Long-term Availability of Sr-90 in Foodstuff after Nuclear Fallout

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INTRODUCTION

Among the many radionuclides possibly released in a severe reactor accident or a nuclear weapon's detonation ^{137}Cs and ^{90}Sr are of particular importance for the long-term exposure. Due to their long physical half-life of 30 and 28.5 years, respectively, the exposure of the population by these nuclides will last for decades and has to be taken into account in estimating the total 50 year-exposure caused by the fallout and in evaluating countermeasures with regard to long-term land utilization. However, as already observed after the weapons' test fallout, their effective decrease in the environment with regard to the population exposure follows a much shorter effective half-life reducing the exposure of the public much quicker than estimated from the physical half-life (1,2).

^{90}Sr contributed to more than 80 % of the exposure after the atmospheric nuclear weapons' tests and also to a significant extent in past accidents. The long-term exposure of a nuclear fallout by this radionuclide therefore, is of particular importance. However, while in the past the long-term bio-availability of ^{137}Cs was investigated (3,4), the long-term decrease of ^{90}Sr was not comparably investigated. It should be noted that the observation of the effective half-life after a short-term nuclear fallout was not easily obtainable after the weapons' tests due to the fact that repeated detonations occurred for a period of several years and the dispersion occurred to the upper atmosphere from where the fallout continued for a long period. The atmospheric deposition, therefore, occurred for several years and blurred the observation of the long-term decrease as caused by the much lower soil-plant transfer.

The long-term availability of ^{90}Sr in milk, therefore, was investigated as a main indicator of the environmental behavior of this radionuclide and as the most important foodstuff with regard to the ^{90}Sr-intake of both the infant and the adult. An estimate of the dose to infants and the adult in central Europe 35 years after the end of atmospheric nuclear weapons testing and 14 years after the reactor accident in Chernobyl is presented and compared to the dose of other long-lived radionuclides in the environment.

Since ^{137}Cs is easier to determine and therefore often used as main indicator for the contamination of the environment, ratios of ^{90}Sr to ^{137}Cs are given for the period after the weapons tests and after the Chernobyl accident up to the present time. The variation in this ratio with time and its causes as well as the differences in environmental decay of these two radionuclides are discussed.

MATERIALS AND METHODS

Samples of milk were taken in different provinces of Austria and investigated with regard to their content of ^{90}Sr and ^{137}Cs. The determination of the ^{137}Cs activity concentration in the milk samples was performed in 5 l-Marinelli-beakers and by using average measurement times of 50000 - 80000 s on a HPGe-detectors with 20% and 30 % relative efficiency. With very low activity samples the measurement times were extended to 200000 s. By this a detection limit of 0,03 Bq/kg ^{137}Cs in dry matter, or of 0,017 Bq/kg ^{137}Cs for the prolonged measurement time, was obtained. Milk samples were leophylisized and ashed after the ^{137}Cs-measurement. Ashing was performed at temperatures of 550 °C to destroy fat fractions which might disturb the dissolution of the sample and analysis of ^{90}Sr.

The ash samples were leached by concentrated HNO_3 and the strontium chemically separated using the crown ether method (5). After chemical separation of the samples the measurement of ^{90}Sr was performed by LSC on a Wallac 1220 QUANTULUS. A special separation and measurement of the ^{90}Y was not performed since no other Sr-isotopes but ^{90}Sr were to expected in the samples. For each sample replicate measurements were performed to assure that no other shorter-lived beta-emitters of equivalent β-energy were present in the sample. By deploying measurement times of 3 x 30000 s a detection limit of 5 - 8 mBq in the sample was obtained. According to the original sample volume of 1 – 5 kg, the minimum detectable activity concentration amounted to 1 – 8 mBq kg^{-1} f.m. which was sufficiently low to detect the extremely low activity levels still present in foodstuffs.

RESULTS

The results of the activity measurements are - grouped according to the different climatic and fallout regions of Austria - given in table 1. The Alpine regions showed the highest activity depositions both after the fallout from nuclear weapons' tests and the Chernobyl accident. This is particularly true for the fallout after the weapons' fallout and for the dominant radionuclide of this fallout, ^{90}Sr, and to a lesser degree to the Chernobyl accident and its dominant radionuclide ^{137}Cs. The ratio of the radionuclides thus varies according to the different regions and varying fallout levels after the two events.

Table 1 Activity concentration of long-lived radionuclides in milk of different sites in Austria in 1997

Site	Activity concentration [Bq kg^{-1}] ^{90}Sr	^{137}Cs	^{40}K	ratio ^{90}Sr/^{137}Cs
Eastern Pannonian region	0.0534 ± 0.0205	0.052 ± 0.034	40.4 ± 1.2	1.03 ± 0.78
Alpine foothills, low fallout	0.0616 ± 0.0349	0.104 ± 0.053	49.0 ± 17.9	0.59 ± 0.45
Alpine foothills, high fallout	0.1218 ± 0.0429	0.596 ± 0.381	52.8 ± 17.7	0.20 ± 0.15
Average	0.078 ± 0.034	0.247 ± 0.329	49.4 ± 14.3	0.79 ± 0.63

The average activity concentration of ^{90}Sr in the investigated samples was 0.078 ± 0.034 Bq kg^{-1}. In higher fallout regions the activity concentrations ranged up to 0.18 Bq kg^{-1}, in low fallout regions values of less than 0.03 Bq kg^{-1} were observed.

The average ratio of ^{90}Sr to ^{137}Cs was found to be 0.79 ± 0.63. However, strong variations in this ratio were observed (6) which are also visible in table 1 for the different climatic and fallout regions. The lowest ratios are observed in the mountainous region with typically high precipitation levels and therefore high deposition values after the weapons' test fallout. In the low rainfall regions of the Eastern Pannonian plains the highest ratios are observed. This may be explained by the fact that the fallout after the weapons' tests was significantly higher in the mountainous region and therefore the ^{90}Sr much higher there, while in the lower Pannonian and Alpine foothill region the fallout generally was lower resulting in lower ^{90}Sr depositions. The Chernobyl fallout, dominated by ^{137}Cs, was not so much depending on the yearly average precipitation in the respective region, but - although strongly varying from site to site due to locally heavy rainfalls - showed rather intensive variations within each of the three regions.

In order to determine the average exposure of the population by ingestion of ^{90}Sr via milk consumption, the number of samples taken and investigated may not seem sufficient. Since a significant increase in sample number was not possible due to the high efforts in sample preparation with each sample, another approach was chosen. By determining the average ratio of ^{90}Sr to ^{137}Cs and multiplying this with the average ^{137}Cs activity concentration, the average activity concentration of ^{90}Sr was to be determined. The average ^{137}Cs activity concentration in milk had been determined before in a large number of samples (about 250 samples) all over Austria (3,7).

Due to the rather high variation of the ratio of ^{90}Sr to ^{137}Cs, however, it seemed reasonable to look at the ratio in detail. Therefore, the ratio in each sample was plotted versus the ^{137}Cs activity concentration as shown in

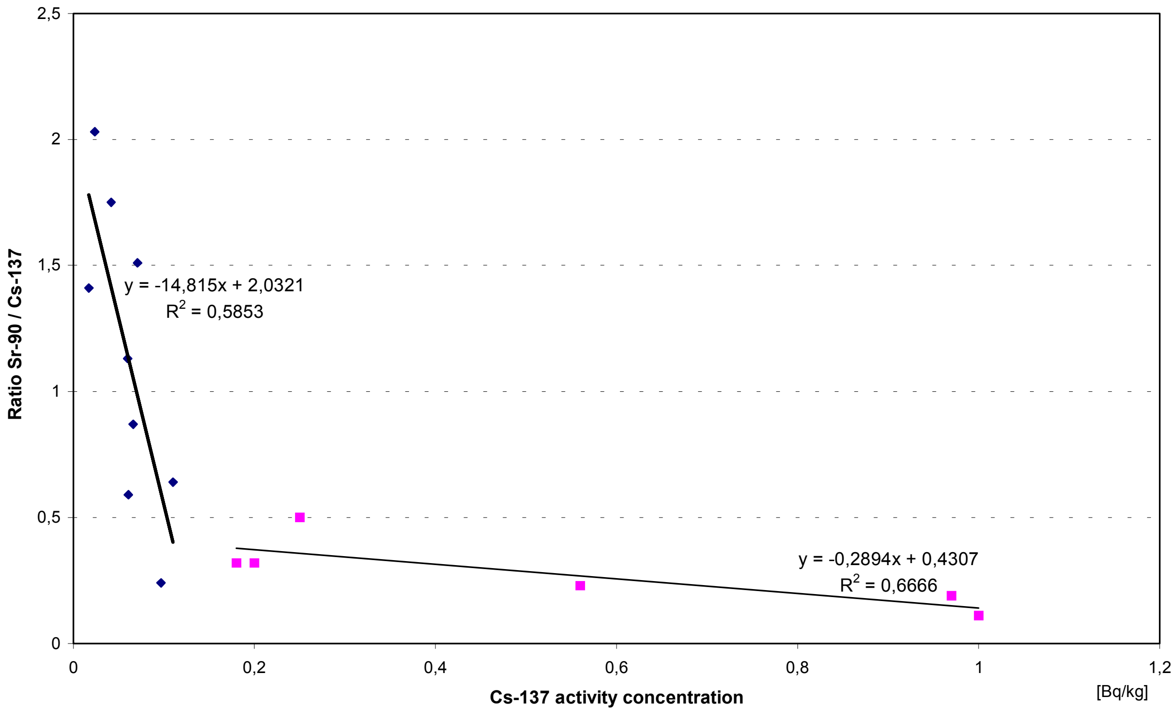

Figure 1 Ratio of ^{90}Sr to ^{137}Cs in milk samples versus the ^{137}Cs activity concentration

figure 1. Obviously, two cohorts of these ratios exist which may be explained by the fact that the deposition in the low precipitation region of Eastern Austria was low both in ^{90}Sr and ^{137}Cs, i.e. both after the weapons' test and the Chernobyl fallout. In the other regions the deposition varied significantly with annual precipitation (^{90}Sr) and with local fallout conditions according to rainfall after the Chernobyl accident (^{137}Cs).

The average ^{137}Cs activity concentration in milk as determined in a nation-wide study in 1997 (7) amounted to 0,50 Bq kg^{-1}. According to figure 1 this yields an average ^{90}Sr / ^{137}Cs ratio of 0,286. From this ratio and the average ^{137}Cs activity concentration an average ^{90}Sr activity concentration of 0,14 Bq kg^{-1} in milk is derived.

The derived average activity concentration is about 1.8 times higher than the value derived from the cohort of samples investigated in this study. This is not a contradiction to the validity of the approach, on the contrary, it demonstrates how essential it may be to have a sufficiently large cohort to obtain average exposure values for the population. Obviously, the sample cohort used for the determination of the ^{90}Sr-content had a bias with regard to the number of the samples from low lying areas of the Pannonian climatic region.

According to Lagoni et al. (8) from the ^{90}Sr activity concentration in milk the activity concentration in other milk products may be derived. This is shown in table 2. Using these values and average foodstuff consumption rates as given by the Austrian Statistical Bureau (9), an annual intake of 19,6 Bq by milk and 3,9 Bq by cheese and 0,08 Bq by other milk products is derived for the adult (6). If the major contribution to the intake comes from transfer from soil (no fallout from air), the contribution of milk and milk products including rennet cheese to the total intake of ^{90}Sr is approximately 57 % (6,10). From this a total ^{90}Sr intake by all foodstuffs in 1997 of 41 Bq may be derived. With a dose conversion factor of 2.8 10^{-8} Sv Bq^{-1} (11) this would yield an internal effective exposure by ^{90}Sr of 1.2 µSv per annum. The exposure of the critical organ (bone) would amount to 12 µSv.

Table 2 Activity concentration of ^{90}Sr in milk products derived from the concentration in milk

milk product	ratio of activity concentration in milk product to activity concentration in milk according to Lagoni et al. (8) ^{90}Sr	^{137}Cs	^{90}Sr activity concentration [Bq kg^{-1}]
milk	1,000	1,000	0,143
cream	0,408	0,842	0,058
butter	0,106	0,403	0,015
butter milk	0,186	0,385	0,027

rennin cheese	30,97	0,601	4,429
fresh cheese	2,645	0,614	0,378

For the 1-year old infant an annual intake of 25,2 Bq by milk and 2,2 Bq by cheese is derived (6). Under the same conditions with no fallout from air the contribution of milk and milk products including rennet cheese to the total intake of ^{90}Sr of the 1-year infant is approximately 80 % (6,10). From this a total ^{90}Sr intake by all foodstuffs in 1997 of 34.3 Bq may be derived. With a dose conversion factor of 7.3 10^{-8} Sv Bq^{-1} (11) an internal exposure by ^{90}Sr of 2.5 µSv (effective dose) per annum for the 1-year infant is derived. The critical organ exposure would amount to 15 µSv (bone).

For the 5-year old child an intake of 11,0 Bq by milk and 2,1 Bq by cheese and other milk products is derived (6) which gives under the same assumptions a total ^{90}Sr intake by all foodstuffs of 19,3 Bq and via a dose conversion factor of 4.7 10^{-8} Sv Bq^{-1} an internal effective exposure by ^{90}Sr of 0.9 µSv in 1997.

These dose values are equivalent to about 0,4 % of the natural internal exposure. These extremely low exposure values demonstrate that basically no exposure remains from weapons' tests and Chernobyl fallout 33 years after the end of above-ground nuclear weapons' testing and 12 years after the Chernobyl accident.

LONG-TERM DECREASE OF ^{90}Sr-ACTIVITY CONCENTRATION

For a contaminated territory the long-term bio-availability of long-lived radionuclides in plants and foodstuffs derived thereof is of utmost importance, both for the long-term utilization of the area and appropriate decisions on land management, relocation of people and resettlement after the "end" of the contamination phase. As a consequence, the activity concentration in foodstuffs and its time course is considered a vital question in the context of highly contaminated territories. Although the contamination of the Austrian territory with ^{90}Sr may not be considered high, neither after the weapons' test fallout nor after Chernobyl plume, it may serve as good indicator of the timely course of the activity concentration. This is particularly relevant as the country is segmen-

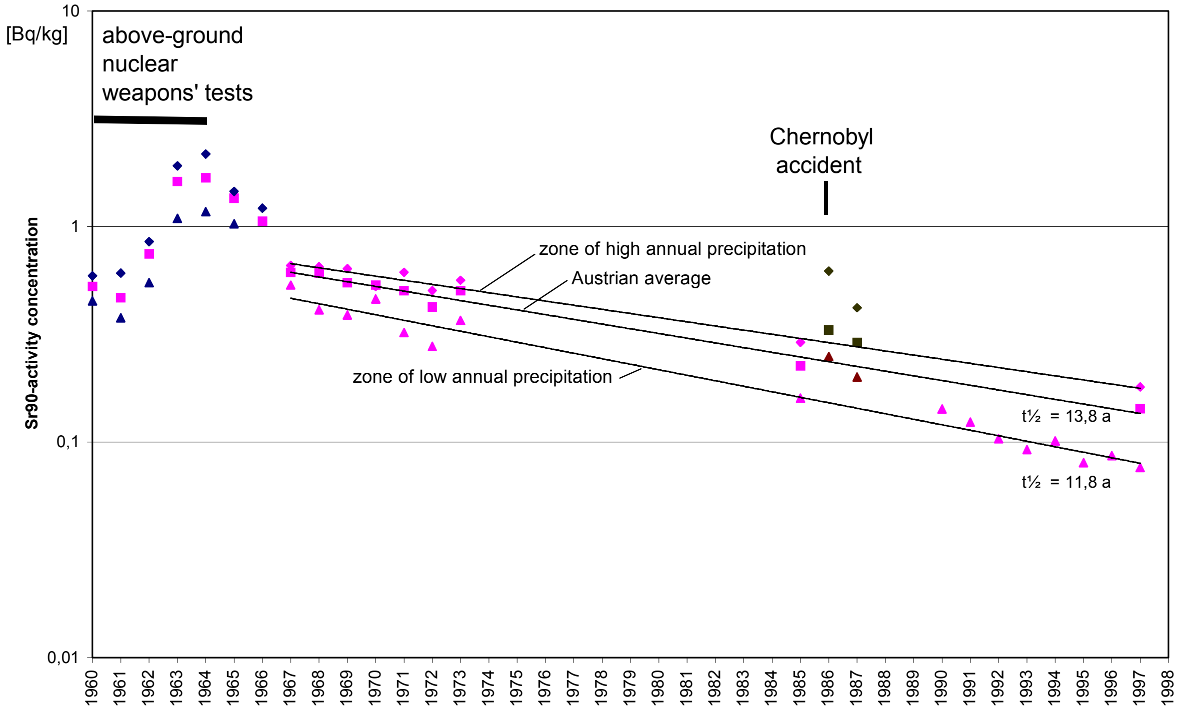

Figure 2 ^{90}Sr activity concentration in milk samples in the years 1960 - 1997

ted in mountainous and hilly parts as well as plains with significantly different ecological parameters with regard to the long-term bio-availability.

The activity concentration in milk from the nuclear weapons' tests until today is given in figure 2. Due to the long time-span the data had to be taken from different sources. Activity concentrations in the years 1960 to 1973 were taken from measurements of the Federal Food Control Laboratory (1), data in the years 1985 - 1987 from an investigation of the ^{90}Sr fraction in the Chernobyl fallout and its contribution to the total dose after the Chernobyl accident in Austria (10). Data for 1997 were recently measured in our own laboratory (6) and the data in 1990 - 1997 were measured by the Food Control Laboratory in a survey of the ^{90}Sr concentration of Viennese consumer milk which is representative for the low precipitation zone (12).

The data in figure 2 are given for the high precipitation zone (Alpine zone) and the low rainfall zone of the lower plains (Pannonia) as well as for the Austrian average (average over all 3 zones). Each zone shows a significant increase in ^{90}Sr content in milk in 1962 - 1964, the time when the most intensive above-ground test series took place. Fallout at that time amounted to approximately 3,3 kBq m^{-2} ^{90}Sr on average during the whole period of which about 0,7 kBq m^{-2} were deposited in 1963 and 0,8 kBq m^{-2} in 1964. In the high precipitation zone the fallout was about 20 % higher than average, in the low zone about 40 % lower.

After the end of the above-ground detonations the activity concentration in milk decreases to about one third of peak activities in the next three years which is equivalent to an effective half-life of 2.7 y. After that period the further activity decrease is slow as the contamination of the plant is virtually only caused by transfer from soil to plant.

The ^{90}Sr activity concentration in 1986 and thereafter are caused by both the fallout of the weapons' tests and the Chernobyl accident. In figure 2 an increase from pre-Chernobyl values by about 50 % is visible as a consequence of the Chernobyl fallout. This implies that still 50 % of the activity concentration in plants after the Chernobyl fallout is still caused by the fallout from the atmospheric weapons' tests. Although the ^{90}Sr from the fallout of the weapons' test cannot be distinguished from that of the Chernobyl fallout as such, it was possible to evaluate the fraction of the Chernobyl ^{90}Sr in the samples taken in 1986 and 1987 (10). In measurements of air by a high volume sampler (3000 m^3 sample) (13) and in rainwater a ratio of ^{90}Sr / ^{137}Cs of 0.040 was established (10). By using this fraction and the fact that the ^{137}Cs from weapons' fallout had decreased to immeasurable levels by 1986, the ^{90}Sr activity concentration in milk was derived in each milk sample from the relative transfer factors for strontium and cesium (14). From this the fraction of ^{90}Sr in milk caused by the Chernobyl accident and that still due to the weapons' fallout could be evaluated (10). These values for weapons' ^{90}Sr are also given in figure 2 for the Austrian average in 1986 and 87. They fit well in the long-term decay of weapons' fallout ^{90}Sr and to the activity concentration determined in 1985.

Despite comparatively high fallout levels in 1986 as compared to other countries, the average deposition of ^{90}Sr in Austria was only 0,9 kBq m^{-2} (15), while after the nuclear weapons' tests it amounted to 3,3 kBq m^{-2} (15,16). Due to this much higher amount after the weapons' tests the activity concentration in milk quickly "returned" to pre-Chernobyl values after the direct contamination phase. The activity concentrations after 1990 (figure 2) are caused to 75 % by the weapons' test fallout and to 25 % by the Chernobyl fallout. Taking this into consideration, the effective half-life of the bio-availability of the ^{90}Sr from weapons' tests fallout would amount to 10,9 y rather than 11,8 y for the low precipitation areas.

ACTIVITY RATIO ^{90}Sr / ^{137}Cs

Since ^{137}Cs is rather easily determined in foodstuffs while the measurement of ^{90}Sr is more difficult and tedious, ^{137}Cs levels are often used to also describe the ^{90}Sr concentrations in foodstuff and its contribution to the ingestion dose after a large-scale fallout. This is only valid if the ratio of ^{90}Sr / ^{137}Cs is well established. In the following, therefore, the ratio of ^{90}Sr / ^{137}Cs shall be discussed in more detail.

In figure 1 it was already shown that this ratio may significantly vary among different fallout areas and climatic zones within a single country. Nevertheless, the variation in Austria with quite significant differences between the low plains and the mountainous Alpine region amounted to less than a factor of 10. This should make it possible to use an average ratio for ^{90}Sr / ^{137}Cs to estimate the contribution of ^{90}Sr to the dose if ^{137}Cs activity concentrations in milk are well established and the ratio of ^{90}Sr to ^{137}Cs is determined in a number of samples and low enough, say < 10 %, to cause only a small additional contribution to the dose.

However, the ratio ^{90}Sr / ^{137}Cs is not constant with time. The average ratio in milk in Austria since the above-ground weapons' testing until today is shown in figure 3. From an initial ratio of 0.2 during the time of nuclear weapons' tests fallout in 1960 - 1964 which corresponds to the activity ratio in air considering the different transfer factors, the ratio steadily increases after the end of above-ground testing to a value of 1.85 in 1985. The reason for this increase is the fact that cesium is more quickly fixed to the soil and, therefore, its bio-availability quicker decreasing than that of ^{90}Sr in the soil. After about 20 years the bio-availability of ^{90}Sr has increased by about 10-times relative to that of ^{137}Cs.

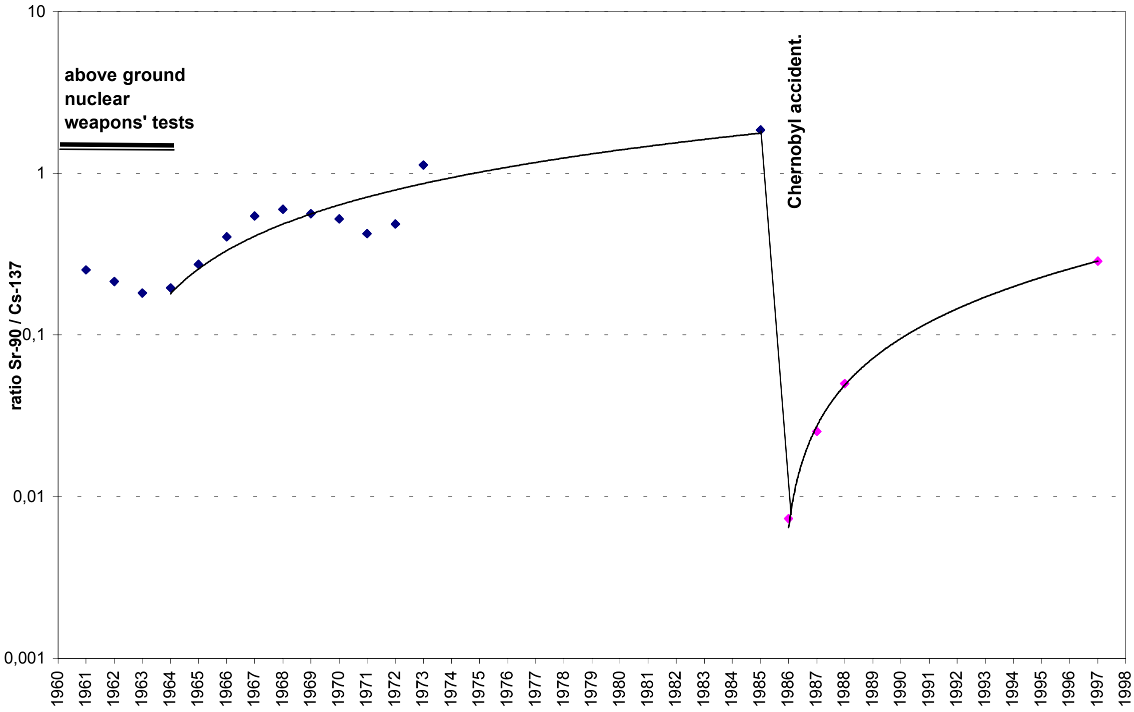

Figure 3 Average ratio of ^{90}Sr to ^{137}Cs in milk in the years 1960 - 1997

Immediately after the Chernobyl accident the ^{90}Sr / ^{137}Cs ratio drops to approximately 0.007. This is due to the low ratio of these two nuclides in the Chernobyl fallout which was only about 4 % at greater distances from the NPP (10). With an increasing relative contribution of the soil-to-plant-transfer to the activity concentration in grass and milk the ratio quickly increases again. This was already observed in the later part of 1986, but the increase becomes dominant in the years after. In 1987 the ratio has already increased by a factor of 3, in 1988 by a factor of 7. In 1997 a ratio of 0.29 is observed which is about 40-times the ratio of the period immediately after the Chernobyl fallout.

From the same reasons as discussed above, it is expected that the ratio ^{90}Sr / ^{137}Cs will further increase in the next years. It has already exceeded the ratio immediately after the end of weapons' testing and eventually will exceed those in 1985.

SUMMARY

The investigation showed that the ^{90}Sr activity concentration in milk has decreased to approximately 8 % on average of the peak values after the above-ground nuclear weapons' tests in 1964. This is equivalent to an effective half-life of roughly 13 years whereas in the first years after the nuclear weapons' fallout an effective half-life of about 2.7 years had been observed. Due to the low fraction of ^{90}Sr in the Chernobyl fallout the contribution of this nuclide from the Chernobyl fallout to the total ^{90}Sr level is comparatively low (about 25 % on average). Only in the first two years after the Chernobyl accident an increase in ^{90}Sr activity concentration in milk is observable. In the following years the concentration decreases at the same rate as before. Subtracting the Chernobyl ^{90}Sr fraction in the activity concentration of 1997, an effective half-life of 12.8 years is derived.

33 years after the fallout of the above-ground weapons' testing the exposure of the Austrian population by ^{90}Sr amounts to approximately 1,2 µSv (adult) and 0,9 µSv (5-year old child), respectively. This corresponds to only about 0,4 % of the ingestion dose by natural radionuclides in foodstuff. For the one-year-infant an exposure of about 2.5 µSv is estimated which is as negligible as the exposure of the 5-year-child or the adult. 75 % of this extremely low exposure is caused by ^{90}Sr from the nuclear weapons' test fallout and 25 % from the fallout after the reactor accident in Chernobyl on average.

The ratio of the activity concentration of ^{90}Sr to ^{137}Cs which is often used to estimate exposure levels by ^{90}Sr after a wide-scale contamination when only ^{137}Cs data, but no actual data on ^{90}Sr are available, shows a strong variation depending on the spatial distribution of the fallout zones. This is due to the fact that two major fallouts occurred with quite different ^{90}Sr/^{137}Cs ratios and significantly different fallout patterns. But even when the average activity ratio over the whole territory is considered, a significant variation with time is observed which ranges from an average ratio of about 0.2 at the time of the weapons' fallout to 1.8 just before the Chernobyl accident, and 0.007 immediately after the Chernobyl accident to about 0.3 only 11 years later. Ratios of these two radionuclides for dose estimates of ^{90}Sr from ^{137}Cs-activity measurements, therefore, have to be

carefully evaluated before applying them to estimate the dose contribution of ^{90}Sr to the total internal dose after a large-scale contamination.

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