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Assessment of Elevated Radionuclide Levels in Soils Associated with an Avian Colony in a High Arctic Environment

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This article presents the results of an investigation into the occurrence of elevated levels of radionuclides in soils associated with a seabird colony in the Arctic. Soils and other materials were collected from a seabird colony (primarily composed of kittiwakes) in Kongsfjorden, located in the High Arctic archipelago of Svalbard. The samples were analyzed for a suite of gamma emitting natural and anthropogenic radionuclides, including ^{137}*Cs and nuclides of the* ^{238}*U and* ^{232}*Th series, to establish the level of enrichment and the behavior of the radionuclides in the immediate area. The results indicate that soils near the colony exhibit enrichment factors of 8 for* ^{137}*Cs, 5 for* ^{238}*U and 2 for* ^{226}*Ra compared to the nuclide content of soils from the general area. The spatial patterns of the nuclides in the soil are consistent with enrichment of the soil via run-off draining from a large accumulation of fecal and nesting material that has developed at the base of the colony.* ^{137}*Cs ingress to the soil appears to have peaked at some point in the past as patterns of enrichment at the colony are different to those exhibited by* ^{238}*U, which must be assumed to be a steady state contribution. The means of introduction of radionuclides to the colony remains unclear but the transfer of* ^{137}*Cs from the marine environment to the terrestrial environment via the food chain and deposition of feces is discussed.*

Keywords Radionuclides, Arctic, radioecology, seabirds, soil.

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Introduction

The role of seabirds in the introduction and environmental fate of anthropogenic contaminants in terrestrial ecosystems has received more attention in recent years, as evidenced by the number of published articles in the literature (Norheim and Kjos-Hanssen, 1984; Schneider *et al.*, 1985; Nettleship and Peakall, 1987; Norheim, 1987; Headley, 1996). To date, the primary contaminants of concern and hence the most frequently reported have been persistent organic pollutants and heavy metals (AMAP, 1998). More recent studies (Headley, 1996) have indicated that seabird colonies may also play a role in the concentration of natural and anthropogenic radionuclides in the Arctic terrestrial environment. Seabirds in high density coastal colonies can have significant impacts on the terrestrial environment. The deposition of large amounts of fecal material can have an influence on the chemical characteristics of the soil. As a result, soils may display changes in both nutrient status, pH and water retention capacity (Smith, 1979; Hogg and Morton, 1983; Bukacinski *et al.*, 1994; Vidal *et al.*, 1998; Garcia *et al.*, 2002). Such changes tend to be most visibly reflected in the diversity and amount of vegetative species to be found at such locations, this being most apparent in nutrient-poor environments found in the Arctic region (Godzik, 1991) where seabird faces often constitute a vital source of nutrients. However, the deposition of fecal material may also result in a significant input of contaminants to the surrounding environment. Heavy metal inputs from seabird colonies have been reported from around the globe, including the Arctic environment (Norheim, 1987; Grodzinska and Godzik, 1991; Headley, 1996). The importance of such processes with respect to radionuclide behavior in the Arctic is the possibility of both local enrichment of radionuclides to levels significantly higher than those of the general background and the transfer of radionuclides within and between environmental compartments. Furthermore, such processes have implications for the behavior of radionuclides in the Arctic environment and the final "sink" for radionuclides in the terrestrial ecosystem. The need for information as to how radionuclides behave in the terrestrial environments of the Arctic in general and Svalbard in particular is exacerbated by the dearth of information relating to the behavior of radionuclides in the terrestrial environment of these regions.

Hallstadius *et al.* (1982), working before the Chernobyl accident, provided a figure for aerial deposition of ^{137}Cs on Svalbard of 2.2 ± 0.3 kBq/m^2 as a result of atmospheric weapons testing in the late 1950's and the 1960's. More recent work on the radionuclide loads in glaciers on Svalbard (Pinglot *et al.*, 1994) provided figures in the range of 0.2–0.54 kBq/m^2 for weapons test fallout (at time of deposition) and assess the Chernobyl contribution as 0.02 kBq/m^2 on West Svalbard. These figures indicate that the primary source of anthropogenic radioactivity on Svalbard is atmospheric fallout, associated with nuclear weapons testing 45 years ago. An assessment of the radioecology of the terrestrial environment of Kongsfjorden, Svalbard in 2001 (Dowdall *et al.*, 2003) indicated that radionuclide levels in soils taken from the West Coast of Svalbard were generally low (mean values: ^{238}U 42 Bq/kg, ^{226}Ra 43 Bq/kg, ^{232}Th 21 Bq/kg, ^{40}K 283 Bq/kg and ^{137}Cs 35 Bq/kg) and displayed a certain degree of spatial homogeneity. However, levels of radionuclides in samples taken from areas associated with seabird colonies in Kongsfjorden displayed anomalous levels of both natural radionuclides and the anthropogenic gamma-emitter ^{137}Cs, relative to average levels for the region. Based on these findings, a more intensive study at one of the sites surveyed in 2001 was undertaken to assess radionuclides found at the bird colony. It was hoped that such a survey would provide information relating to both the nature of the enhancement and the behavior of the radionuclides at the site and provide insight into the radioecological behavior of radionuclides within what is an important ecological system in the Arctic environment.

Materials and Methods

Site Location and Description

The site is adjacent to a seabird colony at Krykkefjellet, on the southeastern shore of Kongsfjorden, Svalbard (78° 53.775′ N, 12° 11.788′ E). The geology of the area consists of sedimentary rocks of Middle and Upper Carboniferous and Permian ages with sandstones and limestones at various locations (Hjelle, 1993). Soils of the general area vary in depth and are generally low in humic materials and are of ages between 9000 and 12000 years (Mann *et al.*, 1986), although the lowland surrounding the specific site consists mainly of gravel deposits with virtually no organic material.

The colony itself consists of approximately 600 pairs of Kittiwakes (*Rissa tridactyla*), 100 pairs of Brunnichs guillemot (*Uria lomvia*) and less than 10 pairs each of Black guillemot (*Cepphus grylle*) and Glaucous gull (*Larus hyperboreus*) (Mehlum and Bakken, 1994). The colony exists on a rock face of some 10 m in height and approx. 30–40 m in length, the base of which is a rock talus covered by accumulated fecal and nesting material, stretching out some 6–7 m from the bottom of the cliff (Figure 1). Water input to the area is largely due to snow and ice melt and where run-off drains from the talus material, a thin (<5 cm) layer of organic soil has developed. No evidence of horizon development was exhibited. In 2002, a series of transects were established for sampling, two parallel to the base of the talus that extended left and right of the slope and three perpendicular to the talus slope, covering an area of approximately 100 m by 30 m. The purpose of this transect system was to determine the extent of influence of the colony on the nearby soil regarding radionuclide levels. At regular intervals along each transect, approximately 4 to 5 kg wet weight of soil was taken using a stainless steel trowel. All samples were sealed in polyethylene bags and returned to the Norwegian Radiation Protection Authority laboratory at Tromsø, Norway. Additional samples were taken of the fecal and nesting material and treated in the same manner as per the other samples.

In the laboratory, transect samples were divided into separate sub-samples consisting of the mineral basal layer and, when present, the overlying humified organic material. All samples were dried at 105°C to constant weight, homogenized in a stainless steel blender, passed through a 2 mm sieve and packed into analytical geometries used for gamma spectrometry. Analytical sample sizes ranged from 20 g to 800 g and samples were counted for a minimum of 24 hours on a high resolution p-type Ge gamma spectrometer (resolution of 1.9 keV at 1332 keV, nominal relative efficiency of 40%) coupled with an 8k multichannel analyzer. The spectrometer was calibrated for energy and photopeak efficiency using traceable standard radionuclide solutions from the Physikalisch Technische Bundesanstalt, Germany. Matrix interferences due to varying sample density and chemical composition were corrected by Gamatool 2.1 from AEA Technology using the mass attenuation coefficients from Hubbel (1982). ^{238}U was assessed using the emissions of the immediate daughter ^{234}Th which, due to its immobility and short half-life, can be assumed to be in secular equilibrium with its parent. The activity ratio between ^{238}U and ^{235}U in environmental materials was then used to correct the peak at 186 keV for the ^{235}U component and obtain a value for ^{226}Ra. Activities of other nuclides were calculated based on their primary emissions. Errors (1σ) included all major contributions and were of the order of less than 20 % for ^{137}Cs and ^{40}K and less than 30% for other nuclides. Samples were analyzed within the internal quality assurance system of the laboratory, using splits, blanks and reference materials and participation in international and national intercomparison exercises.

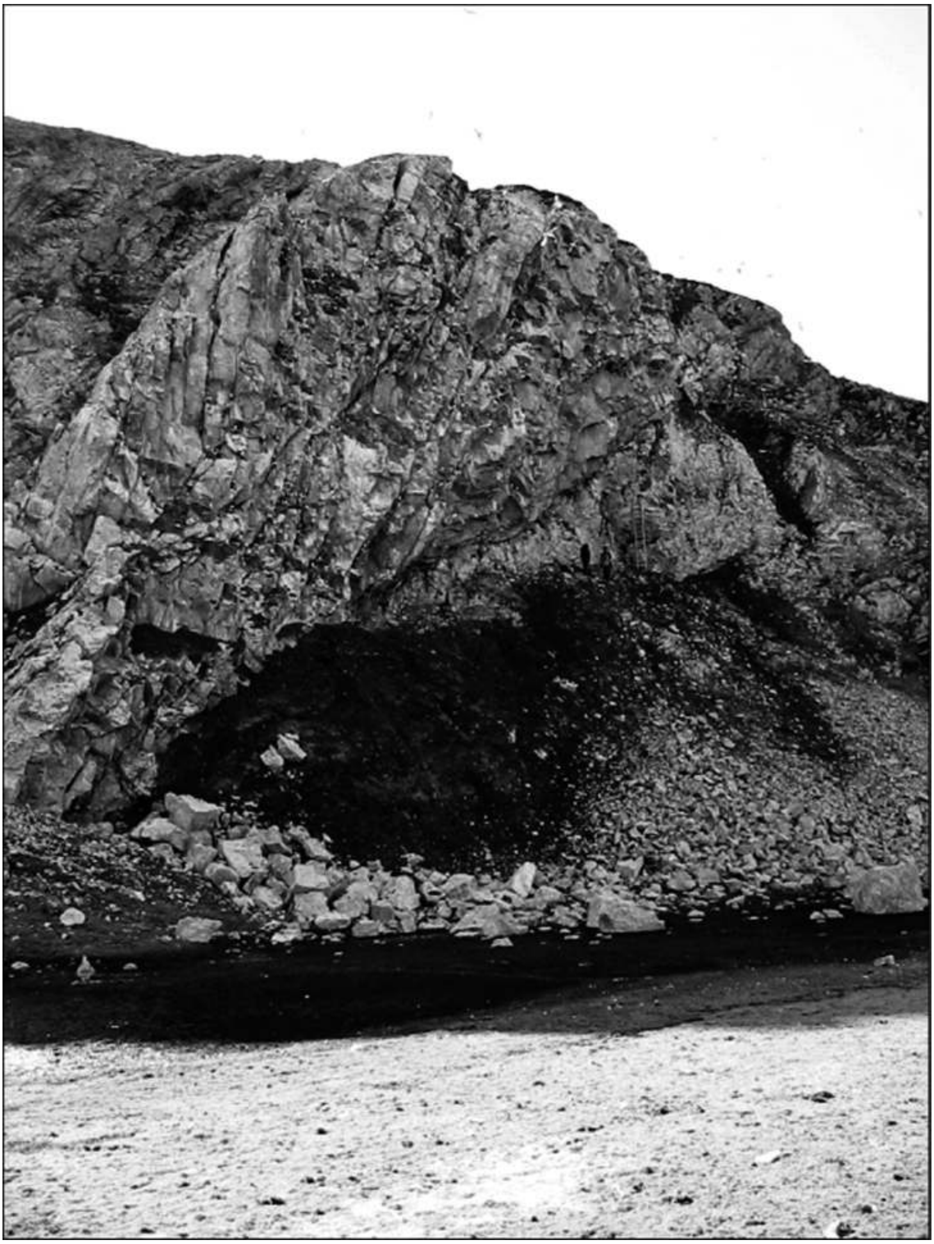

Figure 1. Photograph of study site. The brown sloped area on the talus consists of deposited fecal material and decayed vegetation. The dark area at the base of the talus is organic material overlying a thin stratum of mineral material.

Results

In the talus material, the distribution of natural radionuclides is similar both within and down the talus slope, whereas levels of ^{137}Cs are higher at the base of the slope and within the talus itself (Table 1), as has been reported for a seabird colony elsewhere on Spitsbergen

Table 1

Activity concentrations of ^{137}Cs, ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K in talus material, overlying humified organic layer and underlying mineral soil at the base of the talus. Average values for typical Kongsfjorden soils are shown for comparison (Dowdall *et al.*, 2003). All values dry weight

	^{137}Cs Bq/kg	^{238}U Bq/kg	^{226}Ra Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg
Average values for typical Kongsfjorden soil (n = 11)					
0–3 cm	24.3 ± 35.1	42 ± 33.9	50.2 ± 32	25.9 ± 12.8	342 ± 165
Transect (1.5 m intervals) down slope of talus (n = 1)					
Top of slope	92	77	67	17.3	410
	94.6	76	54	17.3	356
	123.1	58	78	20.7	426
	105	72	64	18.9	364
Base of slope	128	69	88	22	458
Depth profile of talus taken at top of slope (n = 1)					
0–15 cm	84.7	65	64	19.2	394
15–30 cm	69	56	61	17.6	406
30–45 cm	80	73	55	17.8	451
45–60 cm	148	69	94	21.2	534
Overlying humified organic layer at base of talus (n = 19)					
Mean	164.6 ± 67.1	101.3 ± 48.7	47.5 ± 21.4	31.6 ± 6.3	533 ± 95
Range	65–283	<12–244	<4–94	22.7–47.5	340–784
Underlying mineral soil at base of talus (n = 28)					
Mean	34.2 ± 22.9	31 ± 9.2	28.1 ± 6.3	27 ± 1.6	622 ± 32
Range	0.7–91.6	20–56	19–45	24.5–31.4	562–698

(Dowdall *et al.*, 2003). ^{137}Cs is enriched in the talus material up to 6-fold compared to average soil nuclide values for the area (Dowdall *et al.*, 2003), while ^{238}U and ^{226}Ra are enriched up to 2-fold.

Mean activity concentrations of ^{137}Cs and ^{238}U in the humified organic layer at the base of the talus were higher than that observed in the talus materials and relative to the average nuclide content of soils from the area (Dowdall *et al.*, 2003) showed enrichment by up to a factor of 18 for ^{137}Cs and 9 for ^{238}U (Table 1). ^{238}U in the humified layer is not in secular equilibrium with ^{226}Ra and is enhanced in the humified layer relative to ^{232}Th, with the majority of samples exhibiting ^{226}Ra/^{238}U ratios of less than unity (mean of 0.52) and ^{238}U/^{232}Th ratios greater than 8. These values can be compared to the ratios exhibited by soils of the general area of $\sim$1 (^{226}Ra/^{238}U) and $<$3 (^{238}U/^{232}Th) as reported by Dowdall *et al.* (2003).

Average levels of radionuclides in the underlying mineral layers of the soils associated with the bird colony are quite comparable to average soil levels for the area (Dowdall *et al.*, 2003), although average levels of ^{40}K and ^{137}Cs in the mineral layers near the colony appear to be higher (Table 1). The levels in the mineral layers are quite constant between samples, the difference between the first and third quartiles not being greater than 30% of the range of values for any of the nuclides. In comparison, although all levels of the nuclides in the humified layer are normally distributed, they are altogether more variable than compared to the mineral layers, with distribution ranges greater by factors of between 3 and 4 for all nuclides.^{226}Ra and ^{238}U are in approximate secular equilibrium for the majority of the mineral layer samples, with a median ^{226}Ra/^{238}U value of 0.93 while the average ^{238}U/^{232}Th ratio is 1.12, which is significantly different than for ^{238}U/^{232}Th ratios in the humified layers (Mann-Whitney test).

Discussion

In order to identify any significant spatial trends in the data, the mineral and humified data sets were gridded using a multi-quadratic function (Franke, 1982) and contoured (Figures 2 and 3). The spatial pattern of ^{232}Th and ^{40}K suggests that the content of these nuclides in the overlying layers is a function of the nuclide content of the underlying material. The relative

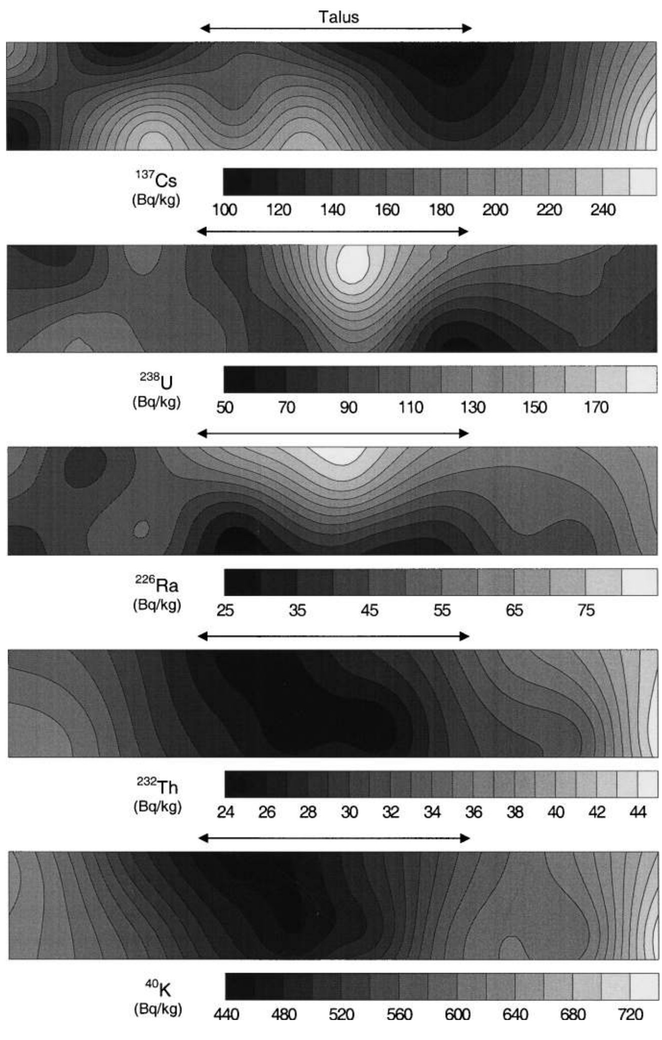

Figure 2. Spatial distribution of ^{137}Cs, ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in humified layer at the base of the fecal and nesting material talus. Position of talus relative to contoured area shown with arrow. Area shown is 25 m × 90 m.

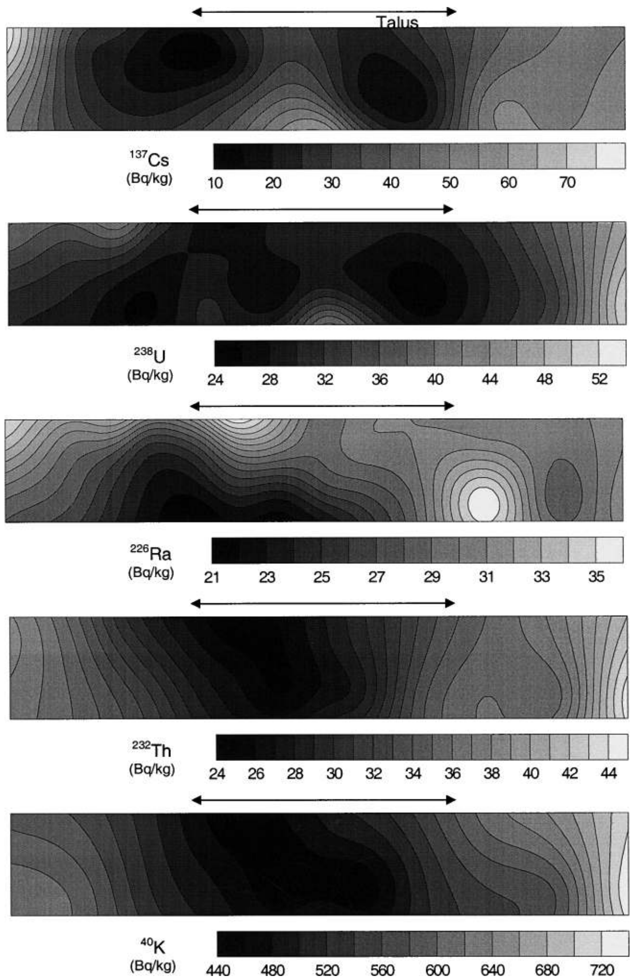

Figure 3. Spatial distribution of ^{137}Cs, ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in mineral layer at the base of the fecal and nesting material talus. Position of talus relative to contoured area shown with arrow. Area shown is 25 m × 90 m.

immobility of ^{232}Th in the biosphere (Sheppard, 1980) precludes the likely introduction of this nuclide by any means other than normal weathering of the underlying substrate. Although ^{40}K can be considered to be more mobile than ^{232}Th in the surficial environment, the extremely slow rate of soil development in the Arctic regions and the very low biological activity may have resulted in the overlying humified layers reflecting the underlying mineral stratum. In contrast to ^{232}Th and ^{40}K, both ^{226}Ra and ^{238}U levels in the humified layer display a strong positive plume out from the base of the talus material towards the center of the study area, which is not replicated in the corresponding mineral layer (Figures 2 and 3). Due to the relative immobility of ^{226}Ra under normal surficial conditions (Taskayev *et al.*, 1978), it is unlikely that the enhanced levels of ^{226}Ra in the humified layers are due to direct inputs of this nuclide but are probably due to ingrowths from such parent ^{238}U as may have accumulated at the base of the talus. The plume of ^{238}U in the humified layer strongly corresponds with the main area of runoff from the colony, indicating the possible leaching of this nuclide from the talus material and into the surrounding soils. A slight but significant positive correlation (Pearson, c = 0.6, $p = 0.012$) is observed between the level of enrichments of the ^{238}U and ^{226}Ra supporting the hypothesis that ^{238}U enrichment is occurring. Furthermore, the even spatial distribution of ^{238}U suggests that the input of this nuclide is a steady state process. In contrast to ^{238}U and ^{226}Ra, ^{137}Cs levels at the site are lowest in both mineral and humified layers nearest the colony, with maximum levels occurring at the periphery of the area that is influenced directly by runoff from the colony. Penetration of ^{137}Cs from the humified layer into the mineral layer is minimal with an average humified/mineral activity ratio of 8.5. ^{137}Cs interacts strongly with soil clay minerals and organic matter and can display relatively low mobility in the soil column (Cornell, 1993; Dumat *et al.*, 1997) so the difference in levels of this nuclide between the organic and non-organic layer is to be expected. Given the spatial distribution of ^{137}Cs within the talus material it would appear that the degree of input and or leaching of this nuclide from the talus material into the surrounding soil has changed over time, an idea further supported by the observed gradient in ^{137}Cs levels away from the base of the talus material.

Although it appears that the primary expression of the seabird colony, on the levels of radionuclides in the surrounding soil, is via levels of ^{137}Cs, ^{238}U and indirectly ^{226}Ra, the source of the radionuclides is unclear. A number of possible sources are, however, worthy of consideration. First and foremost of these is the underlying geology, which through weathering processes may introduce natural radionuclides into overlying soils. Although a sampling program for uranium bearing minerals was not carried out, it is reasonable to assume that the inorganic material present at the site represents the weathered parent bedrock and none of the samples of this material display any evidence of containing uranium or thorium levels that can be considered elevated relative to the surrounding area. However, consideration of the typical geochemical behavior of uranium and radium isotopes in the environment with respect to organic material may provide some insight. Uranium occurs in surface waters primarily as complexes with carbonate, sulphate, silicate and phosphate (Langmiur, 1978), Adams *et al*. (1959) providing an average value for surface fresh waters of 1.2×10^{-3} Bq/l. It has long been established that the organic constituents of soils can adsorb uranium with an enrichment factor ($[U_{\text{organic matter}}]/[U_{\text{water}}]$) of up to 10000/l (Szalay, 1964) via a cation exchange process. It is therefore plausible that elevated uranium levels at the site could be due to the adsorption of low levels of uranium from surface waters onto organic materials that are only present at the site due to nutrient inputs from the colony. In the case of thorium, the lack of enrichment in the organic soil may reflect the fact that thorium is typically present in surface waters at concentrations two to three orders of magnitude lower than uranium, due to its strong affinity for solid phase materials (Adams *et al.*, 1959).

A further source of radionuclides might arise from decaying vegetation within the talus material that was brought to the site by seabirds to be used as nesting material. However, Dowdall *et al*. (2003) report that in general only low levels of the relevant nuclides (average values for typical vascular plants; ^{137}Cs 53 Bq/kg,^{238}U $<$ 25.5 Bq/kg, ^{226}Ra $<$ 32.8 Bq/kg, ^{232}Th $<$ 13 Bq/kg and ^{40}K 138 Bq/kg) are present in vegetation on Svalbard.

As the fecal material of seabirds has been suggested as contributing to heavy metal loading of the terrestrial environment (Norheim, 1987; Grodzinska and Godzik, 1991; Headley, 1996), it is possible that avian feces may additionally be a source of radionuclides. To investigate this idea we have attempted to calculate a rough estimate as the possible flux of ^{137}Cs, based on typical levels in prey species (Brungot *et al.*, 1999), consumption rates by seabirds (Brekke and Gabrielsen, 1994; Ellis and Gabrielsen, 2002), seabird residence times at the study site (Hop *et al.*, 2002), fecal production rates and percentage deposition at study site (Brekke and Gabrielsen, 1994; Mehlum and Gabrielsen, 1995). Calculations were not made for uranium and thorium series isotopes due to a lack of data pertaining to these nuclides in marine biota.

Calculations indicate a potential current input (via prey) by the colony in the region of 24 to 28 kBq ^{137}Cs per season, this figure undoubtedly being higher during the period of peak atmospheric deposition associated with weapons testing some 40 years ago. Unfortunately, neither information on the assimilation of radionuclides by the relevant seabirds, nor the activity of the "raw" feces, is available so it is impossible to assess the proportion of this amount deposited at the colony site each season within fecal material. Calculation of this type, however, indicates that deposition could constitute a significant input of radionuclides to a relatively small area that otherwise would receive a yearly input many orders of magnitude lower. Should the birds of the colony constitute a vector for radionuclides to the area via their feces it would be expected that ^{137}Cs input would have peaked at times when concentrations of ^{137}Cs present in the local marine environment were at their highest. Maximal levels would have occurred following the weapons tests of the 1950's and 1960's and the elevated discharges of ^{137}Cs from Sellafield in the mid 1970's. The concept of a pulsed input of ^{137}Cs through the seabird colony would appear plausible given the spatial distribution of the nuclide in both the soil surrounding the base of the talus and in the talus material. In terms of ^{238}U enrichment, it is possible that seabirds have a role in this process as well, as ^{238}U is present in seawater and may be accumulated by their prey species.

A pulsed input of ^{137}Cs may also have arisen via melting of contaminated snow and ice that exists on Svalbard (Pinglot *et al.*, 1994) due to weapons tests and Chernobyl fallout. Such a flush of ^{137}Cs may have been retained by the greater sorbitive properties of soils at the site, producing elevated local levels, whereas the lower retention capacity of surrounding soils did not retain ^{137}Cs to such an extent.

Irrespective of whether or not the colony inhabitants constitute the source of elevated radionuclides observed at the site, it is obvious that the impact of the colony on the area has a significant effect on the radioecology of the site. The alteration of the soil at the site via the input of nutrients and organic materials from the colony has enhanced the capacity of the soil to retain a variety of radionuclides. It is this capacity for retention that obscures the possible source of the radionuclides, as the ability of organic materials to enrich radionuclides from what are essentially background levels means that identification of the source is made more difficult. The most likely scenario is a number of inputs: melting of contaminated ice and snow, normal levels of uranium series nuclides in surface waters or, potentially, transfer of low levels of radionuclides to the site via the activities of the colony inhabitants. Of some interest, however, is the role that such colonies play in the Arctic terrestrial ecosystem. The input of nutrients tends to produce areas of higher productivity than are typical for high Arctic environments. Indicators of this productivity tend to be the amount and diversity of vegetation growing at such sites. It is therefore of some interest that these sites currently display elevated levels of radionuclides and would, in the event of a future contamination incident, probably constitute a long term sink for contaminant nuclides.

Conclusion

A study of a large accumulation of fecal and nesting material and soils below a seabird colony in a High Arctic environment indicated elevated levels of ^{137}Cs, ^{238}U and ^{226}Ra relative to the levels encountered in the general area. The highest nuclide levels are largely associated with a thin humified organic layer and the results indicate that ingress of these nuclides to the soil is most probably from run-off draining from the accumulated fecal and nesting material that has developed at the base of the colony. ^{137}Cs ingress to the soil appears to have peaked at some point in the past as patterns of enrichment at the colony are different to those exhibited by ^{238}U, which must be assumed to be a steady state contribution. The study confirms the hypothesis that the presence of a seabird colony can have a significant effect on the levels of radionuclides in the High Arctic. The results of this study indicate, however, that this effect manifests itself through an enhancement of the affected soils to adsorb and retain radionuclides rather than seabird colonies constituting a source of radionuclides to the terrestrial environment.

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