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Some Aspects of the Behaviour of ^{90}Sr in the Environment

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Summary

By determining ^{90}Sr apparent half-lives in soil, grass and milk over several years, it was shown that the concentration of Chernobyl ^{90}Sr has rapidly decreased in these samples. Migration in the soil sublayers is certainly the major cause of this disappearance, as shown by profiles. But grass is also involved in this soil decontamination; indeed, the $^{90}Sr/Ca$ ratio is about 4 times higher in grass than in the corresponding soil. An estimation of the ^{90}Sr migration rate is given and an explanation of the phenomenon is discussed.

Introduction

The deposition of ^{90}Sr on land and the transfer to humans by ingestion is the most important pathway for human exposure [1]. Following the nuclear tests in the atmosphere, the survey of ^{90}Sr contamination in environmental samples was undertaken. Since 1976, samples of soil, grass and milk have been regularly collected in 4 locations in Switzerland. After the Chernobyl accident, which brought sufficient radioactivity to Switzerland to be measured, ^{89}Sr and ^{90}Sr were determined in a large range of environmental and food chain samples. Curiously, this Chernobyl ^{90}Sr has vanished rapidly from the upper part of the soil, indicating that its chemical form was different from the one deposited during the sixties. The aim of this work is to try to explain the behaviour of this ^{90}Sr in the environment by analyzing the concentration of ^{90}Sr versus time, its profile in soil and its transfer from soil to grass and from grass to milk.

Experimental

As ^{90}Sr and its daughter ^{90}Y are practically pure β^- emitting radionuclides [2], their measurement must be carried out with thin sources after a radiochemical separation, particularly when dealing with environmental samples. Furthermore, if the samples are contaminated by other fission or activation products, this chemical separation has to be specific for strontium and yttrium.

Samples

Samples of soil (0–5 cm), grass and milk have been collected twice a year (spring and fall) from 4 locations in Switzerland. The milk was taken from cows grazing near the land where soil and grass samples were collected. This allowed the determination of transfer factors.

After the Chernobyl accident, soil samples have been collected in southern Switzerland and ^{90}Sr profiles were determined on 2 cm slices (0–10 cm).

Chemical separation

Details of the separation technique used for determining the ^{90}Sr in environmental samples have been described in an earlier paper [3]. Prior to calcination (12 h, 600 °C), soil and grass samples were air-dried, milk samples were freeze-dried. 5 g of ashes were used in the separation process. In the case of milk, the ashes were directly dissolved in HCl together with a strontium carrier. For soil and grass ashes, an alkaline fusion was necessary to dissolve the sample completely; the carrier and the alkaline earth were precipitated in the carbonate form and dissolved in HCl. The separation of strontium was achieved on two ion exchange columns (Dowex 50) using first a diaminocyclohexane tetraacetic acid (DCTA) solution and then a sodium citrate solution as eluant. This latter solution was kept standing for about 10 days to let ^{90}Y grow. After adding an yttrium carrier, it was separated from strontium on another ion exchange column (Dowex 50) using a sodium malonate solution as eluant. Finally, yttrium oxalate was precipitated and a thin source was prepared by centrifugation on a special device. Both strontium and yttrium separation yields were determined using atomic absorption and complexometric titration respectively.

Activity measurements

To determine ^{90}Sr, the ^{90}Y activity was measured. The higher β^- energy (2.3 MeV) led to a better counting efficiency and the short half-life (64.5 h) of ^{90}Y [2] allowed a radionuclidic purity control. To ensure a low background, the source activity was measured in anticoincidence with a plastic scintillation counter [4] surrounding the gas flow (helium-isobutane) GM counter equipped with a very thin mylar window ($0.9\ mg \cdot cm^{-2}$). Furthermore, the whole counting device was protected by a shielding made of steel, paraffin and a large layer of lead. In these conditions, the background was found to be 0.2 cpm. The source

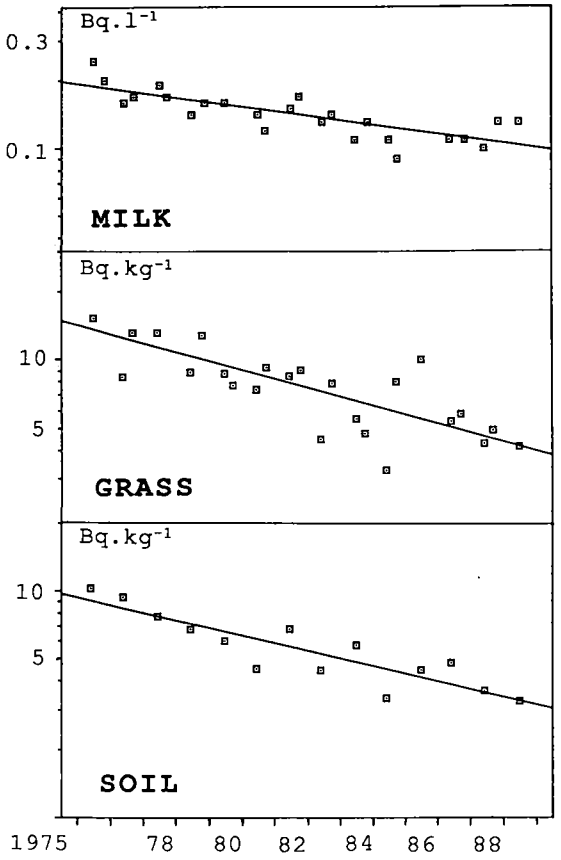

Fig. 1. Typical evolution of ^{90}Sr concentration in soil, grass and milk as a function of time.

was measured typically for 100 hours and intermediate results were automatically taken in by a minicomputer every 6 hours to allow the ^{90}Y purity control. In these conditions, determination limits as low as 0.03 $Bq \cdot kg^{-1}$ can be achieved [5].

Results and discussion

^{90}Sr concentration in soil, grass and milk

Since 1976, samples of soil, grass and milk have been analyzed twice a year in four locations: Grangeneuve and Mühleberg in the western part of Switzerland, Leibstadt and Gösgen in the northern part. At the beginning of the survey, the ^{90}Sr concentration was found to be in the 10 $Bq \cdot kg^{-1}$ range for soil and grass and 10^{-1} $Bq \cdot l^{-1}$ range for milk [6]. These concentrations continuously decreased in all types of samples with an average apparent half-life significantly different from the physical half-life (28.5 y).

Figure 1 shows the results and the typical decay curves obtained for the three kinds of samples. For grass, the results are a little more scattered than those of soil and milk, but the linearity test remains valid, as for all the other curves.

Table 1 gives the apparent half-lives measured at the four locations; the physical half-life was not taken into account. A statistical test demonstrated that all

Table 1. Apparent half-lives

Location	Soil	Grass	Milk
Grangeneuve	12.3±3.6	11.6±3.9	14.8±2.3
Mühleberg	9.0±1.3	7.6±1.3	14.5±2.6
Gösgen	7.8±0.9	6.7±1.1	10.1±2.7
Leibstadt	8.9±1.5	13.3±8.9	12.5±2.5

the curves for a given kind of samples are parallel, allowing thus the calculation of an average apparent half-life. Furthermore, this test showed that the apparent half-lives measured for soil and grass are equal with an average value of about 9 years. In the case of milk, the average apparent half-life seems to be a little higher (13 years) than this latter value.

The analysis of grass and milk in 1986 showed an increase of the ^{90}Sr content in these samples [7]. But during the following years, the values do not show a recollection of this event. The Chernobyl ^{90}Sr seems to have vanished completely from the soil, grass and milk. How and why are the two questions.

^{90}Sr profiles in soil

To vanish from the soil, ^{90}Sr could follow two pathways: rapid underground migration or uptake from the soil by plants, for example. To study these two possibilities, soil samples were taken by slices of 2 cm in the southern part of Switzerland (Genestrerio) which was the area most exposed to the Chernobyl contaminated rain. The location of the sampling was chosen close to a soccer field at the bottom of a flat valley. In contrast to the samples analyzed before, the soil was quite sandy but there was still grass growing on it. Only a few days after the accident, the first set of samples was collected. ^{90}Sr concentration was found in the 30 $Bq \cdot kg^{-1}$ range in the first 2 cm layer. One month later, this concentration had decreased to about 20 $Bq \cdot kg^{-1}$ and two months later the value was still lower. Two more samplings were taken one and two years later, respectively. As the sampling spot was not disturbed, except for mowing, the results are easily comparable.

Figure 2 shows the evolution of the ^{90}Sr profiles. As the total activity of one set of samples (all layers together) can change a little from one sampling to the other, the results are presented as fractions. The error on a single fraction does not exceed 0.02. For the first set (21.05.86), only one layer was taken due to the emergency situation at that time. However, as the ^{90}Sr concentration in the second and third layers of the second sampling (18.06.86) were still very low, the same values were attributed to the first sample. For the lower layer (6–10 cm), the ^{90}Sr concentration did not change from 1986 to 1988; an average value was thus taken for all the samples.

As shown on Figure 2, the ^{90}Sr migrated quite rapidly from the surface to the second and third layers.

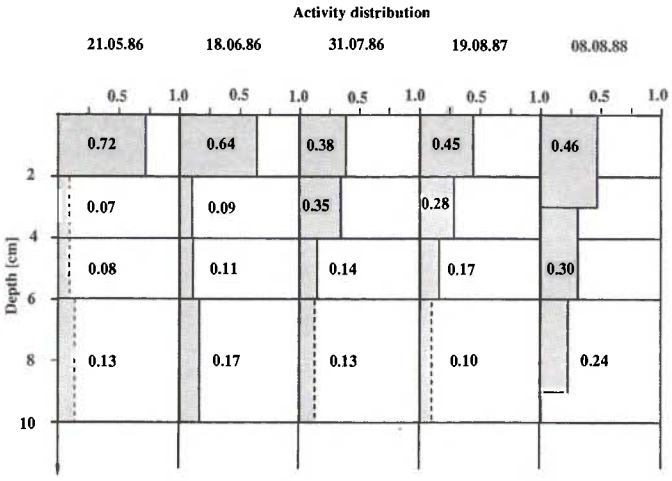

Fig. 2. Profiles of ^{90}Sr activity in a soil from southern Switzerland.

Table 2. Transfer factors normalized to the calcium content of the samples

Locations	Soil-Grass	Grass-Milk
Grangeneuve	2.3+1.9	0.12+0.05
Mühleberg	3.0±1.6	0.14±0.05
Gösgen	4.0+2.5	0.26±0.05
Leibstadt	5.0+2.3	0.21±0.05
Average	3.6+0.6	0.22±0.06

A rough estimation of the migration rate leads to $5 \cdot 10^{-3}$ Bq $\cdot$ kg^{-1} $\cdot$ cm^{-1} $\cdot$ h^{-1} in the first 100 days after the accident.

After one year, the profile shows a small increase of the ^{90}Sr concentration in the first layer, probably due to the soil biologic activity. In the second layer, the ^{90}Sr concentration has slightly decreased while increasing in the third layer, as expected. The migration rate from the second to the third layer is estimated to be about $2 \cdot 10^{-4}$ Bq $\cdot$ kg^{-1} $\cdot$ cm^{-1} $\cdot$ h^{-1}.

Finally, after two years, the situation has stabilized in the three first layers while the ^{90}Sr distribution shows a relative increase in the last layer. These latter results are, however, difficult to compare with the previous ones because the layers' thickness was different.

This study confirms the previous observations given by the apparent decay curves. Indeed, the Chernobyl ^{90}Sr migrated faster than that deposited during the sixties by the nuclear tests fallouts.

^{90}Sr transfer

To test the second way of disappearance of ^{90}Sr from the soil surface, soil-grass and grass-milk transfer factors were determined in the same 4 locations investigated during several years. To make the results easily comparable, the ^{90}Sr activity was normalized to the calcium content of the samples. The transfer factors were thus defined as the ratio of the ^{90}Sr activity corresponding to 1 g of calcium:

$$TF_{soil\text{-}grass} = \frac{(^{90}Sr\,activity \cdot g^{-1}\,Ca)_{grass}}{(^{90}Sr\,activity \cdot g^{-1}\,Ca)_{soil}}$$

and

$$TF_{grass\text{-}milk} = \frac{(^{90}Sr\,activity \cdot g^{-1}\,Ca)_{milk}}{(^{90}Sr\,activity \cdot g^{-1}\,Ca)_{grass}}.$$

Table 2 shows the average results for each location and the average of all individual values.

In the case of ^{90}Sr transfer from soil to grass, it is obvious that the scatter of individual results is large. This is probably due to the dispersion of the calcium content in soil. The average soil-grass transfer factor shows, however, a significant uptake of ^{90}Sr by plants. This fact could again explain partially the vanishing of ^{90}Sr from the soil.

For the transfer factors of ^{90}Sr from grass to milk, the standard deviations are significantly lower, probably because the calcium content both in grass and milk are more steady than in the soil. The average grass to milk transfer factor shows that cows play the role of a decontamination step in the food chain. Part of the ^{90}Sr intake gets certainly its way to the cow's skeleton but this was not investigated. As for humans [8], ^{90}Sr is probably preferentially excreted in urines and in feces.

Conclusion

This study has shown clearly that in the few months following the Chernobyl accident, the freshly deposited ^{90}Sr has migrated underground faster than that from the bombs fallout, already in the soil.

One explanation of this behaviour could be linked to the chemical reactivity of the ^{90}Sr: the temperature at the spot of a nuclear explosion is of course much higher than that in the case of the Chernobyl accident.

The sorption kinetics of ^{90}Sr on the soil material could lead to a further explanation. Indeed, immediately after deposition, ^{90}Sr could behave quite differently than 10 years after the soil contamination. In the case of Chernobyl, the samples were collected right away whereas the first analyses (1975) were undertaken several years after the bomb tests.

Further laboratory investigations are under way to try to better understand the phenomena.

Acknowledgement

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