Sea to land transfer of radionuclides in Cumbria and North Wales: updating of previous investigations

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INTRODUCTION

It is now approximately fifteen years since the last comprehensive sea-to-land transfer studies in coastal regions of the UK, during which time there has been a halving of annual discharge rates for ^{137}Cs and a ten-fold decrease in the annual discharge rate for Pu-α from the Sellafield Ltd site in Cumbria, UK. It is therefore of interest to re-consider the sea-to-land transfer process in order to determine whether the rate of transfer of ^{137}Cs and Pu-α on land has been consistent with previous studies or whether changes have occurred in the interim period. Also during this period, discharges of ^{99}Tc have risen and fallen, peaking in 1995. In order to collect up to date information on the sea to land transfer of ^{137}Cs, Pu-α and ^{99}Tc, Westlakes Scientific Consulting performed two recent investigations where soil samples were taken from a transect running inland from Nethertown beach, 10 km north of Sellafield in Cumbria, UK and from a suite of 10 transects along the north coast of Wales. A summarised outcome of these investigations is given below.

METHOD

Site location and Sampling

Soil samples were taken from permanent grassland sites in a transect running inland, perpendicular to the coast, from Nethertown, Cumbria. An initial sample was taken from Nethertown beach, and subsequent soil samples were taken at intervals of approximately 100 m, 200 m, 500 m, 1, 2, 3, 4, 5, 10, 15 and 20 km inland from the coast. In North Wales, ten transects were sampled, covering the coastline between the Dee estuary and Anglesey, including locations within the Menai Straits (Figure 1). Five soil samples were collected along each transect at intervals of 50, 100, 200, 500 m and 5 to 15 km from the mean high water mark. Surface scrapes were taken for beach samples. All soil core samples were collected using a soil corer to depths of 0-15 and 15-30 cm. Samples were weighed, freeze-dried, re-weighed, disaggregated and passed through a 2 mm sieve prior to analysis. Analytical techniques for the quantification of ^{137}Cs, Pu-α and ^{99}Tc used were those previously described by Bryan et al. (2008).

RESULTS AND CONCLUSIONS

Nethertown transect

Soil samples were analysed for ^{137}Cs, Pu-α and ^{99}Tc (Figure 2). The results showed that ^{137}Cs was not a good tracer for use in determining the sea-to-land transfer process, due to the dominance of Chernobyl-derived deposition. The activity concentrations and inventories of Pu-α were slightly higher than in previous studies, due to the longer time period of deposition. Technetium-99 was detected only in soils within 150 m from the sea, with activity concentrations (0.23 - 0.99 Bq kg^{-1} dry weight) less than one tenth of those found in beach

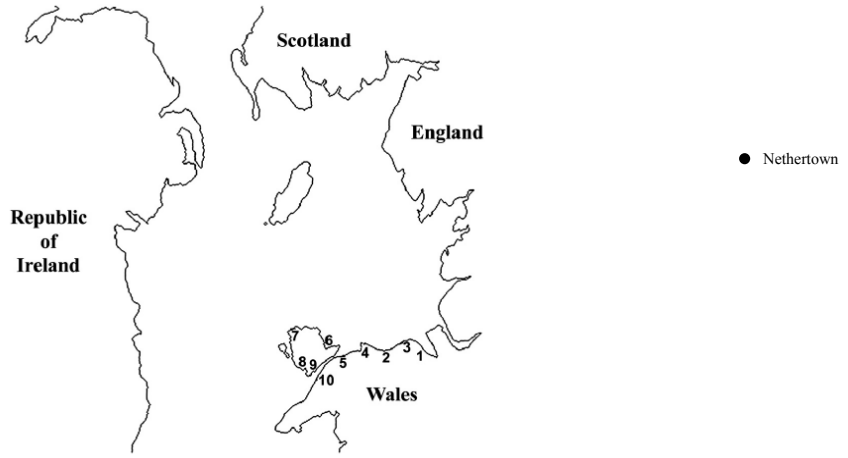
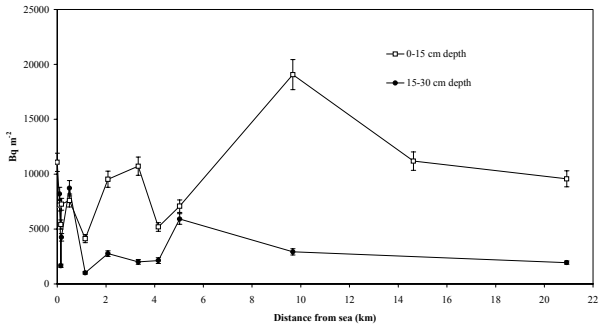
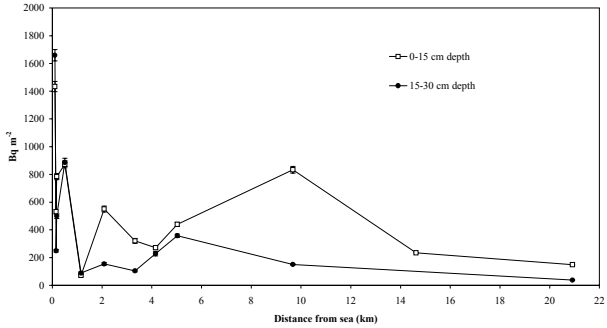


Figure 1. Location of the inland transects at Nethertown and North Wales

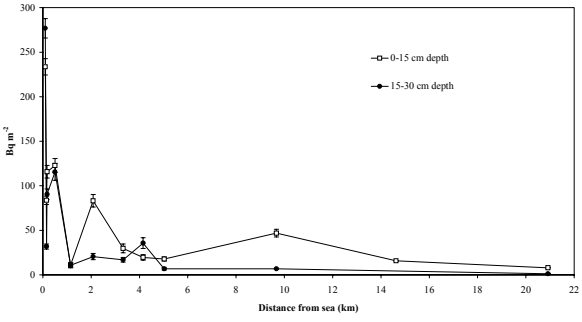
sediment (9.80 Bq kg^{-1} dry weight). At all other sites the activity concentration was less than the analytical limit of detection (0.2 Bq kg^{-1} dry weight). Although the sea-to-land transfer of ^{99}Tc is the only pathway for the deposition of ^{99}Tc in terrestrial environments, other than the application of seaweed as a soil conditioner, the activity concentrations measured were relatively low, and for sites > 150 m from the coast, ^{99}Tc was not detected. Deposition of ^{137}Cs along the Cumbrian



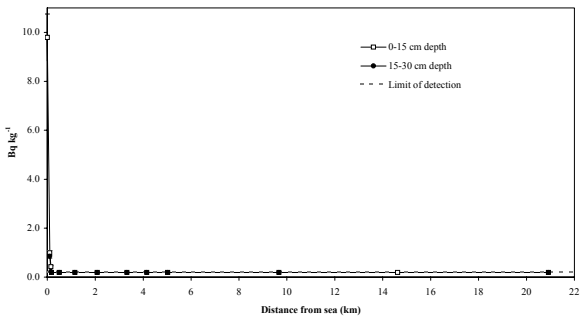
a) Cs-137 deposition (Bq m^{-2})



b) Pu-239,240 deposition (Bq m^{-2})



c) Pu-238 deposition (Bq m^{-2})



d) Tc-99 activity concentration (Bq kg^{-1})

Figure 2. Inland transect deposition trends at Nethertown, UK

coast is primarily derived from Chernobyl fallout whilst the deposition of ^{238}Pu and ^{239,240}Pu was found to have a strong contribution from Sellafield atmospheric discharges at distances beyond 1 km from the coast. The measured deposition of ^{99}Tc was unrelated to atmospheric discharges or global fallout and therefore interpreted to be entirely derived from sea-to-land transfer.

Calculations were made of the quantities of radioactivity transferred from the sea to the Cumbrian coast since 1951. Overall emission rates were calculated to be: 9.09 GBq of ^{99}Tc; 228 GBq of ^{137}Cs; 21.7 GBq of ^{238}Pu; and 106 GBq of ^{239,240}Pu.

Model predicted deposition rates showed that the current deposition of ^{137}Cs and Pu-α derived from the sea, are lower than those at the time of previous studies performed in the 1980's (Eakins *et al*., 1981). This is due to the decreases in liquid discharge activities from the Sellafield site over the last two decades, which have resulted in lower seawater concentrations of these radionuclides. In the case of ^{99}Tc, the recent decrease in discharges has meant that the current sea-to-land deposition rate is comparable with rates estimated for the early 1990's, prior to the peak discharges of the last fifteen years.

North Wales transects

^{137}Cs

For the majority of the transects (2, 3, 5, 6, 7, 8, 9 and 10), there was either little variation or no correlation between distance from the coast and ^{137}Cs activity concentration. Transects 1 and 4 recorded a decline in ^{137}Cs deposition within the first 200 m from the mean high water mark. Subsequent increases in the inventories at 500 m and in particular at the 5 to 10 km background locations are likely to reflect increased atmospheric fallout, due to higher rainfall experienced by upland locations, either from weapons testing or the Chernobyl accident (Cawse et al., 1988). The deposition inventories measured for all of the locations were consistent with the ranges reported by Cawse et al. (1988) (4000 – 30 000 Bq m^{-2}) and Howorth et al. (1991) (862 – 13 657 Bq m^{-2}).

Pu-alpha

The deposition of ^{238}Pu and ^{239,240}Pu in soils ranged between 3 to 36 Bq m^{-2} and 21 to 227 Bq m^{-2} respectively. These are comparable to the ranges reported by Howorth et al. (1991) for soils within 100 m of the North Wales coastline (7.9 to 67.4 Bq m^{-2} and 29.8 to 404 Bq m^{-2} for ^{238}Pu and ^{239,240}Pu respectively). The results show that there is some evidence for enhanced deposition close to the sea. However, the effect is not as obvious as for data described within the Nethertown transect.

^{99}Tc

Of the ten intertidal sediment samples, seven recorded ^{99}Tc activity concentrations greater than the limit of detection. With one exception, the activity concentrations were relatively low compared to those reported for sediments closer to Sellafield (BNGSL, 2006). The exception to this was sediment sampled from the mean high water mark at transect 9. The value detected at this location (1320 Bq kg^{-1}) was 30 times higher than the next highest activity concentration, which was measured in sediment collected from the mean high water mark of transect 1. Of the remaining 9 transects, ^{99}Tc was detected in only a single soil sample, that collected from 68 m inland at transect 1. The activity concentration (2.00 ± 1.26 Bq kg^{-1}) was close to the limit of detection. Hence, there is no evidence of significant ^{99}Tc transfer on to coastal land in North Wales via the mechanism of deposition from sea spray.

From the more distant Welsh transects, the ^{137}Cs, ^{238}Pu and ^{239,340}Pu field data were consistent with what had been reported 15 years previously. Therefore there has been no increase in the supply of these Sellafield-derived radionuclides to the terrestrial environment of the North Wales coast. The ^{99}Tc data in Welsh sediments were consistent with reported values within annual monitoring programmes, however, a relatively high activity concentration was measured in one sediment sample. This site was further investigated to determine the reason

why such a high value was found. At present there is no clear evidence as to why this elevated concentration should be present, but the role of seaweed and its capacity in accumulating ^{99}Tc and transferring it to sediment is of interest. The enhanced activity concentration of ^{99}Tc measured in the sediment is insignificant with respect to CED (0.0001% of the annual dose limit to members of the public).

A highly conservative assessment of the modelled CED values for members of the public in North Wales was determined by assuming that individuals obtain all of their foodstuffs from fields within 100 m of the sea and also reside at the same location. Foodstuff concentrations attributable to sea to land transfer were modelled using the dynamic compartmental model LANDFOOD, based on the NRPB FARMLAND code (Brown and Simmonds, 1995). The assessment considered three different age groups: Adults, Children (1-15 years of age) and infants (0-1 year old).

The year with the highest CED values from sea to land transfer for all age groups, under critical group conditions, was modelled to be 1985, with CED values of 1.46 μSv, 1.01 μSv and 0.60 μSv to adults, children and infants respectively. CEDs for 2004 were found to be considerably lower at 0.59 μSv, 0.40 μSv and 0.26 μSv.

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