Airborne ^{137}Cs, ^{90}Sr and total beta activity in northern Finland in the 1960s

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Abstract

Air filter samples collected at Sodankylä (67°22'N, 26°39'E) were analyzed for determining concentrations of ^{137}Cs, ^{90}Sr and total beta in surface air at northern Finland during 1963. Activity concentrations of ^{137}Cs, ^{90}Sr and total beta varied seasonally being highest in spring and summer, due to enhanced transportation of aerosols containing radioactive particles from stratosphere to troposphere during warm seasons. Activity ratio ^{137}Cs/^{90}Sr in air of Sodankylä was 0.68(±0.24) – 13(±4), the median value 1.97 suggests the radioactive contamination source to be global nuclear test fallout in 1963. No direct deposition from Novaya Zemlya was detected.
Keywords: cesium, strontium, air, nuclear tests

1 Introduction

Global radioactive fallout had a maximum value in 1963, originating from atmospheric nuclear weapons testing in 1950s and 1960s, conducted mainly by the USA and Soviet Union [1]. The most powerful atmospheric nuclear test (50-58 megatons) was exploded on 30 October 1961 at the height of 4000 m above Novaya Zemlya (approximate coordinates 73.85°N, 54.50°E). The air filters collected within monitoring programme of Finnish Meteorological Institute (FMI) in 1963 were analysed for determining the activity concentrations of ^{137}Cs, ^{90}Sr, total beta and plutonium isotopes during the year. The possibility of some direct deposition from Novaya Zemlya tests to Sodankylä was also examined, because the distance between Sodankylä and Novaya Zemlya is only 1000 km. The possibility of direct atmospheric transport of ^{137}Cs and total beta activity from Novaya Zemlya to Norway has already been investigated, but no clear evidence of such direct deposition to Norway was obtained [2].

2 Materials and methods

2.1 Air sampling and measurements of total beta and gamma activity

Daily aerosol samples were collected by filter sampling at the FMI's Sodankylä meteorological observatory (67°22'N, 26°39'E, h = 179 m above sea level). The total beta activities of the air filter samples were measured in the FMI's laboratory five days after the end of sampling, when the short-lived ^{222}Rn and ^{220}Rn had decayed into ^{210}Pb and stable lead, respectively. In 1963 the measured total beta activity was mainly due to artificial beta emitters and ^{210}Pb with its daughter nuclide ^{210}Bi [3]. In 2007, the daily archived air filter halves from year 1963 were analysed with HPGe gamma spectrometry to determine ^{137}Cs.

Then the filter samples were combined to 176 samples, each covering 1-3 days, for radiochemical analysis.

2.2 Radiochemical separation and measurement of ^{90}Sr

Sr-carrier was added to samples before radiochemical separation procedure for determining chemical recovery of Sr. The filters were ashed in 400 °C and the residue was wet-ashed with concentrated acids. Sr was separated from other radionuclides and matrix by extraction chromatography with TRU® and Sr-resin® (Eichrom Technologies). Finally, separated Sr-fractions were in 15 ml of 0.05 M HNO_3. Sr-fractions were stored for two weeks in order to have radioactive equilibrium between ^{90}Sr and its daughter nuclide, ^{90}Y, in samples. The activity of ^{90}Sr in samples was determined by measuring the activity of ^{90}Y with low-level liquid scintillation counter Quantulus 1220 (Wallac Ltd). The recovery of Sr was determined by measuring concentration of stable Sr with ion chromatography. The separation procedure has been published in detail elsewhere [4].

3 Results and discussion

3.1 Activity concentrations of ^{137}Cs, ^{90}Sr and total beta in air filters

Activity concentrations of ^{137}Cs, ^{90}Sr and total beta in air of Sodankylä were <50 – 13800(±2700) μBq/m^3, <10 –5340(±290) μBq/m^3 and 3700 – 477300 μBq/m^3, respectively, in 1963. Seasonal variation in air concentrations of ^{137}Cs, ^{90}Sr and total beta were similar compared to corresponding variation of plutonium isotopes [4], activity concentrations were higher in spring and summer than in autumn and winter (Figures 3.1 a and b). This is due to the increased transportation of aerosols containing radioactive particles from stratosphere down to troposphere during spring. The maximum total beta activity concentrations are three orders of magnitude higher compared to the natural level due to ^{210}Pb [5].

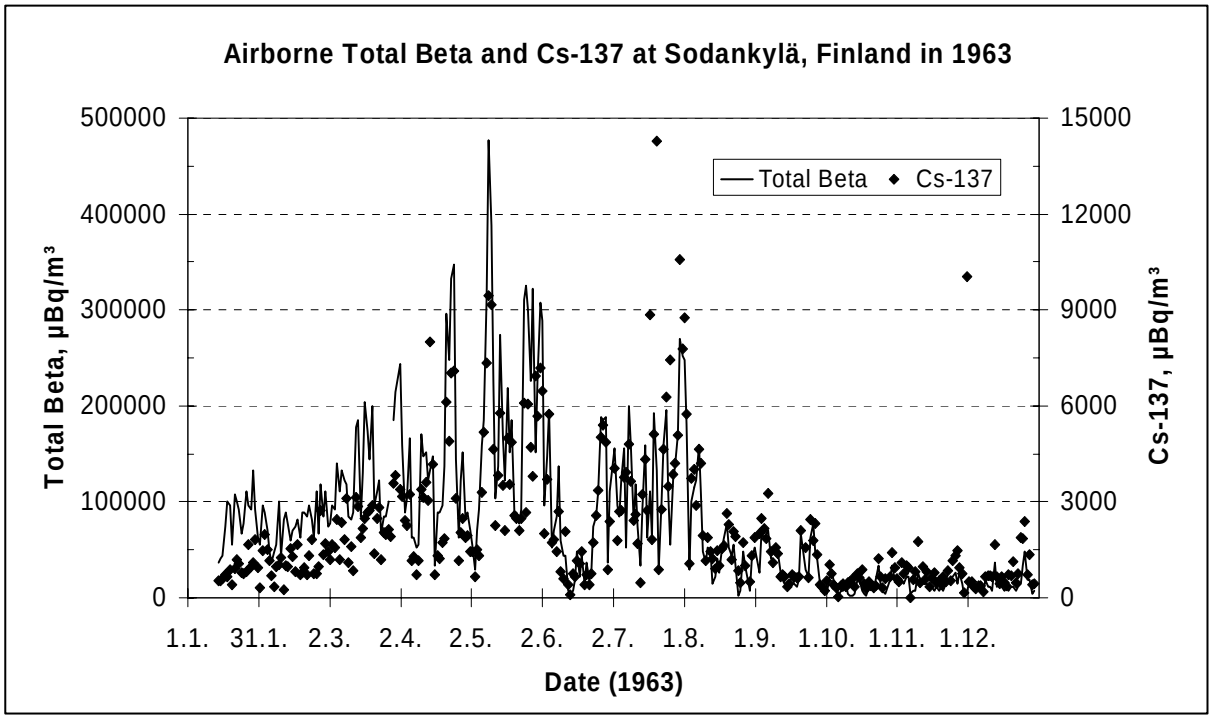

Figure 3.1 a: Activity concentrations of ^{137}Cs and total beta at Sodankylä in 1963.

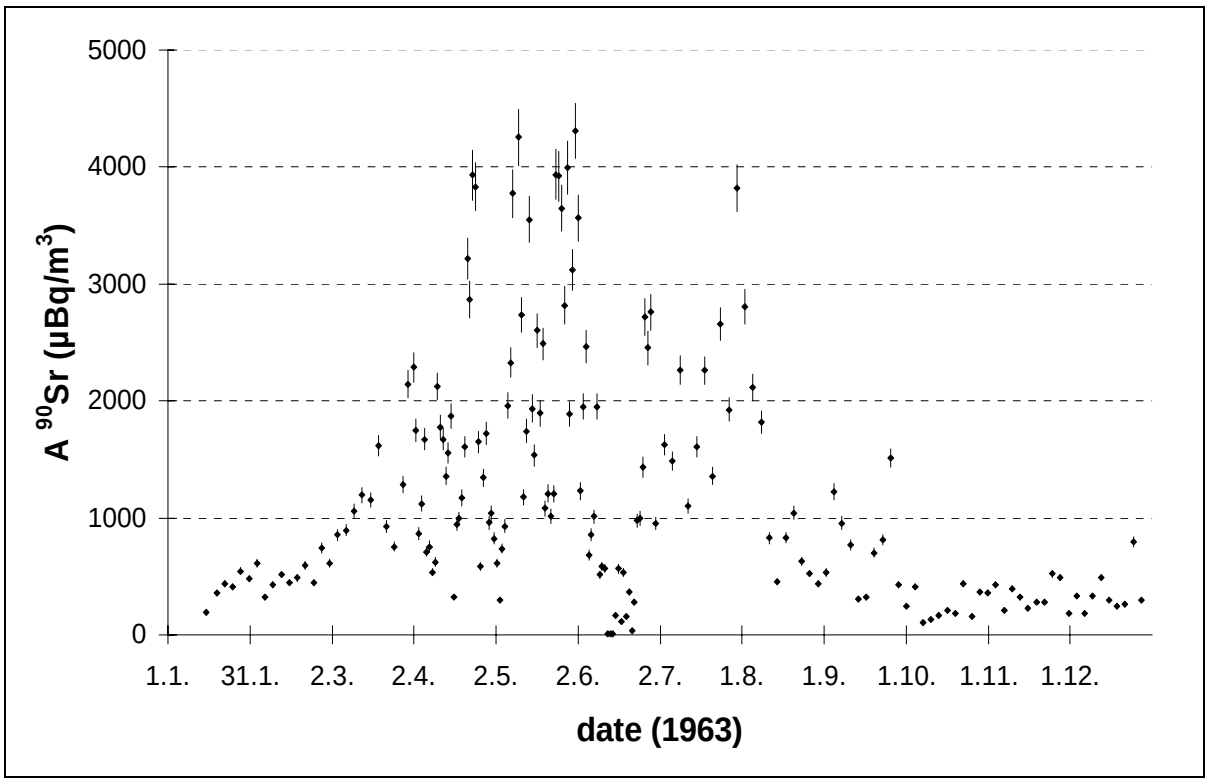

Figure 3.1 b: Activity concentration of ^{90}Sr in air at Sodankylä.

3.2 Activity ratio ^{137}Cs/^{90}Sr in air filters

^{137}Cs/^{90}Sr activity ratio varied greatly in air filter samples, from 0.68(±0.24) to 13(±4), the median value being 1.97 (Figure 3.2). ^{137}Cs/^{90}Sr activity ratio is estimated to be ~1.5 in global fallout from nuclear weapons testing [1], therefore the median value for ^{137}Cs/^{90}Sr represents global contamination from nuclear tests in Sodankylä. Nuclear test sites, such as Novaya Zemlya, "Kraton-3" in Yakutia and Semipalatinsk test site have been found to often have ^{137}Cs/^{90}Sr activity ratio less than one, significantly different from activity ratio in global fallout, and the ratio ^{137}Cs/^{90}Sr at these nuclear test sites can vary between <1 and 20 [6]. Some low values for ^{137}Cs/^{90}Sr ratio in Sodankylä could indicate traces of direct deposition from Novaya Zemlya, but this should be confirmed with other methods, for example by determining ^{240}Pu/^{239}Pu mass ratio for air filter samples, before making any definitive conclusions.

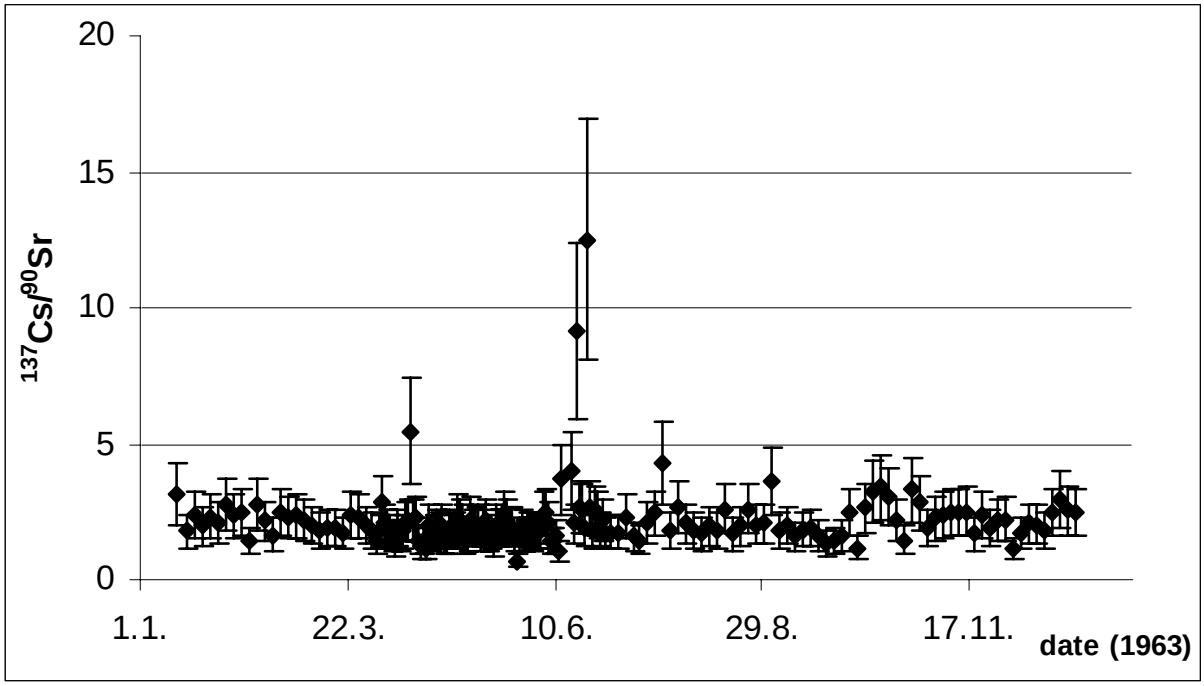

Figure 3.2: Activity ratio $^{137}Cs/\ ^{90}Sr$ in air filters in 1963.

3.3 Activity ratio total beta/(^{137}Cs+^{90}Sr) in air of Sodankylä

The activity ratio total beta/(^{137}Cs+^{90}Sr) was decreasing continuously during 1963 (Figure 3.3), due to decay of short-lived fission products from nuclear tests by Soviet Union in the end of 1962. The origin of radioactive particles in surface air of Sodankylä is different in January-March and April-December, as the half-lives of total beta activity are clearly different for the two time periods. The artificial radioactivity in the northern tropospheric air during the first months of 1963 was dominated by the nuclear tests by Soviet Union at Novaya Zemlya in the end of 1962. On the other hand, in spring 1963 the exchange of air between stratosphere and troposphere increased due to solar heating, and the enhanced vertical mixing in the troposphere brought an aged mixture of radionuclides from other nuclear tests conducted by Soviet Union and USA in large quantities to the ground-level air.

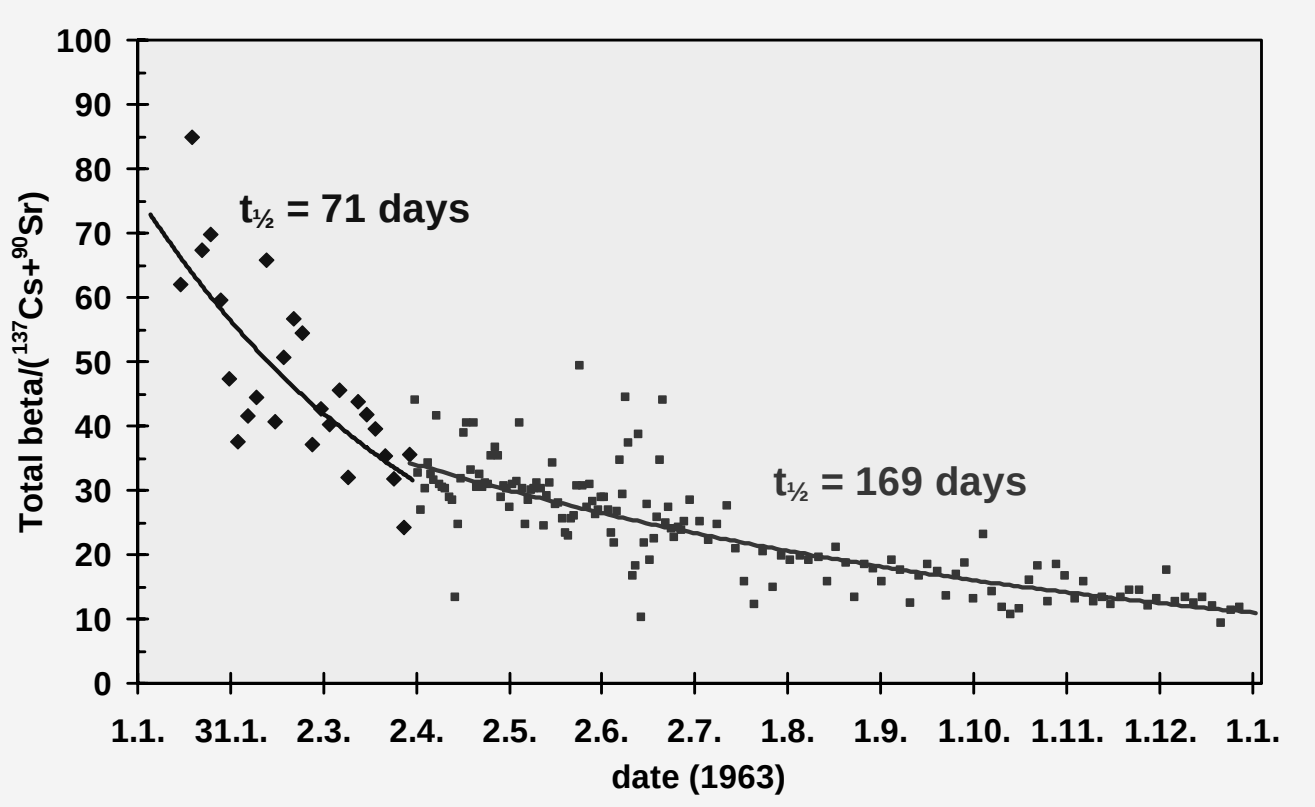

Figure 3.3: Activity ratio total beta/(^{137}Cs+ ^{90}Sr) in air at Sodankylä. The half-life of total beta activity vs. ^{137}Cs+^{90}Sr has been determined for periods January-March and April-December.

4 Conclusions

^{137}Cs, ^{90}Sr and total beta had the highest activity concentrations in air of Sodankylä, in 1963, during spring and summer due to seasonally increased air mass transport from stratosphere to troposphere. The source of artificial radionuclides in ground-level air of Sodankylä in 1963 was the global nuclear weapons testing fallout, although some low ^{137}Cs/^{90}Sr activity ratios were observed during 1963.

References

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