated sediments transported by tidal processes.

The maximum exposure for a member of the public was calculated to be 0.200 µSv h⁻¹ by applying a conversion coefficient of 0.85 Sv Gy⁻¹ to the highest total dose rate recorded in the 2007 survey of 0.235 µGy h⁻¹. This was measured at a location within the inner Esk Estuary. A member of the public would need to be present at the maximum exposure location for 57% of the time to exceed the 1 mSv annual dose limit from this external exposure pathway alone. However, this calculation is based on a total (measured) dose rate, which includes cosmic radiation, anthropogenic and natural radioactivity contributions, whereas the 1 mSv statutory annual dose limit applies to exposures from anthropogenic sources only. Even considering the contribution of other pathways such as radionuclide intake through food, it seems very unlikely that the annual dose limit for individuals living near the Esk Estuary will be exceeded.

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1 Introduction

Estuaries are the dynamic aquatic interface between freshwater and marine environments. They often become sinks for fine sediments from both freshwater and marine waters due to the complex interplay of physical and chemical processes that occur when freshwater rivers meet tidal saltwater. Particle reactive contaminants tend to be deposited in association with fine sediments, so estuaries also become sinks for a range of environmental contaminants including radionuclides. Given the range of commercial and recreational activities that are supported by estuaries, it is important to understand how contaminants accumulate, and to be able to assess risks to humans and the environment through source-pathway-receptor linkages.

The Esk Estuary (West Cumbria, England) is a recognised sink for radionuclides (Emptage & Kelly, 1990). The estuary has received radionuclide inputs from the nearby Sellafield complex via regulated discharges of radioactive waste into the marine environment. Other sources of radionuclides are from weapons testing and nuclear accidents (from direct inputs and from surface water run-off from land where radionuclides were deposited).

Over the last 30 years the Sellafield site has reduced the amount of radioactivity it discharges in liquid effluent (Figure 1.1), and radionuclide activity concentrations in the environment around Sellafield have declined (RIFE, 1996 - 2008). Monitoring results from the nearby Esk Estuary show a less clear trend, with activity concentrations of some radionuclides fluctuating from year to year (e.g. RIFE, 2008) and fluctuations in the total gamma dose rates measured (Table 1.1; RIFE, 1996 - 2008). This is thought to be due to the dynamic nature of sediment distribution in the estuary: sediment is redistributed under the influence of the prevailing tidal and freshwater conditions, and older, deeper, more contaminated sediments are exposed or buried.

The objectives of this study were to:

- assess the current external gamma radiation dose rates in the Esk estuary; and
- assess changes in the measured dose rates relative to a survey of the estuary that was undertaken in 1989.

This report presents the findings of work undertaken by scientists from the University of Liverpool's Institute for Sustainable Water Integrated Management and Ecosystem Research (SWIMMER).

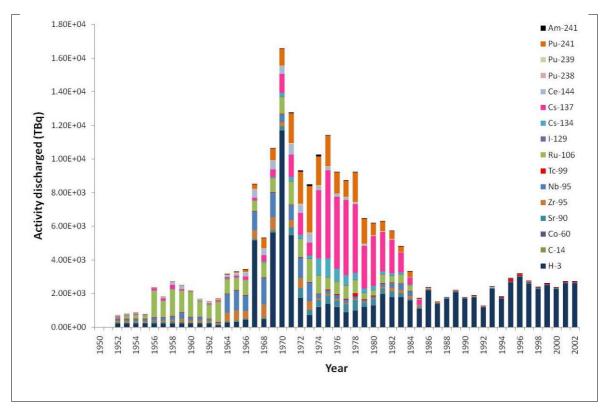


Figure 1.1 Temporal variation in the radionuclide activity profile of the total annual discharge to the marine environment from the Sellafield complex (data from Jackson et al., 2000 and BNFL annual monitoring reports)

Table 1.1 Temporal variation in total gamma dose rates (μGy h⁻¹) measured at locations around the Esk Estuary (Data from RIFE 1996 – 2008)

| Location and ground type | 1995 | 1996 | 1997 | 1998 | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 |
|------------------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Carleton Marsh salt marsh & mud | - | - | - | - | - | - | - | - | 0.160 | 0.130 | 0.160 | 0.150 | 0.160 |
| Carleton Marsh grass & mud | - | - | - | - | - | - | - | - | 0.100 | 0.160 | 0.160 | 0.160 | 0.160 |
| Carleton Marsh salt marsh | 0.260 | 0.250 | 0.250 | 0.230 | 0.220 | 0.200 | 0.180 | 0.170 | 0.180 | 0.175 | 0.160 | 0.140 | 0.150 |
| River Mite estuary grass & mud | - | - | - | - | - | - | - | - | 0.160 | 0.140 | - | 0.180 | 0.180 |
| River Mite estuary salt marsh | 0.230 | 0.230 | 0.210 | - | - | - | - | 0.200 | 0.200 | 0.170 | 0.150 | 0.180 | 0.180 |
| Raven Villa grass & mud | 0.120 | 0.130 | 0.130 | 0.130 | 0.120 | 0.120 | 0.110 | 0.100 | 0.160 | 0.160 | 0.140 | 0.150 | 0.150 |
| Raven Villa salt marsh | 0.240 | 0.220 | 0.210 | - | 0.180 | 0.180 | 0.170 | 0.165 | 0.170 | 0.155 | 0.170 | 0.160 | 0.150 |
| Boat area sand | 0.065 | 0.071 | 0.069 | - | - | - | - | - | 0.092 | 0.100 | 0.100 | 0.100 | 0.110 |
| Boat area pebbles & sand | - | - | - | - | - | - | - | 0.110 | 0.110 | 0.100 | 0.100 | 0.099 | 0.100 |
| Boat area mud & sand | 0.092 | 0.096 | 0.095 | 0.110 | - | - | - | - | - | - | - | - | - |
| Ford mud & pebbles | - | - | - | - | - | - | - | - | - | 0.120 | 0.110 | 0.098 | 0.110 |
| Ford sand | 0.110 | 0.110 | 0.100 | 0.110 | 0.110 | 0.100 | 0.100 | 0.102 | 0.120 | 0.101 | 0.110 | 0.120 | 0.110 |
| Muncaster Bridge grass & mud | - | - | - | - | - | - | - | - | - | 0.130 | 0.130 | 0.120 | 0.120 |
| Muncaster Bridge grass | - | - | - | - | - | - | - | 0.130 | 0.130 | 0.140 | 0.130 | 0.120 | 0.120 |
| Muncaster Bridge salt marsh | 0.220 | 0.240 | 0.230 | 0.200 | - | - | - | - | 0.120 | - | - | - | - |
| Salmon Garth mud & pebbles/sand | 0.140 | 0.130 | 0.120 | 0.120 | 0.120 | 0.120 | 0.110 | 0.110 | 0.110 | - | - | 0.110 | 0.110 |
| Salmon Garth mussel bed | 0.092 | 0.095 | 0.092 | 0.097 | 0.094 | 0.088 | 0.083 | 0.082 | 0.085 | 0.100 | 0.110 | 0.110 | 0.100 |
| Salmon Garth pebbles & sand | 0.086 | 0.091 | 0.093 | 0.093 | 0.091 | 0.089 | 0.089 | 0.084 | 0.106 | 0.086 | - | 0.110 | 0.100 |
| Salmon Garth pebbles & stones | - | - | - | - | - | - | - | - | 0.097 | 0.099 | - | 0.098 | 0.100 |
| Eskmeals Nature Reserve mud | - | - | - | - | - | - | - | - | - | 0.160 | 0.098 | 0.100 | 0.110 |
| Eskmeals Nature Reserve salt marsh & mud | - | - | - | - | - | - | - | - | - | 0.160 | 0.150 | 0.120 | 0.140 |
| Eskmeals Nature Reserve salt marsh | 0.250 | 0.250 | 0.220 | - | - | - | - | - | - | 0.150 | - | 0.120 | 0.130 |
| Carleton Marsh salt marsh & mud | - | - | - | - | - | - | - | - | 0.160 | 0.130 | 0.160 | 0.150 | 0.160 |
| Carleton Marsh grass & mud | - | - | - | - | - | - | - | - | 0.100 | 0.160 | 0.160 | 0.160 | 0.160 |

Notes: - indicates that no result was reported in RIFE

2 Methodology

2.1 Field study location and sampling strategy

The Esk Estuary is located on the west-Cumbrian coastline, UK (Ordnance Survey National Grid Reference: SD 082 963). It is approximately 10 km south of the Sellafield nuclear fuel reprocessing plant and 2 km south of the low-level radioactive waste repository operated by Low-level Repository Ltd. The estuary is at the confluence of the rivers Irt, Mite and Esk, and it discharges into the Irish Sea through a narrow (approximately 400 m wide) channel between the Drigg and Eskmeals sand dune ranges (Figure 2.1).

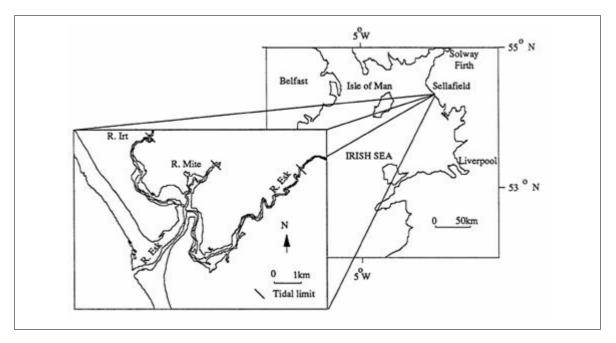


Figure 2.1 Location of the Esk Estuary survey area (reproduced from Kelly & Emptage, 1991)

The 1989 survey of gamma dose rates around the Esk Estuary (Emptage & Kelly, 1990) was based on an irregular grid pattern with transects orientated perpendicular to the low-water channel. The layout of the grid was established using a 1:25000 scale map before the start of the survey. Precise measurement locations were decided by the surveyors at the time of measurement, to ensure coverage of all major sediment facies within the estuary.

The monitoring plan for the 2007 survey was developed to allow the results to be compared with those from the 1989 survey. Ordnance Survey National Grid References for the measurement locations used in the 1989 survey were entered into a Geographical Information System (GIS) software package (ArcGIS) and overlaid onto a base map of the Esk Estuary (Figure 2.2). This map was used to define the spatial extent of the survey to be undertaken in 2007 (Figure 2.3), with the aim of ensuring representative measurement coverage across all areas of the estuary that were included in the 1989 survey. The zones of the estuary (labelled a – e in Figures 2.2)

and 2.3) correspond to: (a) Irt Estuary; (b) Mite Estuary; (c) outer Esk estuary; (d) middle Esk Estuary; and (e) inner Esk Estuary.

The 2007 survey of the estuary was undertaken over 14 days between July and August, and 576 gamma dose rate measurements were made. The 1989 survey was predominantly undertaken in June and included 869 gamma dose rate measurements followed by 21 additional measurements taken in May 1990 from a small area of the River Esk, upstream of Muncaster Bridge (SD 113 964). The 1989 and 2007 surveys are therefore broadly comparable, both spatially and seasonally.

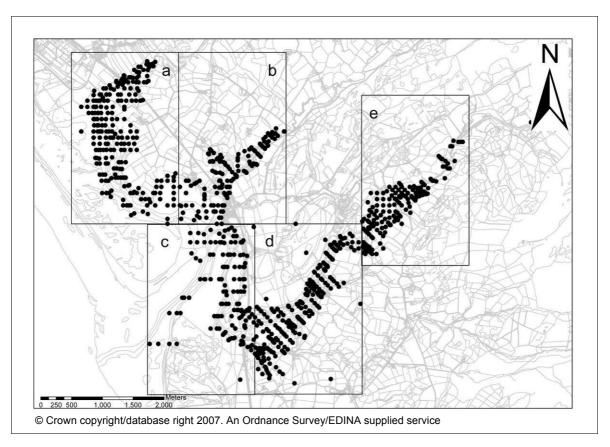


Figure 2.2 Measurement locations for the 1989 survey showing the five zones of the estuary (a – e) that were used for data comparison

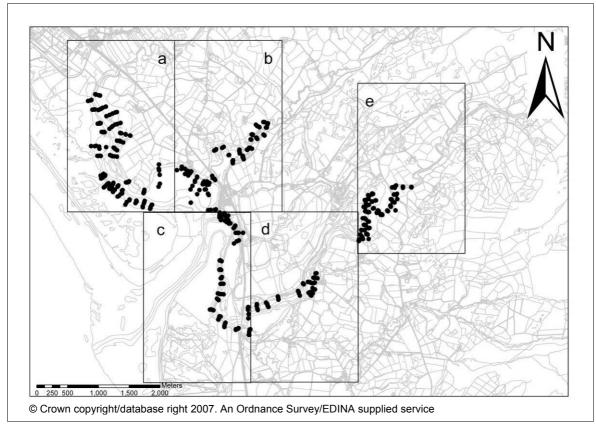


Figure 2.3 Measurement locations for the 2007 survey showing the five zones of the estuary (a – e) that were used for data comparison

2.2 *In situ* measurement of gamma dose rates

Gamma dose rates in air (µGy h⁻¹) are numerically equivalent to gamma air kerma rates (HMIP, 1995); air kerma rate being a measure of the radiation energy that is absorbed per unit mass of air over a given time period (*kerma* is an acronym for kinetic energy released per unit mass). Therefore, gamma dose rates in air can be calculated from *in situ* measurements of gamma air kerma rates. This is the approach that was used to determine gamma dose rates in the Esk Estuary during the 1989 survey (Emptage & Kelly, 1990) and is the standard approach adopted for many routine monitoring and survey applications (e.g. RIFE, 2008).

2.2.1 Field measurement

Measurements of gamma ray air kerma rates were made *in situ* using a Mini-Instruments Environmental Monitor Type 6-80 fitted with an MC71 Geiger-Muller (G-M) tube, which is energy compensated for gamma radiation entering radially (Saint-Gobain Crystals & Detectors UK LTD, 2000). Thus, to detect contamination immediately below the monitoring position it may be preferable to mount the energy compensated tube in the horizontal plane. However, to enable comparison with other surveys, the standard methodology described in HMIP (1995) was followed.

The MC71 G-M tube was mounted vertically on a light-weight low attenuation tripod (to ensure rotational axis symmetry) and the G-M tube centre positioned at a height of 1 m (±0.2 m) above the sediment surface. This provides a field of view of approximately 8m radius (200 m²) at 662 keV for a typical exponential decrease in specific activity with depth (Tyler et al., 1996). A count time of 600 seconds was used, in order to ensure detection of a minimum of 700 counts (HMIP, 1995), and the total counts per second (counts s⁻¹) measured over the counting period were recorded. Measurements were not made during heavy rainfall, to reduce the likelihood of radon progeny washout influencing the results (HMIP, 1995), or immediately after heavy rainfall when moisture in the surface sediments may attenuate the terrestrial gamma photons (Thompson et al., 1999). For the same reason, measurements immediately after exposure of sediments following tidal inundation were avoided.

To enable maximum coverage of the estuary within the 14 day survey period, two Mini-Instruments Environmental Monitor Type 6-80s with MC71 G-M tubes were used. Both instruments were calibrated by VT Nuclear Services (United Kingdom Accreditation Service (UKAS) accredited laboratory number 0542) using a ²²⁶Ra source to determine instrument-specific radium calibration factors (Table 2.1) at air kerma rates between 1 and 3 μGy h⁻¹. During calibration a ¹³⁷Cs source was also used to derive a conversion coefficient for situations in which anthropogenic radionuclides are the dominant contributor to air kerma rate. However, previous work on the relative contribution of natural and anthropogenic radionuclides to measured external gamma dose rate in the Esk Estuary suggests that the anthropogenic radionuclide contribution is approximately 30% of the total dose rate measured (McDonald et al., 2005). Therefore, the ²²⁶Ra conversion coefficient was considered to be the most appropriate for use in this study and is in line with the approach adopted for the 1989 survey (Emptage & Kelly, 1990).

To ensure that there was no significant difference in the performance of each instrument, duplicate measurements were made at 10 locations covering a total gamma dose rate range of $0.08-0.24~\mu Gy~h^{-1}$. A Shapiro-Wilk test confirmed normality of the measurement data (P >0.05) and the Levene test confirmed

homogeneity of variances (P > 0.10). A paired-sample t-test confirmed no significant difference (P > 0.05) between the means of the measurements from the two instruments and the linearity of comparative performance of the instruments across the dose rate range for the survey was confirmed by regression analysis ($r^2 = 0.93$). It was concluded that the response of the two instruments was comparable across the gamma dose rate range encountered within the survey and no systematic bias needed to be accounted for in processing the resultant survey data. Fixed point measurements were performed daily at a non-tidal location to check for instrument drift or meteorological influences, and the measurements were stable throughout the survey period.

Table 2.1 Characterisation of instruments used for the Esk Estuary survey in

| Instrument | Mini-Instruments Environmental Monitor Type 6-80 (Serial No. FN 0000141) with an MC71 G- M tube (Serial No. 1736) | Mini-Instruments Environmental Monitor Type 6-80 (Serial No. FN 0000140) with an MC71 G-M tube (Serial No. 1738) |
|---------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------|
| K ¹ (²²⁶ Ra source) | 18.6 | 18.7 |
| K ¹ (¹³⁷ Cs source) | 14.3 | 14.6 |
| P^{2} (counts s ⁻¹) | 1.158 ± 0.066^3 | 1.081 ± 0.043 ⁴ |
| Intrinsic detector background (counts s ⁻¹) | 0.2 | 0.2 |

Notes: ¹ Instrument-specific calibration factor to convert counts s^{-1} to air kerma rate (µGy h^{-1}); ² Sum of cosmic and intrinsic detector count rates (counts s^{-1}); ³ Mean \pm standard deviation (n = 5); ⁴ Mean \pm standard deviation (n = 7)

2.2.2 Calculation of gamma dose rates

That calculations described in this section are for calculating gamma air kerma rates but, as noted in the previous section, gamma air kerma rates are numerically equivalent to gamma dose rates. Therefore, to simplify the terminology, the remainder of this report refers to gamma dose rates rather than air kerma rates.

There are different sources of radiation that have to be considered when calculating gamma dose rates. The total gamma dose rate measured by an instrument includes intrinsic radiation (from the material used to construct the detector), cosmic radiation (from space), and radiation from natural and anthropogenic radionuclides in the environment. The anthropogenic component is usually the quantity of interest but, as it is not possible to measure this directly, this quantity has to be calculated.

To avoid confusion in the terms used here to describe gamma dose rate, they are defined as follows:

• Measured total gamma dose rate ($D_{I,C,N_t,A}$) is to the sum of the dose rates from intrinsic (I), cosmic (C), natural terrestrial (Nt) and anthropogenic (A) radiation. This is the measurement represented by the number of counts that the monitoring equipment records.

- Total gamma dose rate ($D_{C,N_{t,A}}$) is the sum of the dose rates from all sources external to the instrument. It is the measured total gamma dose rate ($D_{I,C,N_{t,A}}$) with the intrinsic radiation contribution subtracted.
- Terrestrial gamma dose rate ($D_{Nt,A}$) is the sum of the dose rates from terrestrial radiation, both natural and anthropogenic. It is the total gamma dose rate ($D_{CNt,A}$) with the cosmic radiation contribution subtracted.
- Anthropogenic gamma dose rate (D_A) is the does rate from anthropogenic radiation only. It is the terrestrial gamma dose rate ($D_{Nt,A}$) with the natural radiation component subtracted.

Different studies adopt different reporting approaches, some report total gamma dose rate ($D_{C,Nt,A}$) and some report terrestrial gamma dose rate ($D_{Nt,A}$). To facilitate comparison with other surveys, both quantities are reported in this study.

The following equation is used to calculate the terrestrial gamma dose rate (D_{NtA}):

$$D_{Nt,A} = \frac{N - P}{K}$$

Where $D_{\scriptscriptstyle Mr,A}$ is the terrestrial gamma dose rate (μ Gy h⁻¹); N is the measured count rate (counts s⁻¹); P is the sum of cosmic and intrinsic detector count rates (counts s⁻¹); and K is the instrument-specific calibration factor for converting counts s⁻¹ to gamma dose rate in air (μ Gy h⁻¹).

A typical value for P is 1.00 counts s⁻¹ (0.2 counts s⁻¹ due to the intrinsic detector background and 0.8 counts s⁻¹ due to the contribution of cosmic radiation (HMIP, 1995; Thompson et al., 1999)). However, these background values are location and instrument specific so, to determine the appropriate value of P to use for each detector, measurements were performed over Esthwaite water (Ordnance Survey National Grid Reference: SD 359 969), a large water-body in Cumbria with a water depth in excess of 10 m and remote from surrounding high ground. Measurements were taken from a plastic boat positioned close to the middle of Esthwaite water to ensure that the majority of counts detected would be due to the intrinsic detector background and cosmic radiation contributions alone (Ambrosi, 2009). Esthwaite water is approximately 65 m above sea-level. Whilst it is recognised that gamma dose rate increases with altitude, the difference in the cosmic contribution to measured gamma dose rate between sea-level and 65 m elevation is minimal (Thompson et al., 1999). The P values determined at Esthwaite water are presented in Table 2.1. The mean cosmic and intrinsic detector background was 0.059 \pm 0.004 μ Gy h⁻¹ (n = 12).

To calculate the total gamma dose rate ($D_{C,Nt,A}$), the assumed intrinsic detector background (0.2 counts s⁻¹) was used in place of P in the equation given above.

2.3 Radionuclide distribution in sediment

2.3.1 Determining radionuclide depth distribution

In order to investigate the relationship between radionuclide activity concentration depth profile in estuarine sediments/soils and measured gamma dose rates, three cores were collected from each of three locations around Muncaster Bridge (SD 113 964) where a range (0.055 μ Gy h⁻¹ – 0.113 μ Gy h⁻¹) of terrestrial gamma dose rates ($D_{N_{I,A}}$) had been measured. Location 1 was on the north bank of the Esk, on the east side of the bridge. Locations 2 and 3 were on the west side of the bridge - Location 2 on the north bank and Location 3 on the south bank.

The sediment/soil surface was cleared of vegetation and stones. Any surface litter was removed by gently scraping it away with a spatula. Cores were taken to 40cm depth using a stainless-steel split-blade corer and sectioned in the field into the following depth sections using a stainless-steel knife: 0-5 cm, 5-10 cm, 10-15 cm, 15-20 cm, 20-30 cm and 30-40cm. Core sections were transferred to labelled plastic sample bags, double-bagged and packed into a cool box to maintain sample integrity during transport to the laboratory.

Soil samples were dried at 80°C to constant mass, passed through a 2 mm sieve to remove debris and ground using a 'Grinder La Minervia' rotary mill to produce a homogenised uniform sample matrix. The samples were then packed into either 330 ml Marinelli beakers or 150 ml plastic containers (depending on the sample size) and analysed on EG & G Ortec hyper-pure germanium gamma detectors. The detectors were calibrated for the energy range 30 keV to 2,000 keV and the spectra were analysed for a suite of naturally occurring and Sellafield-relevant anthropogenically-derived radionuclides. Typical count times ranged from 7 h to 24 h depending on the activity of the sample; the count times for each sample being long enough to make sure that there were sufficient counts in the main peaks for the radionuclides to be quantified.

Radionuclides known to be potentially significant contributors to gamma dose rates in air were quantified and are reported, specifically those radionuclides with a kerma rate per unit activity per unit mass (µGy h⁻¹ per Bq kg⁻¹) greater than 0.04 based on the values reported by the International Commission on Radiological Units and Measurements (ICRU, 1994). These were ²⁰⁸Tl and ²²⁸Ac from the ²³²Th series, ²¹⁴Bi from the ²³⁸U series, ⁴⁰K, ⁶⁰Co and ¹³⁷Cs. Results were also reported for ²⁴¹Am. Although not a significant contributor to gamma dose rates in air, due to its low-energy gamma photons (59 keV), ²⁴¹Am is an important component of the Sellafield discharge and, as a particle reactive actinide, its environmental behaviour is similar to that of Pu. ²⁴¹Am is also generated *in situ* by ingrowth from ²⁴¹Pu. Isotopes of Pu, especially ²³⁹Pu and ²⁴¹Pu, have been significant contributors to the total annual activity discharged from Sellafield (Figure 1.1) but these two isotopes are principally alpha and beta emitters respectively, and so they are not detected by the gamma spectrometry technique. Therefore, the ²⁴¹Am data provide an indication of the distribution of other actinides in the estuary.

2.3.2 Estimating external gamma dose rates from measured activity concentrations

Measured activity concentrations were used to estimate terrestrial gamma dose rates ($D_{Nt,A}$) at 1 m above the sediment surface using the ICRU methodology (ICRU, 1994).

For natural radionuclides a uniform distribution was assumed. The mean activity concentration was calculated for each natural radionuclide and multiplied by the appropriate ICRU conversion factor (Table 2.2). For anthropogenic radionuclides, a non-uniform depth was assumed and the estimation of the terrestrial gamma dose rate contribution was obtained by calculating β , the relaxation mass per unit area, and using this to derive a core-specific conversion factor as described in the ICRU methodology.

Table 2.2 ICRU conversion factors for natural radionuclides homogenously distributed with depth

| Radionuclide | Kerma rate per unit activity per unit mass (μGy h ⁻¹ per Bq kg ⁻¹) |
|--------------------------------|-------------------------------------------------------------------------------------------|
| ²²⁸ Ac ¹ | 0.221 |
| ²¹⁴ Bi ² | 0.401 |
| 40 K | 0.0417 |
| ²⁰⁸ TI ¹ | 0.326 |

Notes: ¹Radionuclide from the ²³²Th series; ²Radionuclide from the ²³⁸U series

2.4 Particle size analysis of estuarine sediments

At 60 of the dose rate measurement locations, samples of surface soil were collected for particle size analysis (PSA). The locations (Figure 2.4) were selected to ensure representative coverage of the geographical extent of the survey area and of the various sediment facies. PSA samples were collected to 5cm depth using a stainless-steel Eijkelkamp corer with a sample collection ring diameter of 5 cm. Samples were transferred to labelled plastic sample bags, double-bagged and packed into a cool box to maintain sample integrity during transport to the laboratory.

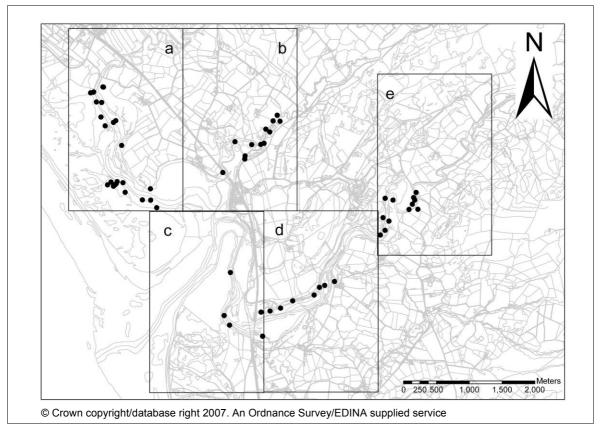


Figure 2.4 Locations for which particle size analysis was undertaken as part of the 2007 survey

Given the potential for fine particulates to comprise a large proportion of the sediment composition in estuarine environments, PSA was performed using a wet-sieving method. Each core was split in half longitudinally. One half was used for PSA and the other retained for archive purposes. The PSA samples were mixed for 12 hours in an end-over-end shaker with 10% sodium hexametaphosphate dispersant solution (CalgonTM). Following this they were wet sieved through a sieve stack, consisting of four sieves (2 mm, 600 μ m, 212 μ m and 63 μ m) and a sample collection container, which were mechanically agitated for 600 seconds. This method enabled the following particle size fractions to be determined based on the Wentworth classification scheme (Wentworth, 1922): Coarse sand (600 μ m – 2 mm), Medium sand (212 μ m – 600 μ m), Fine sand (63 μ m – 212 μ m) and Silt (< 63 μ m).

2.5 Data handling

2.5.1 Preparation of data

At locations where both instruments were used to take a measurement, the arithmetic mean of the two measurements was calculated and used as the representative gamma dose rate for that location.

The 1989 survey reported measured total gamma dose rates ($D_{I,C,Nt,A}$) (Emptage & Kelly, 1990). However, a measured value for the cosmic and intrinsic detector background at the time of the 1989 survey is also reported (0.049 \pm 0.006 μ Gy h⁻¹, n =

2). Terrestrial gamma dose rates ($D_{Nt,A}$) were estimated in the 2007 survey, the purpose of which was to investigate the temporal change in gamma dose rates in the estuary due to changes in sediment activity concentrations. To enable direct comparison between the two surveys, 0.049 μ Gy h⁻¹ was subtracted from the measurements reported by Emptage & Kelly to convert the measured total gamma dose rate ($D_{LC,Nt,A}$) to terrestrial gamma dose rate ($D_{Nt,A}$).

2.5.2 Statistical analysis of dose rate data

Statistical analyses were performed using SPSS 16.0 for Windows (Release 16.0.1) to determine whether there were significant differences between the 1989 and 2007 survey data for the estuary as a whole and for each zone of the estuary (Figure 2.3). Exploratory analyses were used to quantify the basic parameters of the data sets (n, arithmetic mean, standard deviation and range) as well as test for normality of data distribution (Shapiro-Wilk test) and homogeneity of variance (Levene's test). Analyses were performed on raw data, log transformed and square root transformed data. For each data set comparison, the outcome of the exploratory analyses was used to inform the choice of statistical procedure to test the null hypothesis that there was no significant difference between the 1989 and 2007 survey data sets.

In all cases, neither the raw or transformed data met the assumptions for the application of parametric statistical techniques. Therefore, the Mann-Whitney *U*-Test (a non-parametric technique) was used to test the hypothesis that there was no significant difference between the 1989 and 2007 measurements for each data set comparison.

3 Results

3.1 Estuary-level comparison

The total ($D_{C,N_{I},A}$) and terrestrial ($D_{N_{I},A}$) gamma dose rate data for the 1989 and 2007 surveys are summarised in Table 3.1. The reduction in dose rates between 1989 and 2007 was highly statistically significant (Mann-Whitney U-Test, P < 0.01). Although dose rates in the estuary had declined since 1989, the areas of the estuary with elevated dose rates were broadly comparable between the 1989 and 2007 surveys (Figure 3.1).

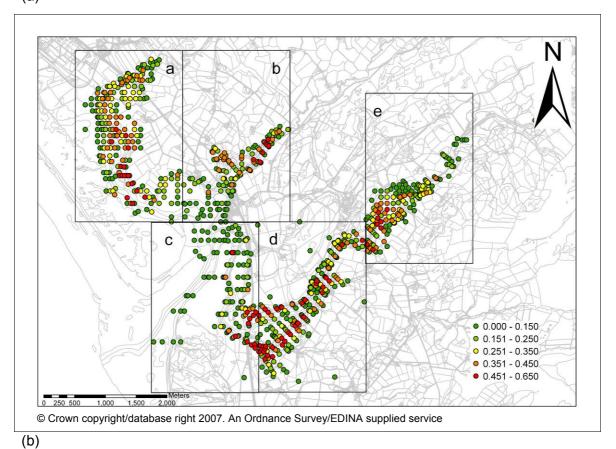
Table 3.1 Summary statistics for the estuary-level comparison of the 1989 and 2007 surveys

| Year of | Statistic | Dose rate (μGy h ⁻¹) | | | |
|---------|-----------|----------------------------------------|-----------------------------------------------|--|--|
| survey | | Total gamma dose rate $(D_{C.Nt,A})^1$ | Terrestrial gamma dose rate ($D_{N_{t,A}}$) | | |
| | | C,NI,A | | | |
| 1989 | n | 890 | 890 | | |
| | Mean | 0.232 | 0.183 | | |
| | Median | 0.178^2 | 0.129^2 | | |
| | SD | 0.135 | 0.135 | | |
| | Min | 0.070 | 0.021 | | |
| | Max | 0.608 | 0.559 | | |
| 2007 | n | 576 | 576 | | |
| | Mean | 0.123 | 0.073 | | |
| | Median | 0.115^2 | 0.066^2 | | |
| | SD | 0.034 | 0.034 | | |
| | Min | 0.064 | 0.016 | | |
| | Max | 0.235 | 0.186 | | |

Notes: 1 The 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; 2 Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney *U*-Test, P < 0.01)

When making comparisons between surveys which report external gamma dose rates, it is important to understand any differences in the approach used to convert the measured count rate (counts s⁻¹) to dose rate. Decisions made regarding the value of P and the handling of intrinsic detector background when reporting gamma dose rates can make a notable difference to the results obtained (Figure 3.2). For example, applying a correction for intrinsic detector background (assumed to contribute 0.2 counts s⁻¹) when reporting total gamma dose rate ($D_{C,Nt,A}$) can reduce the reported values by 0.011 μ Gy h⁻¹. Emptage & Kelly (1990) did not apply this correction to the 1989 survey data. However, even if the 1989 summary data are reduced by 0.011 μ Gy h⁻¹, they are still notably higher than the dose rates measured in 2007.

(a)



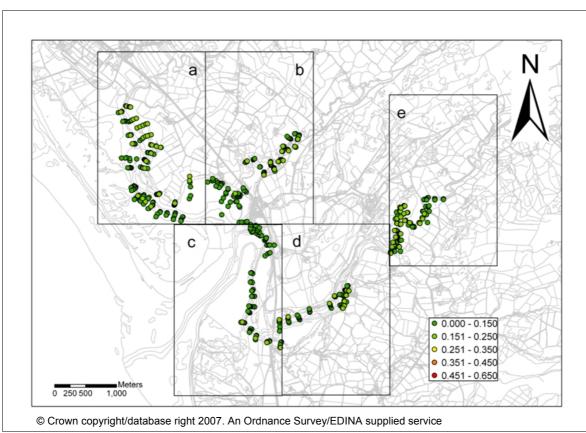


Figure 3.1 Total gamma dose rate measurements ($D_{C,Nt,A}$) (µGy h⁻¹) at the Esk Estuary in (a) 1989 and (b) 2007

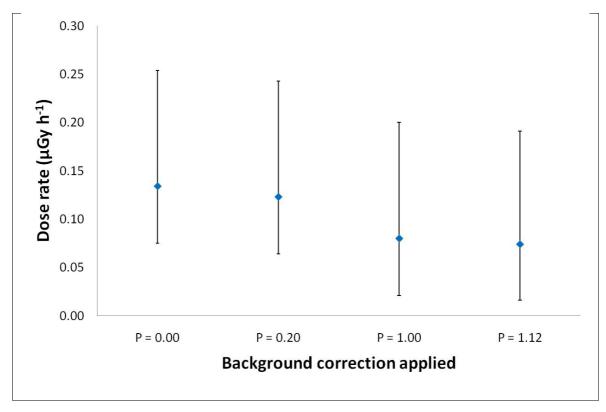


Figure 3.2 Effect of P value selection on calculated dose rate

3.2 Esk Estuary zones

3.2.1 Zone A (Irt Estuary)

In the Irt Estuary, there was a highly statistically significant decrease in the median total $(D_{C,Nt,A})$ and terrestrial $(D_{Nt,A})$ dose rates between the 1989 and 2007 surveys (Mann-Whitney *U*-Test, P < 0.01). Total dose rate $(D_{C,Nt,A})$ ranges were 0.072 – 0.602 μ Gy h⁻¹ in the 1989 survey and 0.064 – 0.227 μ Gy h⁻¹ for the 2007 survey (Table 3.2). Although there were fewer measurement locations in the 2007 survey the areas of elevated dose rates appeared broadly comparable (Figure 3.3).

Table 3.2 Summary statistics for the Esk Estuary Zone A comparison of the 1989 and 2007 surveys

| Year of | Statistic | Dose rate (μGy h ⁻¹) | | | |
|---------|-----------|----------------------------------|--------------------------------------------|--|--|
| survey | | Total gamma dose rate | Terrestrial gamma dose | | |
| | | $(D_{C,N_{t,A}})^{1}$ | $rate\ (D_{{\scriptscriptstyle Nt},A}^{})$ | | |
| 1989 | n | 228 | 228 | | |
| | Mean | 0.244 | 0.195 | | |
| | Median | 0.218^2 | 0.169^2 | | |
| | SD | 0.129 | 0.129 | | |
| | Min | 0.072 | 0.023 | | |
| | Max | 0.602 | 0.553 | | |
| 2007 | n | 160 | 160 | | |
| | Mean | 0.124 | 0.075 | | |
| | Median | 0.115^2 | 0.067^2 | | |
| | SD | 0.039 | 0.039 | | |
| | Min | 0.064 | 0.016 | | |
| | Max | 0.227 | 0.175 | | |

Notes: 1 The 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; 2 Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney *U*-Test, P < 0.01)

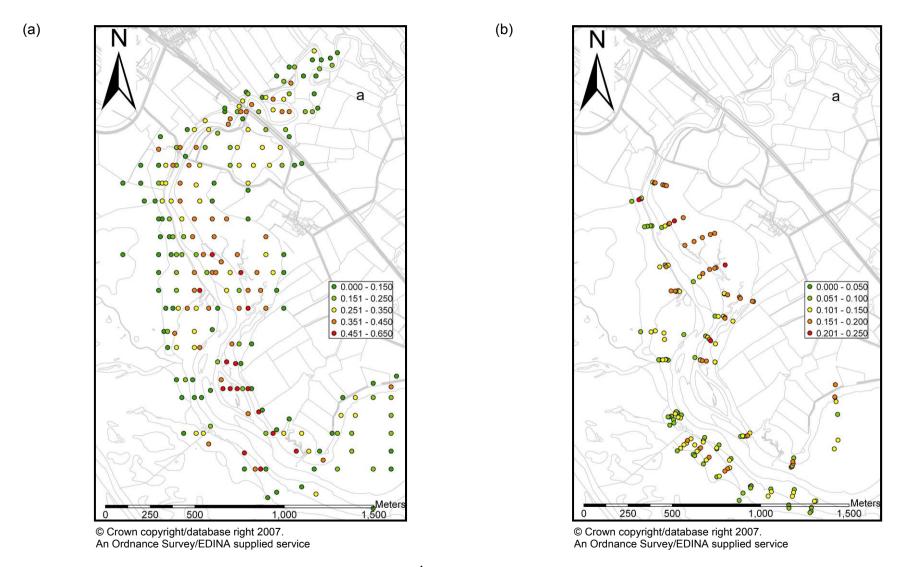


Figure 3.3 Total gamma dose rate measurements (μGy h⁻¹) in Zone A of the Esk Estuary in (a) 1989 and (b) 2007 (N.B. the ranges of dose rates, and the associated colour codes, differ between the two diagrams)

3.2.2 Zone B (Mite Estuary)

The total gamma dose rate ($D_{C,N_{t,A}}$) range in the Mite Estuary was 0.078 – 0.530 µGy h⁻¹ in 1989 and 0.068 – 0.198 µGy h⁻¹ in 2007 (Table 3.3). There was a highly statistically significant decrease in the median total ($D_{C,N_{t,A}}$) and terrestrial ($D_{N_{t,A}}$) dose rates between the 1989 and 2007 surveys (Mann-Whitney *U*-Test, P < 0.01). The areas of elevated dose rates appeared broadly comparable between the two surveys (Figure 3.4), with higher dose rates being measured in the upper (more inland) part of the Mite Estuary.

Table 3.3 Summary statistics for the Esk Estuary Zone B comparison of the 1989 and 2007 surveys

| Year of survey | Statistic | Dose rate (μ Gy h ⁻¹) Total gamma dose rate $(D_{C,Nt,A})^1$ | Terrestrial gamma dose rate ($D_{\mathit{Nt,A}}$) |
|----------------|-----------|----------------------------------------------------------------------------------|-----------------------------------------------------|
| 1989 | n | 134 | 134 |
| | Mean | 0.224 | 0.175 |
| | Median | 0.183^2 | 0.134^2 |
| | SD | 0.122 | 0.122 |
| | Min | 0.078 | 0.029 |
| | Max | 0.530 | 0.481 |
| 2007 | n | 124 | 124 |
| | Mean | 0.120 | 0.071 |
| | Median | 0.109^2 | 0.062^2 |
| | SD | 0.035 | 0.035 |
| | Min | 0.068 | 0.017 |
| | Max | 0.198 | 0.149 |

Notes: 1 the 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; 2 Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney U-Test, P < 0.01)

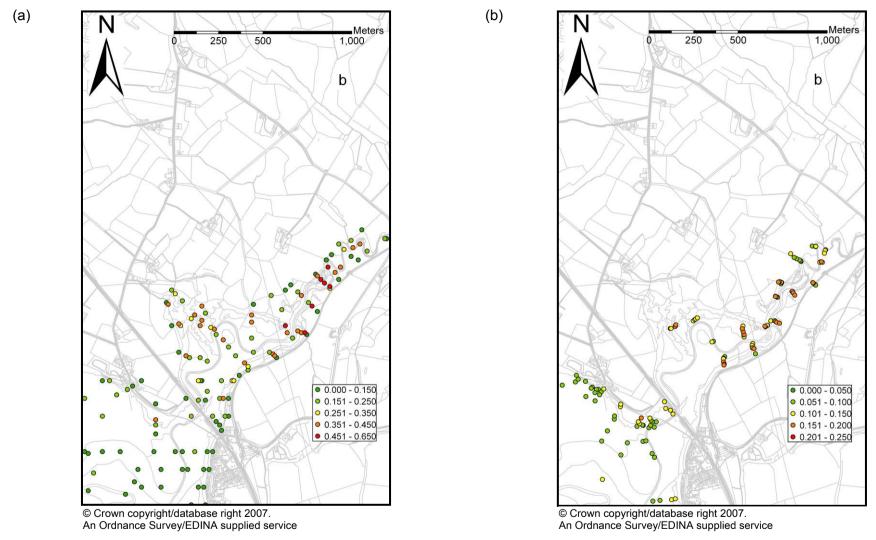


Figure 3.4 Total gamma dose rate measurements (μGy h⁻¹) in Zone B of the Esk Estuary in (a) 1989 and (b) 2007 (N.B. the ranges of dose rates, and the associated colour codes, differ between the two diagrams)

3.2.3 Zone C (outer Esk Estuary)

A Mann-Whitney *U*-Test confirmed a highly statistically significant (P < 0.01) decline in both the total ($D_{C,N_{I},A}$) and terrestrial ($D_{N_{I},A}$) median dose rates between 1989 and 2007 for the outer Esk Estuary. In 1989 the total dose rate ($D_{C,N_{I},A}$) range was 0.070 – 0.608 μ Gy h⁻¹ whereas the range in 2007 was 0.072 – 0.196 μ Gy h⁻¹ (Table 3.4).

The spatial coverage of the outer Esk Estuary was more constrained in the 2007 survey (Figure 3.5) with fewer measurements being taken (n = 137 for 1989 and n = 90 for 2007). Although the 2007 survey did not extend into the channel area between the Eskmeals and Drigg coastal sand dunes, this is the narrow channel through which the Esk Estuary exchanges water with the Irish Sea. As a result, this is likely to be an energetic section of the estuary with relatively high water velocities and minimal fine sediment accretion, assumptions which are supported by the low terrestrial dose rates ($D_{Nt,A}$) recorded for this channel area in 1989. Therefore, it is unlikely that the more constrained coverage of the outer Esk Estuary in the 2007 survey has resulted in areas of high dose rates being missed within the channel area. For the section of the outer Esk Estuary covered by both surveys, there is good agreement in the areas in which elevated dose rates were recorded.

Table 3.4 Summary statistics for the Esk Estuary Zone C comparison of the 1989 and 2007 surveys

| Year of survey | Statistic | Dose rate (µGy h ⁻¹) Total gamma dose rate | Terrestrial gamma dose | |
|----------------|-----------|-----------------------------------------------------------|------------------------|--|
| | | $(D_{C,Nt,A})^1$ | rate $(D_{Nt,A})$ | |
| 1989 | n | 137 | 137 | |
| | Mean | 0.185 | 0.136 | |
| | Median | 0.124^2 | 0.075^2 | |
| | SD | 0.136 | 0.136 | |
| | Min | 0.070 | 0.021 | |
| | Max | 0.608 | 0.559 | |
| 2007 | n | 90 | 90 | |
| | Mean | 0.105 | 0.056 | |
| | Median | 0.099^2 | 0.051^2 | |
| | SD | 0.023 | 0.024 | |
| | Min | 0.072 | 0.024 | |
| | Max | 0.196 | 0.149 | |

Notes: 1 The 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; 2 Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney *U*-Test, P < 0.01)

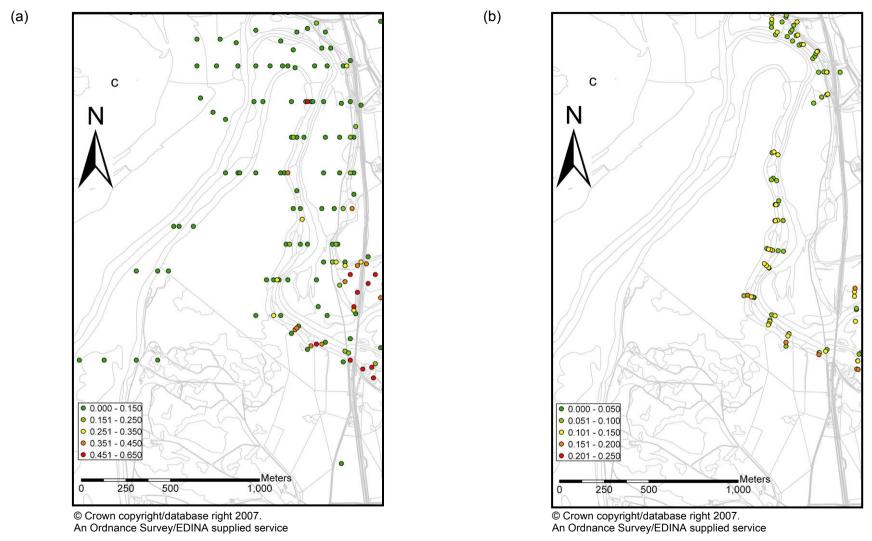


Figure 3.5 Total gamma dose rate measurements (μGy h⁻¹) in Zone C of the Esk Estuary in (a) 1989 and (b) 2007 (N.B. the ranges of dose rates, and the associated colour codes, differ between the two diagrams)

3.2.4 Zone D (middle Esk Estuary)

Spatial coverage of the middle Esk Estuary was also more constrained in the 2007 survey than in the 1989 survey (Figure 3.6). This was due to problems gaining access permissions for the south bank area in particular. However, it is evident that locations with elevated dose rates are comparable between 1989 and 2007 in the survey area that is common to both surveys.

In 1989, the total dose rate ($D_{C,N_{I},A}$) range was 0.086 – 0.606 μ Gy h⁻¹. This had reduced to 0.080 – 0.204 μ Gy h⁻¹ in 2007. The reduction in median dose rates was highly statistically significant (Mann-Whitney *U*-Test, P < 0.01), confirming that, as with other sections of the estuary, there has been a general reduction in dose rates over time.

Table 3.5 Summary statistics for the Esk Estuary Zone D comparison of the 1989 and 2007 surveys

| Year of survey | Statistic | Dose rate (μ Gy h ⁻¹) Total gamma dose rate $(D_{C,Nt,A})^1$ | Terrestrial gamma dose rate ($D_{\mathit{Nt,A}}$) |
|----------------|-----------|----------------------------------------------------------------------------------|-----------------------------------------------------|
| 1989 | n | 222 | 222 |
| | Mean | 0.260 | 0.211 |
| | Median | 0.199^2 | 0.150^2 |
| | SD | 0.151 | 0.151 |
| | Min | 0.086 | 0.037 |
| | Max | 0.606 | 0.557 |
| 2007 | n | 59 | 59 |
| | Mean | 0.133 | 0.084 |
| | Median | 0.129^2 | 0.081^2 |
| | SD | 0.034 | 0.035 |
| | Min | 0.080 | 0.028 |
| | Max | 0.204 | 0.156 |

Notes: ¹ The 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; ² Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney *U*-Test, P < 0.01)

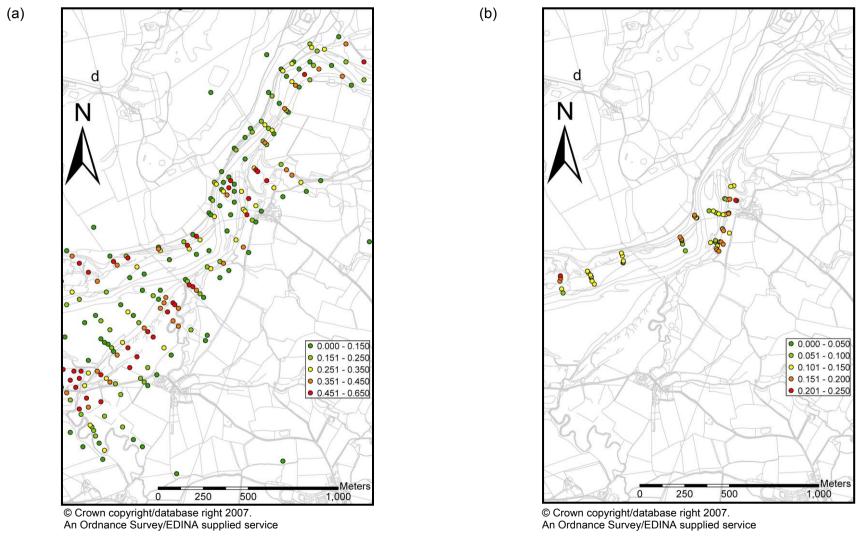


Figure 3.6 Total gamma dose rate measurements (μGy h⁻¹) in Zone D of the Esk Estuary in (a) 1989 and (b) 2007 (N.B. the ranges of dose rates, and the associated colour codes, differ between the two diagrams)

3.2.5 Zone E (inner Esk Estuary)

The inner Esk Estuary spans Muncaster Bridge and incorporates the area used for collecting the cores used to investigate the depth distribution of gamma emitting radionuclides within the sediment of the Esk Estuary (see Sections 2.3 and 3.3). There was good agreement between the areas of elevated dose rates determined in the 1989 survey and those identified in the 2007 survey (Figure 3.7).

A highly statistically significant decrease in median total dose rates ($D_{C,Nt,A}$) was observed between the 1989 and 2007 surveys (Mann-Whitney *U*-Test, P < 0.01). In 1989 the total dose rate range was 0.090 – 0.543 μ Gy h⁻¹ whereas the 2007 survey range was 0.079 – 0.235 μ Gy h⁻¹ (Table 3.6).

Table 3.6 Summary statistics for the Esk Estuary Zone E comparison of the 1989 and 2007 surveys

| Year of survey | Statistic | Dose rate (μGy h ⁻¹) | | | |
|----------------|-----------|--------------------------------------------------------------|---------------------------------------------------|--|--|
| | | Total gamma dose rate $\left(D_{C,\mathit{Nt},A}\right)^{1}$ | Terrestrial gamma dose rate $(D_{\mathit{Nt,A}})$ | | |
| 1989 | n | 165 | 165 | | |
| | Mean | 0.226 | 0.177 | | |
| | Median | 0.201^2 | 0.152^2 | | |
| | SD | 0.120 | 0.120 | | |
| | Min | 0.090 | 0.041 | | |
| | Max | 0.543 | 0.494 | | |
| 2007 | n | 142 | 142 | | |
| | Mean | 0.130 | 0.081 | | |
| | Median | 0.125^2 | 0.074^2 | | |
| | SD | 0.029 | 0.029 | | |
| | Min | 0.079 | 0.028 | | |
| | Max | 0.235 | 0.186 | | |

Notes: ¹ The 1989 total gamma dose rate data reported by Emptage & Kelly (1990) include the intrinsic radiation contribution; ² Highly statistically significant difference between the 1989 and 2007 median values (Mann-Whitney *U*-Test, P < 0.01)

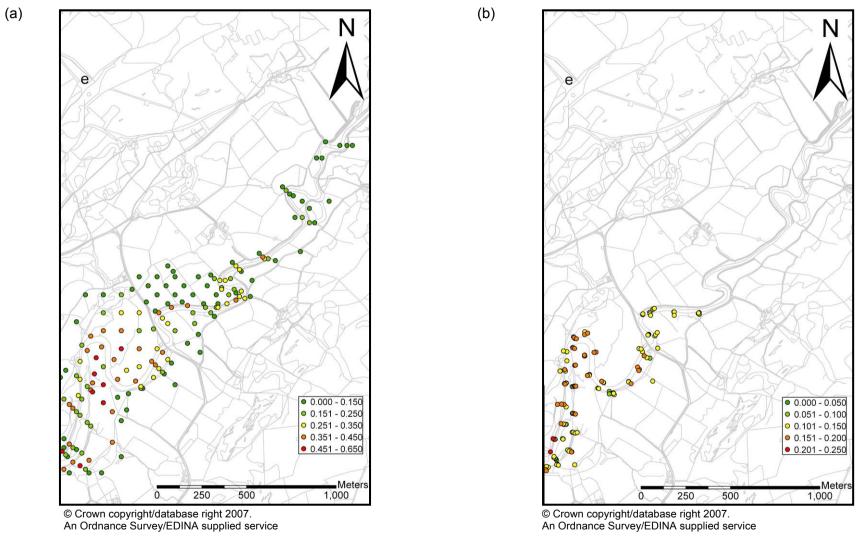


Figure 3.7 Total gamma dose rate measurements (μGy h⁻¹) in Zone E of the Esk Estuary in (a) 1989 and (b) 2007 (N.B. the ranges of dose rates, and the associated colour codes, differ between the two diagrams)

3.3 Sediment activity concentrations and dose rate contributions

There was notable variation in the activity concentrations of the three cores collected at each location, especially in the 241 Am and 137 Cs activity concentrations (Figure 3.8 - 3.13). As would be expected, the depth distribution of natural radionuclides (228 Ac, 214 Bi, 40 K & 208 Tl) was relatively uniform at all three sites, whereas the activity concentrations of anthropogenic radionuclides (241 Am, 137 Cs & 60 Co) varied according to depth. These observations support the approach taken for applying the ICRU methodology (ICRU, 1994) for estimating terrestrial gamma dose rates ($D_{N_{I},A}$) in air from measured soil activity concentrations, that is assuming the depth distribution for natural and anthropogenic radionuclides to be uniform and non-uniform depth respectively. Also, from the 241 Am and 137 Cs activity concentration data, there is evidence that sub-surface maxima existed at the three locations (between 10 and 15 cm at Locations 1 & 3 and between 5 and 10 cm at Location 2), so to assume a classical exponential decrease in activity concentrations with depth would be inappropriate. Calculating β , the relaxation mass per unit area, and using this to determine the conversion factor to apply when estimating the dose rate from anthropogenic radionuclides (see ICRU, 1994) is a more suitable approach for Locations 1 – 3.

The radionuclide activity concentration data for each core were used to estimate terrestrial dose rates ($D_{Nt,A}$) (μ Gy h⁻¹) at 1 m above the soil surface (Table 3.7) and the relative contributions of the radionuclides to the terrestrial dose rate (Table 3.8). With the exception of Core C from Location 3, 137 Cs was dominant, contributing approximately 60 – 70 % to the terrestrial dose rates. Estimated terrestrial dose rates were much higher (approximately 0.030 µGy h⁻¹) than those measured using the in situ monitoring equipment (Table 3.9). However, the in situ-derived dose rates were converted from measured count rates (counts s⁻¹) using the instrument-specific calibration factor (K) for ²²⁶Ra (see Section 2.2.2). This was based on the assumption that natural radionuclides were the dominant contributor to dose rates within the estuary (McDonald et al., 2005). Although this is in line with approach used in the 1989 survey (Emptage & Kelly, 1990), this assumption does not appear valid for Locations 1 - 3. The only core for which this assumption may be correct is Core C from Location 3, for which ¹³⁷Cs is estimated to contribute approximately 30 % of the terrestrial dose rate. Recalculating the measured in situ-derived terrestrial dose rates for the three locations using K for ¹³⁷Cs (see Table 2.1) results in terrestrial dose rates which are in much better agreement with the dose rates estimated from the activity concentration data (Table 3.9). There are still differences between the in situ-derived and estimated dose rates but, given that the in situ-derived dose rates varied by more than 0.050 µGy h⁻¹ over short distances (a few m) at these locations, this may be due to the limited number of cores collected (n = 3) at each location.

These observations raise questions over the appropriateness of using K for 226 Ra in calculating dose rates for the entire estuary, although doing so makes the results of the 2007 survey directly comparable with the results of the 1989 survey. It would appear from the Location 3 Core C data and McDonald et al. (2005) that terrestrial dose rates below 0.070 μ Gy h⁻¹ are likely to be dominated by contributions from natural radionuclides. However, the data reported suggest that at higher terrestrial dose rates (> 0.100 μ Gy h⁻¹) anthropogenic radionuclides may be the dominant contributor and K for 137 Cs should be used. Applying K for 137 Cs rather than K for 226 Ra to the measured count rates from the 2007 survey increases the dose rates by approximately 30% on average. This highlights the need for transparency in the approach adopted for

| reporting measured gamma radiation dose rates, and some of the pitfalls which may be encountered when comparing results between different surveys. | Эе |
|----------------------------------------------------------------------------------------------------------------------------------------------------|----|
| | |

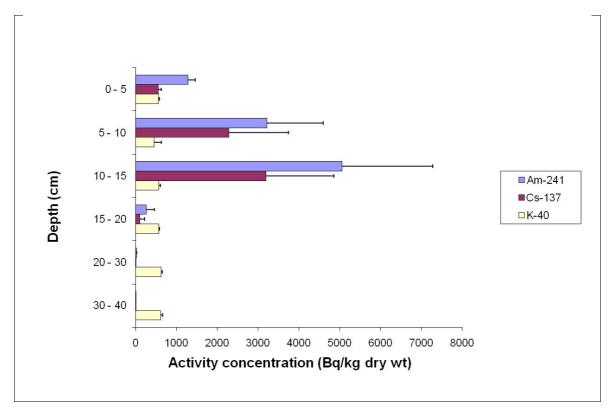


Figure 3.8 Mean depth profiles for ²⁴¹Am, ¹³⁷Cs and ⁴⁰K at Location 1. Error bars show standard deviation.

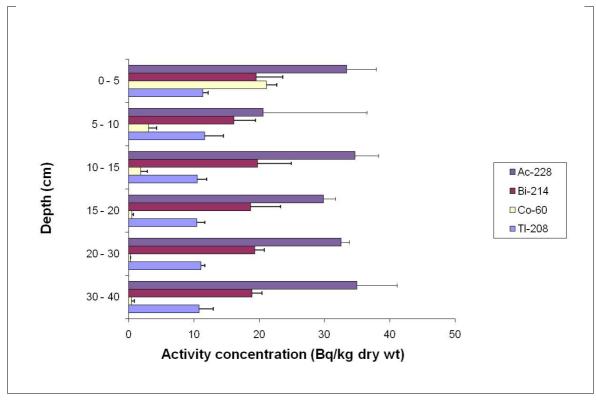


Figure 3.9 Mean depth profiles for ²²⁸Ac, ²¹⁴Bi, ⁶⁰Co and ²⁰⁸Tl at Location 1. Error bars show standard deviation.

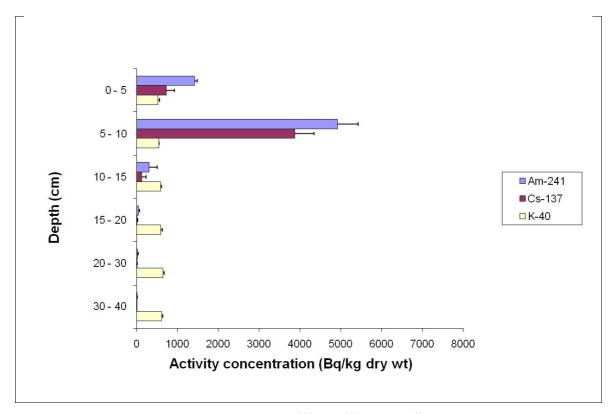


Figure 3.10 Mean depth profiles for ²⁴¹Am, ¹³⁷Cs and ⁴⁰K at Location 2. Error bars show standard deviation.

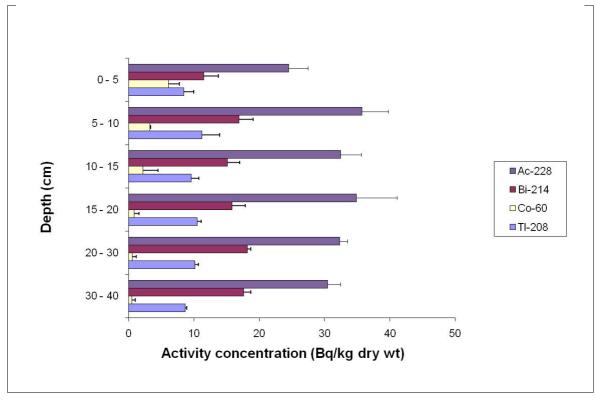


Figure 3.11 Mean depth profiles for ²²⁸Ac, ²¹⁴Bi, ⁶⁰Co and ²⁰⁸Tl at Location 2. Error bars show standard deviation.

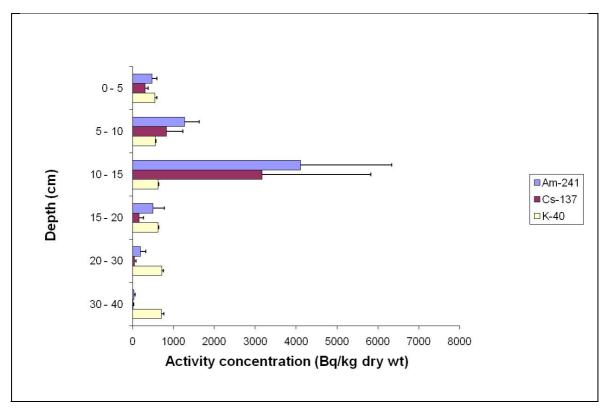


Figure 3.12 Mean depth profiles for ²⁴¹Am, ¹³⁷Cs and ⁴⁰K at Location 3. Error bars show standard deviation.

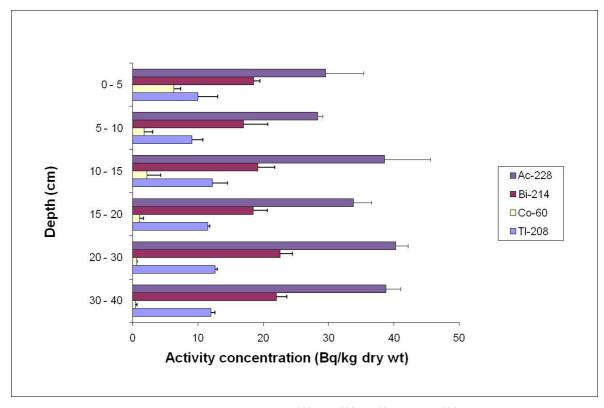


Figure 3.13 Mean depth profiles for ²²⁸Ac, ²¹⁴Bi, ⁶⁰Co and ²⁰⁸Tl at Location 3. Error bars show standard deviation.

Table 3.7 Calculated contributions of natural and anthropogenic radionuclides to terrestrial dose rate ($D_{N_{l,A}}$) (μ Gy h⁻¹)

| Location | Core | ²³⁸ U Series | ²³² Th Series | ⁴⁰ K | ¹³⁷ Cs | ⁶⁰ Co | Total Terrestrial |
|----------|------|----------------------------|-----------------------------|-----------------|-------------------|------------------|----------------------|
| 1 | Α | 0.006 | 0.010 | 0.024 | 0.063 | 0.002 | 0.105 |
| | В | 0.006 | 0.010 | 0.025 | 0.092 | 0.002 | 0.135 |
| | С | 0.007 | 0.011 | 0.024 | 0.094 | 0.002 | 0.137 |
| 2 | Α | 0.007 | 0.011 | 0.022 | 0.069 | 0.001 | 0.110 |
| | В | 0.007 | 0.011 | 0.024 | 0.062 | 0.001 | 0.104 |
| | С | 0.008 | 0.010 | 0.025 | 0.073 | 0.001 | 0.117 |
| 3 | Α | 0.008 | 0.010 | 0.025 | 0.089 | 0.001 | 0.133 |
| | В | 0.008 | 0.012 | 0.027 | 0.070 | 0.001 | 0.119 |
| | С | 0.007 | 0.011 | 0.026 | 0.020 | 0.001 | 0.066 |

Notes: ¹ Dose rates calculated from measured activity concentrations using the methodology described in ICRU (1994).

Table 3.8 Relative contributions¹ of natural and anthropogenic radionuclides to terrestrial dose rate ($D_{Nt,A}$) (%)

| Location | Core | ²³⁸ U Series | ²³² Th Series | ⁴⁰ K | ¹³⁷ Cs | ⁶⁰ Co |
|----------|------|-------------------------|--------------------------|-----------------|-------------------|------------------|
| 1 | Α | 6 | 9 | 23 | 60 | 2 |
| | В | 4 | 7 | 18 | 68 | 2 |
| | С | 5 | 8 | 17 | 69 | 1 |
| 2 | Α | 6 | 10 | 20 | 63 | 1 |
| | В | 7 | 10 | 23 | 59 | 1 |
| | С | 7 | 8 | 21 | 63 | 1 |
| 3 | Α | 6 | 8 | 19 | 67 | 1 |
| | В | 7 | 10 | 23 | 59 | 1 |
| | С | 11 | 17 | 39 | 31 | 1 |

Notes: ¹ Dose rates calculated from measured activity concentrations using the methodology described in ICRU (1994).

Table 3.9 Measured terrestrial dose rates ($D_{N_{I},A}$) and calculated dose rate ranges at Locations 1 – 3 (μ Gy h⁻¹)

| Site | Measured (based on <i>K</i> for ²²⁶ Ra) | Measured (based on <i>K</i> for ¹³⁷ Cs) | Calculated from measured activity concentrations ¹ |
|------------|----------------------------------------------------|----------------------------------------------------|---------------------------------------------------------------|
| Location 1 | 0.055 - 0.103 | 0.070 - 0.132 | 0.105 – 0.137 |
| Location 2 | 0.068 - 0.078 | 0.087 - 0.100 | 0.104 – 0.117 |
| Location 3 | 0.075 – 0.113 | 0.097 - 0.145 | 0.066 - 0.133 |

Notes: ¹Dose rates calculated from measured activity concentrations using the methodology described in ICRU (1994).

3.4 Particle size analysis

The particle size distribution in surface sediments collected from the Esk Estuary in 2007 indicates that medium (212 $\mu m-600~\mu m$) and coarse (600 $\mu m-2~\mu m$) grained sands are the dominant size fractions, with mean contributions to the sediment composition of 38 % and 33 % respectively (Table 3.7). Silt (< 63 μm) generally contributes < 10 % to the sediment mass but this can exceed 25% in some areas, especially in the Mite estuary (Zone B). The previous 1989 survey identified a strong positive correlation between areas of fine-grained sediment deposition and measured external gamma dose rates (Emptage & Kelly, 1990) but that relationship was not evident from the 2007 survey PSA data (Figure 3.14). The r^2 for the regression analysis (0.01) indicated no statistically significant relationship between the proportion of silt in the surface sediments and the dose rate measured.

Table 3.10 Percentage (by mass) of four Wentworth particle classification categories in surface sediments collected from the Esk Estuary in 2007

| Statistic | Coarse sand (600 µm – 2 µm) | Medium sand (212 μm – 600 μm) | Fine sand (63 μm – 212 μm) | Silt (< 63 μm) |
|-----------|--------------------------------|----------------------------------|-------------------------------|-------------------|
| n | 60 | 60 | 60 | 60 |
| Mean | 32.64 | 37.82 | 20.49 | 9.05 |
| Median | 38.40 | 33.79 | 16.43 | 8.65 |
| SD | 15.38 | 18.17 | 14.26 | 6.39 |
| Min | 0.02 | 0.00 | 5.74 | 0.03 |
| Max | 59.22 | 90.67 | 69.60 | 27.13 |

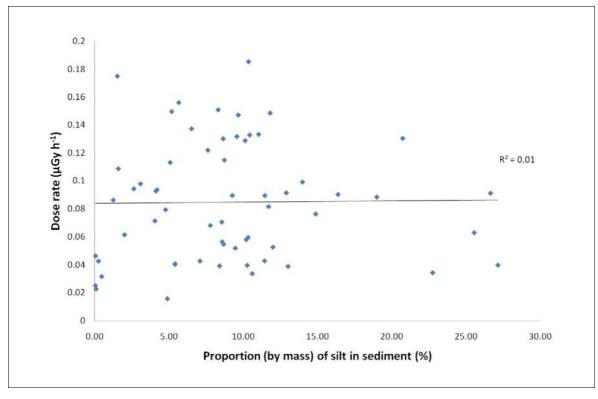


Figure 3.14 Relationship between the composition of surface sediments and measured external gamma dose rates

3.5 External exposure estimation

The calculated total and terrestrial dose rates (μ Gy h⁻¹) were converted to external exposure (μ Sv h⁻¹) using a Gy to Sv conversion coefficient of 0.85 for a rotational geometry and photon energies in the range 50 keV – 2 MeV (ICRP, 1996). This is in agreement with the approach adopted for regulatory monitoring in the United Kingdom (e.g. RIFE, 2008) but other studies adopt different conversion coefficients and this must be recognised when comparing the external exposure estimates of these studies. For example, Green et al. (1989) use the UNSCEAR (1982) conversion coefficient of 0.70 Sv Gy⁻¹, which may actually be more appropriate for situations in which the measured gamma dose rate is derived from a mixture of anthropogenic and natural radionuclides (RIFE, 2008), and Emptage & Kelly (1991) use a conversion coefficient of 0.87 (Spiers et al., 1981). Summary statistics for external exposure are presented in Table 3.11.

The maximum total external exposure in the estuary had reduced from $0.517~\mu Sv~h^{-1}$ in 1989 to $0.200~\mu Sv~h^{-1}$ in 2007. Thus, if exposure was solely from external radiation in the estuary, a member of the public would have to be present at the maximum exposure location (in the inner Esk Estuary) for 5,000 hours (0.57 years) to reach the 1 mSv annual dose limit. Although other exposure pathways are not considered, it is evident that this level of occupancy at the location of maximum external exposure is highly unlikely; the actual maximum annual occupancy in the estuary (combining recreational and occupational occupancies) having been determined through habits survey as 424 hours (Doddington et al., 1990). Also, the above calculation is based on a 'worst case' total dose rate, which includes cosmic, anthropogenic and natural terrestrial radioactivity contributions, whereas the statutory 1 mSv annual dose limit is applies to exposures from anthropogenic sources only.

Table 3.11 Summary statistics for external exposure (μSv h⁻¹) in the Esk Estuary in 2007

| Year of survey | Statistic | Dose rate (µSv h ⁻¹) | | |
|-------------------|-----------|----------------------------------|----------------------|--|
| | | Total exposure | Terrestrial exposure | |
| 1989 ¹ | n | 890 | 890 | |
| | Mean | 0.197 | 0.156 | |
| | Median | 0.151 | 0.110 | |
| | SD | 0.115 | 0.115 | |
| | Min | 0.060 | 0.018 | |
| | Max | 0.517 | 0.475 | |
| 2007 | n | 576 | 576 | |
| | Mean | 0.105 | 0.062 | |
| | Median | 0.098 | 0.056 | |
| | SD | 0.029 | 0.029 | |
| | Min | 0.054 | 0.014 | |
| | Max | 0.200 | 0.158 | |

Notes: ¹For the purposes of comparison, exposure values for the 1989 survey were recalculated using the 0.85 conversion factor proposed by the ICRP (ICRP, 1996)

4 Discussion

The reduction in terrestrial dose rates ($D_{\it Nt,A}$), and therefore external exposure, throughout the estuary, is likely to be due to the combined influence of decreasing radionuclide discharges from the Sellafield site (Figure 1.1), radioactive decay of Chernobyl-derived ¹³⁷Cs ($t_{1/2}$ = 30 years), and burial of contaminated sediment as indicated by the sub-surface maxima for ¹³⁷Cs and ²⁴¹Am (Figures 3.8, 3.10 & 3.12). This burial of contaminated sediments would increase the attenuation of gamma photons by the overlying sediment, (especially lower energy gamma photons), and reduce the measured external gamma dose rates.

For ⁶⁰Co, an exponential decrease with depth was observed, rather than the presence of sub-surface maxima (Figures 3.9, 3.11 & 3.13). This is likely to be due to differences in the discharge history of ⁶⁰Co and, for example, ¹³⁷Cs. Marine discharges of ¹³⁷Cs from Sellafield peaked in the mid-1970s and had decreased markedly by the mid-1980s. Discharges of ⁶⁰Co had a peak in the mid-1980s followed by a second peak of similar magnitude in the late 1990s. Therefore, upper layers of stable sediment areas in the estuary are more likely to have been subject to recent inputs of ⁶⁰Co than ¹³⁷Cs. This would result in the different depth profiles observed for these anthropogenic radionuclides.

The depth profile of ²⁴¹Am in the sediments, which is similar to that of ¹³⁷Cs, is likely to be representative of the depth profile for the Pu isotopes that have been discharged from Sellafield. This suggests that, in the stable sediment areas of the estuary, there may be ongoing burial of contaminated deposits, reducing exposure to members of the public. This theory is supported by the lack of correlation between surface sediment particle size and measured dose rates (Figure 3.14). Given the dynamic nature of estuaries, periodic movements of surface sediments and redistribution of grain sizes in the surface sediments may be expected.

The terrestrial gamma dose rates ($D_{N_{I},A}$) calculated from the sediment activity concentrations (Table 3.7) demonstrate that anthropogenic radionuclides (specifically ¹³⁷Cs) were the dominant contributor to the dose rate at sites with elevated dose rates (> 0.100 μ Gy h⁻¹), accounting for 60 – 70 % of the dose rate (Table 3.8)..

Although there is considerable heterogeneity in the sediment radionuclide activity concentrations (and therefore external gamma dose rates), over small distances (< 10 m) in the estuary (see Section 3.3), the data from the 2007 survey suggest that in addition to radioactive decay, the reduction in observed dose rates may be closely linked to burial of contaminated sediments. On a large scale there appears to be little evidence of significant redistribution of contaminated sediment within the estuary (Figures 3.1 & 3.3-3.7). In considering potential future long-term radiation exposures in the Esk Estuary, it may be prudent to consider the potential for remobilisation of contaminated sediments (for example as tidal regimes alter under the influence of climate change and sea-level rise).

5 Conclusions

This study has demonstrated a highly statistically significant decrease in the external gamma dose rates within the Esk Estuary between 1989 and 2007. The measured total gamma dose rate ($D_{I,C,Nt,A}$) range in 2007 was 0.064 – 0.235 μ Gy h⁻¹ with a mean of 0.123 μ Gy h⁻¹. This equates to a maximum exposure of 0.200 μ Sv h⁻¹ and a member of the public would need to be present at the maximum exposure location for 57% of the time in order for them to receive the 1 mSv annual dose limit from this external exposure pathway alone. However, this calculation is based on a total dose rate, which includes cosmic, anthropogenic and natural terrestrial radioactivity contributions, whereas the 1 mSv annual dose limit is a regulatory limit that for exposure from anthropogenic sources. Even considering the contribution of other pathways such as radionuclide intake through food, it is unlikely that the annual dose limit for individuals living near the Esk Estuary will be exceeded.

There is limited evidence to suggest that part of the reason for the reduction in measured dose rates is the burial of contaminated sediment deposits by uncontaminated sediments transported by tidal processes.

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