Measurements of Sr-90 Radionuclide in Slovenian Soils before and after Chernobyl Accident

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ABSTRACT

Strontium-90 is a long-lived fission product (half life of 28.7 years) that is globally dispersed in the environment. It had been transported by air masses from the nuclear weapon tests sites in the period of 1951-1980 and also from Chernobyl (1986) and deposited elsewhere, especially over northern hemisphere. Contamination of surface layer (0-10 cm) of undisturbed soil in Slovenia was measured in the middle of the seventies (1973-75) and recently (2002). In parallel, long-lived radionuclide, Cs-137 was measured too, the second campaign was performed ten years after Chernobyl accident. Maps on Sr-90 and Cs-137 contents in soil were elaborated, showing different distributions of area contamination and different levels for a case of nuclear weapon tests and due to Chernobyl accident for both radionuclides. The past contamination from atmospheric nuclear tests Sr-90 and Cs-137 in Slovenian territory was characterised with the high values on western part of the country (with the exception of the coastal region) and typical values of 1.5-2 kBq/m² for Sr-90 and 3-4 kBq/m² for Cs-137. The Chernobyl accident raised the contamination with Cs-137 mostly in northwest part (Alpine region), with an average value of 20-25 kBq/m² for the country. Contamination with Sr-90 was much lower, the existing levels increased for about 0.2 kBq/m². Recently measured levels of Sr-90 in the upper layer of soil hardly approach to 0.3 kBq/m².

1 INTRODUCTION

Sr-90 is radiologically a very important radionuclide that enters into the food chain and accumulates in human bones like calcium. It still nowadays contributes to the majority of the public ingestion exposure from artificial radionuclides (60-80 %) related to global
contamination. It was released into the environment very intensively during 1955-63 as the consequence of military nuclear tests in the atmosphere and during the Chernobyl accident in 1986. Total activity of released Sr-90 was estimated to 630 PBq and from this figure only about 8 PBq is due to Chernobyl accident [1]. The nuclear weapon releases resulted in a cumulative radioactive deposition of 460 PBq reaching the maximum in 1970. An average deposition of Sr-90, estimated for the northern hemisphere (with latitudes 40° -50°) in the middle of the seventies, was about 2 kBq m⁻². This value was estimated on the basis of Cs-137 data for deposition taking into account the ratio of total released activities of both radionuclides (Sr-90: 622 PBq and Cs-137: 948 PBq). Calculated ratio between totally released activities is therefore 1 : 1.6.

The first radioactive contamination of the ground with Sr-90 and Cs-137 in Slovenia was surveyed three decades ago (in 1973-75) by Institute of Occupational Safety [2]. The contents of both radionuclides in the upper layers of soils (0-10 cm) were determined. Almost two hundred of soil samples throughout the Slovenian territory were analysed, on radioactivity content. A half of them was also characterised from the pedologic point of view. In further period only at some points of Slovenian territory (Ljubljana, Kobarid, Murska Sobota, and Krško) the soil contamination measurements were followed on yearly basis within the national monitoring programme of the environmental radioactivity. Long-lived radionuclide Cs-137 was measured in the second campaign performed ten years after Chernobyl accident [3]. This survey was performed by the Institute of Geology using in-situ gamma spectrometry measurements. In the research study that the Institute of Occupational Safety carried out in 2002, soil samples were taken from 16 locations and analysed for Sr-90.

The aim of this paper is to compile the results obtained in all those studies in order to present the territorial distribution levels of Sr-90 and Cs-137 in Slovenian soils in the pre-Chernobyl period and after the accident. The maps with the data on the pre-Chernobyl values were elaborated using the same technique as for results of the subsequent analyses.

2 METHODS

The first sampling strategy was in principle to collect samples from undisturbed grass land over the country and was not based on the uniform grid system. Sampling points were therefore focused on agricultural land use, and so there appeared a lack of samples in the forest and rocky alpine regions. In spite of that, the distribution of sampling points is widespread enough and enables creating the map with data on surface activities of Sr-90 and Cs-137. In the two series, altogether 170 samples were collected. After Chernobyl accident 49 samples was taken for Cs-137 and 16 samples for Sr-90 analyses.

A core sampler was used for sampling of 0-5 cm and 5-10 cm thick layers. An average sample was taken from an area of 100 m². Samples were dried to constant weight, ground, sieved through 2x2 mm² sieve and homogenised. For radiochemical analysis two parallel samples were prepared. For gamma-spectrometric measurements about 0.5 kg of sample was put in Marinelli beaker and waited for at least three weeks to reach the equilibrium between radon and its short-lived decay products.

Strontium was separated from the sample by successive fuming nitric acid precipitations. After chemical separation of the samples the measurement of Sr-90 was
performed with low background anti-coincidence proportional counter. A special separation and subsequent measurement of the daughter radionuclide Y-90 was not performed since no other strontium isotopes were to be expected in the samples. In the first campaign the content of Cs-137 was determined by laboratory gamma-spectrometric measurements with 3x3” NaI(Tl) in units kBq m$^{-2}$. The post-Chernobyl samples were analysed in-situ with gamma-spectrometry (3x3” NaI(Tl)) at 48 locations. Soil profile measurements of Cs-137 activity for typical soils were performed with the high resolution HPGe detector. In-situ data were corrected for the assumed depth profile, previously determined in a limited number of different soils. The maps of Sr-90 and Cs-137 deposition in upper soil layer were elaborated using the SURFER ver. 8 software.

3 RESULTS AND DISCUSSION

Maps on Sr-90 and Cs-137 content in the pre-Chernobyl soils in Slovenia are presented for the first time. The results of Sr-90 in soils in the period 1973-75 showed the local distribution with higher values in the north-western part of Slovenia, as shown on the map (Fig. 1). Higher deposition was found in the regions with high precipitation levels; corresponding correlation factor of nearly $r=0.8$ means well expressed relationship. The lowest results were obtained in the Coastal region and in the northeast (Pannonian basin). The most frequent results were distributed in the interval of 0.5 to 2.2 kBq m$^{-2}$ with the highest values of 3-4 kBq m$^{-2}$ in pasture grassland of the Slovenian Alpine region. The control measurements of the soil layers showed that less than two thirds of the total activity is present in the upper 10 cm, so these values have to be multiplied by 1.6 to get real values of deposited activity. Calculated real values are within the interval of 0.7-3 kBq m$^{-2}$ with maximum of 4-5 kBq m$^{-2}$ in the Alpine region. The average value of 2 kBq m$^{-2}$ (in 1973) is not far from the expected figure of 2.2 kBq m$^{-2}$.

![Figure 1. Map of Sr-90 in the upper layer of soil (0-10 cm) in Slovenia in the period 1973-75](image.png)
If we take into the account only radioactive decay (and neglect penetration into deeper layers or leaching from the surface with precipitation) then these values should have at present a half of the quoted values.

In the time frame of the Chernobyl accident (1986) the levels were 0.5 kBq m\(^{-2}\) measured in Ljubljana and up to 1.1 kBq m\(^{-2}\) in the Soča river basin (near Kobarid) [3].

Measurements in 2002 showed that surface contamination of the undisturbed ground in the depth of 0 to 10 cm significantly decreased in one half life of Sr-90, mostly to the values of 0.1-0.4 kBq m\(^{-2}\) (Fig. 2). The highest value was detected in the upper basin of the Soča river (near 0.7 kBq m\(^{-2}\) ). In general, the results fit quite well the distribution pattern from the seventies, but levels are much lower now. These results are reflected also in a decrease of Sr-90 in milk [6]. Its concentrations in milk declined for 3-4 times from the seventies till nowadays, that is pretty faster than only due to radioactive decay. A migration to the deeper soil layers evidently has an effect of the same order of magnitude.

![Map of Sr-90 in the upper layer of soil (0-10 cm) in Slovenia in 2002](Legend:

<table>
<thead>
<tr>
<th>Color</th>
<th>Activity (Bq/m(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Green</td>
<td>100-200</td>
</tr>
<tr>
<td>Blue</td>
<td>200-300</td>
</tr>
<tr>
<td>Purple</td>
<td>300-400</td>
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<tr>
<td>Orange</td>
<td>400-500</td>
</tr>
<tr>
<td>Red</td>
<td>600-700</td>
</tr>
</tbody>
</table>

Figure 2. Map of Sr-90 in the upper layer of soil (0-10 cm) in Slovenia in 2002

A map of Cs-137 was elaborated too, showing for more than twice higher values as for Sr-90 and almost the same distribution type (Figure 4). Atmospheric nuclear tests gave rise to higher contents in west and northwest part of the country. In the middle of the seventies the average value for Slovenia was 3.5 kBq m\(^{-2}\) in the upper layer of soil (0-10 cm) with maximum values up to 6-8 kBq m\(^{-2}\). The depth profile of the soil showed that of Cs-137 was accumulated in this layer up to 80 % of the deposited activity. The average deposition thorough the country of about 4 kBq m\(^{-2}\) was close to modelled prediction of 3.5 kBq m\(^{-2}\) for northern hemisphere between 40\(^\circ\)-50\(^\circ\) and equals to results from Bavaria [5].
The Chernobyl accident raised the contamination with Cs-137 mostly in northwest and in north part of Slovenia with an average value of 20-25 kBqm$^{-2}$ for the whole country [4] and with maximum values in the Alpine region, exceeding 50 kBqm$^{-2}$ [4], [8]. But ten years after the accident, the values dropped significantly, mostly due to migration of caesium into deeper layers. An intensive use of fertilizers forwarded the penetration downwards.

Figure 3. Map of Cs-137 in the upper layer of soil (0-10 cm) in Slovenia during 1973-75

Figure 4. Map of Cs-137 in the upper layer of soil (0-10 cm) ten years after the Chernobyl accident (1996)
On the assumption that all Cs-137 was deposited at the ground surface, the median value from in-situ measurements was calculated to be 2 kBq m\(^{-2}\). Taking into account the experimentally determined depth profile the median value of 6 kBq m\(^{-2}\) seems to be more realistic (Fig. 4).

4 CONCLUSIONS

The area contamination with Sr-90 and Cs-137 shows different distributions and different levels for the cases of nuclear weapon tests and for the Chernobyl accident for both radionuclides.

The past contamination from atmospheric nuclear tests Sr-90 and Cs-137 in Slovenian territory was characterised with the high values in the west part of the country (with the exception of the coastal region) with average values for the whole territory of 1,5-2 kBq m\(^{-2}\) for Sr-90 and 3-4 kBq m\(^{-2}\) for Cs-137.

The Chernobyl accident raised the contamination with Cs-137 mostly in northwest part (Alpine region), with an average value for the whole country of 20-25 kBq m\(^{-2}\). Contamination with Sr-90 was much lower and increased the existing levels for only about 0,2 kBq m\(^{-2}\), that is for about 10 %. Recently measured levels of Sr-90 in the upper layer of soil (0-10 cm) hardly exceed 0,3 kBq m\(^{-2}\). Chernobyl accident considerably changed the surface contamination of the ground with Cs-137. The surface layer of an undisturbed soil still nowadays contains twice as much Cs-137 as before Chernobyl. In spite of that Chernobyl Sr-90 enhanced the previous values for a quite small proportion, it is - due to its high radiotoxicity - nevertheless responsible for the majority of public exposure caused by the global contamination.

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