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Case Study on Radioactive Contamination in Western Arctic Tundra

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1. INTRODUCTION

The High Arctic, a region characterised by its remote and largely inaccessible tundra terrain, was the focus of international field campaigns in 1999, 2012, and 2013. The initial expedition pertained to the freshwater systems of the Canadian Arctic, while the subsequent two projects addressed subjects concerning the tundra vegetation of the shoreline of the Western Arctic. The research material comprised lake sediments and soils, the majority of which were sourced from the Arctic Archipelago. In addition, the material included various species of lichens and mosses, which were collected from southwest Greenland, the Canadian Arctic, and Alaska. The objective of the study was to re-evaluate and supplement the existing knowledge relating to radioactivity contamination levels, trends and patterns in the terrestrial environment of the Western Arctic during the early 21st century. In order to produce a database that would facilitate such an assessment, both deposition and isotopic composition for anthropogenic radioisotopes were analysed. The primary focus was on ⁹⁰Sr, ^{134, 137}Cs, ^{238, 239, 240}Pu, and ²⁴¹Am. In addition to the spatial distribution, the temporal variations of pollution for lake sediments dated using ²¹⁰Pb dating models were also investigated.

2. RESULTS AND DISCUSSION

2.1 Level of anthropogenic radioactivity

The comparison of mean activity concentrations (\pm SE, 1 σ) of chosen radioisotopes was possible for lichens and mosses. The highest levels of ¹³⁷Cs were noted in the Arctic Archipelago and southern Greenland sites, where it reached 75 \pm 42 (69°N 96°W) and 80 \pm 29 Bq/kg (63°N

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 51°W), respectively (Cwanek et al. 2020a,b). The maximum concentrations of 39 ± 14 Bq/kg for ^{90}Sr (70°N 96°W), 4.6 ± 1.2 Bq/kg for $^{239+240}\text{Pu}$, and 1.52 ± 0.47 Bq/kg for ^{241}Am (75°N 95°W) were observed in the northern Arctic Archipelago (Cwanek et al. 2020a,b). Despite the heterogeneity of the findings across different locations, the identification of significant and pervasive geographical dependence proved to be a challenging task. The values of the Pearson's correlation coefficients (ranging from 0.34 to 0.51) indicated positive but rather low correlations between radionuclide concentrations and latitudes, especially in the case of ^{137}Cs . The observed spatial trends are likely to be attributable to the impact of the effective half-life, T_{eff} . It is evident that the deposition of anthropogenic radionuclides, which are derived from the predominant source of radioactivity on Earth – that is, atmospheric nuclear tests – has been estimated to be greater at southerly sites. However, at northern latitudes, where precipitation and temperatures are lower, biological turnover is significantly reduced, leading to a prolonged T_{eff} and, consequently, elevated levels of accumulated pollutants over time.

A further comparison of 137 Cs and $^{239+240}$ Pu inventories (\pm SE, 1 σ) made for lake sediments and soils revealed the following essential quality of Canadian Arctic ecosystems. Specifically, the levels of integrated depositions of man-made radioisotopes were found to be elevated in lake sediments relative to soils from the catchment areas. For instance, the greatest values in the sediment amounted to $3488 \pm 40 \text{ Bq/m}^2$ for ^{137}Cs and $101 \pm 2 \text{ Bq/m}^2$ for $^{239+240}\text{Pu}$ (71°N 123°W), whereas these results in soil reached only $1095 \pm 22 \text{ Bg/m}^2$ and $37.4 \pm 2.4 \text{ Bg/m}^2$ (71°N 123°W), respectively (Cwanek et al. 2021). The process, which was found to be the most probable cause of such differentiation, was determined to be sediment focusing. This phenomenon may occur within the lake basin, indicating the internal resuspension of matter in shallow zones by water currents, followed by transport and descent into deeper zones. Consequently, the sedimentary material present within the lake basin is generally greater in quantity than that resulting from direct atmospheric deposition. The quantitative assessment of the focusing effect of ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu in the reservoirs under study was conducted by calculating the focusing factor, F, as the ratio of inventory in sediment and soil at a given site. As anticipated, the most effective sediment focusing occurred at the lake in the eastern Arctic Archipelago (67°N 81°W). The mean focusing factor (\pm SE, 1 σ) was as high as 6.36 ± 0.48 for 137 Cs and 6.61 ± 0.52 for ²³⁹⁺²⁴⁰Pu (Cwanek et al. 2021).

The research findings indicated the presence of ^{134}Cs traces (Cwanek et al. 2020a), which were exclusively detected in lichen samples collected at the Alaskan site (65°N 165°W). The activity concentration of ^{134}Cs averaged at 14.0 ± 2.9 Bq/kg (\pm SE, 1 σ ; decay corrected to the day of sampling), whereas the mean of the $^{134}\text{Cs}/^{137}\text{Cs}$ isotopic ratio was 1.04 ± 0.14 (\pm SE, 1 σ ; decay corrected to 2011-03-11). The latter value corresponded to the Fukushima fallout signature of 1.033 ± 0.006 .

2.2 Radioactive pollution sources

The analysis of the isotopic ratios of 238 Pu/ $^{239+240}$ Pu, 240 Pu/ 239 Pu, 241 Am/ $^{239+240}$ Pu, $^{239+240}$ Pu/ 137 Cs, and $^{239+240}$ Pu/ 90 Sr was employed to deduce the primary source of contamination within the entire study area. The results were then subjected to comparison with global fallout reference levels (+SNAP 9A).

The slopes of linear trends, fitted to experimental data for lichens and mosses, amounted to 0.0318 ± 0.0013 for 238 Pu/ $^{239+240}$ Pu and 0.364 ± 0.022 for 241 Am/ $^{239+240}$ Pu, corresponding to the global fallout (+SNAP 9A) reference values of 0.025-0.04 and 0.38, respectively (decay corrected to 2013). Furthermore, the values for 240 Pu/ 239 Pu demonstrate consistency with the aforementioned conclusion (Cwanek et al. 2020a,b).

The findings for the Western Arctic vegetation were consistent with those established for the Canadian Arctic about the 238 Pu/ $^{239+240}$ Pu and 241 Am/ $^{239+240}$ Pu ratios for soils, as well as the

²³⁸Pu/²³⁹⁺²⁴⁰Pu temporal variations for lake sediment (Cwanek et al. 2021) (67°N 81°W). An investigation was also conducted into the lake sediments and soils with respect to their ²³⁹⁺²⁴⁰Pu/¹³⁷Cs isotopic composition. In general, for the majority of the sites, the ²³⁹⁺²⁴⁰Pu/¹³⁷Cs results were found to be in rather good agreement with the global fallout signature. Deviations of ²³⁹⁺²⁴⁰Pu/¹³⁷Cs in sediments towards both higher and lower values may be attributable to the migration of radionuclides downward in the cores. Such movement would provide a rationale for the presence of radiocaesium or plutonium beneath 1945 at locations 75°N 99°W and 67°N 81°W, respectively. However, a clear excess of ¹³⁷Cs above the global fallout, dated around the mid-1980s, was noted at site 71°N 123°W, where migration of radionuclides was not detected. It is evident that the presence of radiocaesium in the Canadian Arctic region, as a consequence of the Chernobyl accident in 1986, has had a significant impact on the lake in question (Cwanek et al. 2021).

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