(Aus dem Bereich Lebenswissenschaften des Österreichischen Forschungszentrum Seibersdorf Ges. m. b. H.)

Trends in caesium activity concentrations in milk from agricultural and semi-natural environments after nuclear fallout

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(With 3 figures)

Summary

The radiocaesium contamination of milk and milk products is directly related to that in grass or hay and therefore the time trend to the effective half-life in these fodders.

In the early phase the half-life in grass predominantly depends on effects such as dilution due to plant growth, translocation and weathering effects. The average effective half-life during this period (growing season) lies between 5 and 18 days. In upland pastures values of up to 25 days are observed. Studies performed on a great number of sites in particular countries after the Chernobyl accident showed half-lives for ^{137}Cs in grass from 7.9±1.5 d to 10.5±1.4 d for the period of May to July. An equivalent biological half-life for ^{131}I was observed. Only one measurement of half-lives in winter was performed up to now indicating a substantially longer value (50 d). No reliable data on effective half-lives at other periods of the year (late summer, fall) are available and would require further research.

The long-term decline is determined by soil properties. Soils with low fixation capacity and low pH show higher aggregated transfer factors into milk than others. In certain semi-natural alpine pastures these factors remain high for years which cannot be explained by extreme soil properties only. Other reasons such as water logging, little dilution due to low plant growth, cycling of radionuclides within living and dead plant biomass and runoff effects have to be considered as main causes. A classification system for the long-term trend in Cs-availability to milk is proposed, but further research on the differencies and possible measures with respect to seasonal variations and climatic conditions is required.

Key-words: fallout, contamination, biological half-life, semi-natural ecosystems, Caesium 137.

Zeitlicher Verlauf der Cäsiumaktivitätskonzentration in Milch in agrarischen und naturnahen Ökosystemen nach einem radioaktiven Fallout

Zusammenfassung

Die Kontamination von Milch und Milchprodukten durch Radiocäsium ist direkt abhängig von der in Gras oder Heu und damit der zeitliche Verlauf von der effektiven Halbwertszeit in diesen Futtermitteln.

In der Anfangsphase hängt die Halbwertszeit vor allem von Effekten wie der Verdünnung durch Pflanzenwachstum, Translokation und Abwitterungseffekten ab. Die durchschnittliche effektive Halbwertszeit liegt in diesem Zeitraum während der Wachstumsperiode bei 5–18 Tagen. In Almzonen werden Werte bis zu 25 Tagen beobachtet. Untersuchungen an einer Vielzahl von Standorten in einzelnen Ländern nach dem Tschernobylunfall zeigten Halbwertszeiten für ^{137}Cs in Gras von 7,9 ±1,5 d bis 10,5 ±1,4 d im Zeitraum Mai bis Juli. Für ^{131}J wurde eine vergleichbare biologische Halbwertszeit beobachtet. Nur eine einzige Bestimmung der Halbwertszeit im Winter wurde bisher durchgeführt, wobei sich ein substantiell längerer Wert ergab (50 d). Keine zuverlässigen Daten existieren für andere Zeiträume des Jahres (Spätsommer, Herbst). Weitere Untersuchungen in diesem Bereich sind erforderlich.

Das Langzeitabklingverhalten wird durch die Bodeneigenschaften bestimmt. Böden mit geringer Fixierungskapazität und niedrigen pH-Werten weisen einen höheren Cs-Transfer in Milch auf als andere. In bestimmten Almen bleiben die Transferfaktoren über einen Zeitraum von Jahren hoch, was nicht alleine durch extreme Bodeneigenschaften erklärbar ist. Andere Gründe wie Stauwasser, ein geringer Verdünnungseffekt durch Pflanzenwuchs, das Zyklieren des Radiocäsiums zwischen der lebenden und toten Pflanzenmasse sowie Einträge durch oberflächliche Verlagerung kontaminierter Bodenpartikel sind als die wesentlichen Gründe anzusehen. Ein Klassifikationssystem für das Langzeitverhalten der Cäsiumverfügbarkeit in Milch wird daher vorgeschlagen. Es ist jedoch weitere Forschung bezüglich der Unterschiede in der saisonalen Variation und den klimatischen Bedingungen erforderlich.

Schlüsselworte: Fallout, Kontamination, biologische Halbwertszeit, naturnahe Ökosysteme, Cäsium 137.

1. Introduction

After a nuclear fallout a large fraction of the exposure of the population is caused by the exposure due to consumption of contaminated foodstuff. An important fraction of foodstuff consumed by man is produced by feeding on grass: virtually all milk and milk products and a large fraction of meat (mutton and a major part of beef) as well as meat produced by feeding milk from these animals (lamb, veal). These foodstuffs comprise a significant contribution to the total ingestion dose after a large-scale fallout (Mück et al. 1992). Therefore, trends in activity concentration in grass play a significant role both in the exposure period immediately after fallout as well as in the long-term exposure by long-lived radionuclides (Mück 1989).

If the fallout of radioactivity occurred during the vegetation period, the radionuclide concentration of soluble radionuclides in milk will be directly related to that in grass. The grass contamination, on the other hand, will depend on the fallout level, the contribution by wet and dry deposition and the growth stage. The short-term time variation of the Cs-concentration in grass will be predominantly controlled by weathering effects and by dilution due to plant growth.

The long-term contamination, on the other hand, is a result of both the interception during the fallout period and the following translocation within the plant as well as the transfer from soil to plant. The decline in activity concentration in the plant is mainly caused by radionuclide export to deeper soil layers and export due to leave removal. As grass is grown under intensive and extensive cultivation, this may give rise to different decay behaviour characterized by different effective half-lives of caesium in the plant. In the long term, therefore, the main influence on contamination levels may be attributed to site-specific mobility and the availability of radiocaesium with regard to uptake by plant roots which is influenced by soil properties, water logging, climatic conditions and cultivation.

The present paper focusses on the state-of-the-art knowledge on short- and long-term "environmental decay" of radiocaesium in milk from agricultural and semi-natural environments.

2. Effective half-life of radionuclides in grass shortly after fallout

In the first year the ingestion dose is significantly influenced by the total deposition and the subsequent variation of radionuclide concentrations in plants. With a fallout during the growing season the rise in activity levels immediately after fallout is followed by a decline shortly afterwards which is predominantly caused by plant growth and weathering effects (Mück et al. 1994). It may be described by an exponential relationship (Nielsen 1981) characterized by one or more effective half-lives which will mainly depend on the rate of biomass growth. This in turn is dependent on the type of plant and, in particular, on the season of the year.

Up to the Chernobyl accident values for effective half-lives have been taken from observations after atomic bomb tests in arid areas of the USA (Martin 1964), from experiments with artificial contaminations (Milbourn and Taylor 1965) or from assumptions on plant growth, weathering and consequent dilution effects in the plant (Pröhl 1988). By using the Chernobyl fallout, the measurements were based on a realistic fallout scenario as to be expected after serious reactor accidents or nuclear weapons' detonations in contrast to artificial contaminations as commonly used in absence of real environmental contaminations (Mück et al. 1994).

In contrast to the fallout after the weapons' testing of the sixties, the deposition after the Chernobyl accident was short, lasting only for a few days. Thus, effective half-lives of ^{137}Cs observed after the Chernobyl accident are less influenced by subsequent radionuclide deposition than with the nuclear weapons' testing, for which the deposition lasted for several years due to continuous blasting and was continued even after the ban of atmospheric testing by the super powers by smaller nuclear weapons' countries. Thus, the evaluations after Chernobyl give more accurate results on the actual half-life in plants than those observed after weapons' testing.

Therefore, the determination of the effective decay constant after the Chernobyl fallout was of great interest. It was determined in Austria by observing the activity concentration in grass at various sites widely distributed over the country for a time period of three months after the accident (Mück et al. 1994). Such investigations were also performed in a number of other countries and a comparison of the various results shall be given.

2.1 Measurements performed in Austria

At nine different sites samples of grass, plant bases and litter were taken over a period of three months (OBERLÄNDER et al. 1986). The sites were chosen to cover the whole territory of Austria to give a representative value for the nation-wide grassland and to ensure that local variations due to different soil types or climatic conditions would not influence the overall result. The samples were taken starting on 6 May 1986, i.e. six days after the major fallout, and just at the end of the total fallout, until 21–23 July 1986, or 83 days after the major fallout. The interval of sampling was 2–4 days at the beginning and increased up to 45 days at the end of the survey period. At ten other sites, samples were taken only twice in the middle of the surveillance period to evaluate the nation-wide average activity concentration in grass and to compare the effective half-lives. They showed no significant deviations from the average of the nine sites.

Since "old" caesium from the nuclear weapons testing decayed only by a factor of about 1.7 since the fallout, it could distort the results obtained. However, average deposition of ^{137}Cs in Austria after the Chernobyl fallout was about four times higher than after nuclear weapons testing. Furthermore, the caesium of the weapons' fallout penetrated into deeper soil layers in the subsequent 20 years (MEISEL et al. 1991). The amount still to be found in the sod is negligibly small as compared to the Chernobyl fallout. From the measurement of samples taken before the accident it was estimated at less than 0.2 % (MÜCK et al. 1990 a).

2.2 Present state of knowledge

The variation of the ^{137}Cs-activity concentration in grass d.m. with time as observed after the Chernobyl accident is given in figure 1 for the Austrian sites.

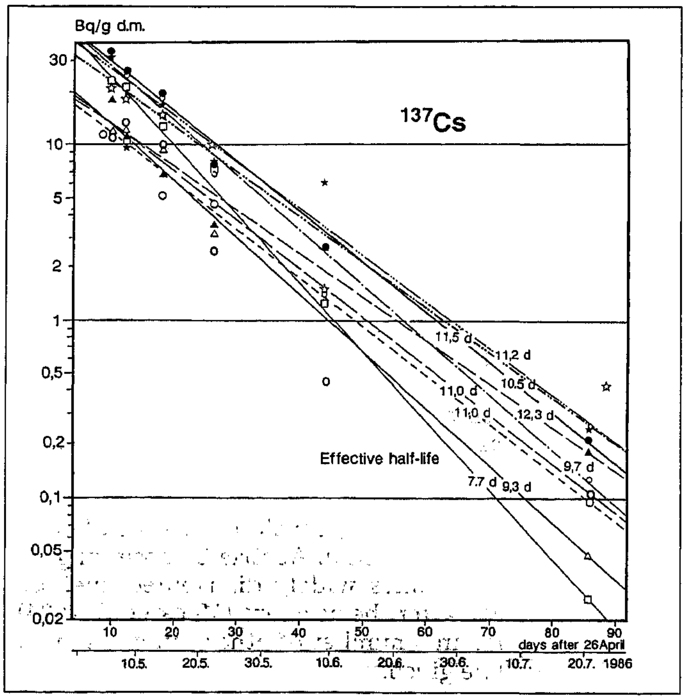

Fig. 1: Time trend in ^{137}Cs-activity concentration in grass (d.m.) at nine sites in Austria (MÜCK et al. 1994).

The decrease in concentration during the total period of 83 days is given by an effective half-life which ranges between 7.7 d and 12.3 d with an average value observed at the nine sites of 10.5±1.4 d.

Measurements of the effective half-life of ^{131}I were also performed at the same sites. They showed comparable results to those obtained for ^{137}Cs. The observed value of 4.6±0.6 d, equivalent to a biological half-life of 10.7±3.2 d, was, within the error limits, equal to that observed for ^{137}Cs.

Before the Chernobyl accident a number of studies was performed mostly using articificial contamination. A study in the UK after the Windscale accident showed an effective half-life both in field and laboratory experiments of 13.9 days (ARCRL 1960). MARTIN reported for ^{89}Sr and ^{90}Sr in plants a quite longer half-life of about 26 d and for ^{131}I of 13 d. The values were, however, observed in the desert of Nevada (MARTIN 1964). The growth rate in this arid region may not necessarily be transferable to European climate. MILBOURN and TAYLOR found a slightly shorter half-life of nine days by using ^{89}Sr as contaminant (MILBOURN and TAYLOR 1965). KIRCHMANN et al. (1966) reported a value of 30 d on pastures, while CHAMBERLAIN (1970) found a value of 14 d during the growing season. In a parallel study a co-worker and he found an effective half-life of 19 d during growing season, but 50 d in winter (CHADWICK and CHAMBERLAIN 1970). KRIEGER and BURMANN (1969) reported a half-life of 7–10 d, but demonstrated that "laboratory experiments" without realistic rainfall conditions resulted in values of 15–18 d emphasizing the fact that greenhouse measurements would tend to result in overestimates. From all these data NIELSEN recommended in a review paper (NIELSEN 1981) a half-life of 15 d for particulates and 10 d for iodine. According to the observations after the Chernobyl accident (MÜCK et al. 1994) such differences between the two radionuclides, at least in the growing season, don't seem realistic.

After the Chernobyl fallout numerous observations on effective half-lives using the Chernobyl fallout were performed. However, many experiments incorporated only one site which results in large variations in these values between different authors. ERIKSSON (1991) reported a half-life of 5.7 d at one site in Sweden while from the activity concentrations in grass at two sites in Northern Holland reported by DE MEIJER et al. (1990), although not specifically stated in the text, an effective half-life of 12–13 d may be derived. MARTIN et al. (1989) report a half-life of 22±2 d on a pasture in NE Scotland over the first 15 weeks after fallout. They suggest a shorter value of 15 d in the first three weeks after fallout. Table 1 indicates the large variations in observed half-lives.

It may seem doubtful whether experimental results for ^{89}Sr or ^{90}Sr may be readily transferred or compared to ^{137}Cs since ^{89}Sr is not as easily taken up into the leaf as ^{137}Cs and therefore may be more easily washed off at a later stage than Cs. This conclusion may be also drawn from experiments by KRIEGER and BURMANN (1969) who reported a half-life of 7–10 d for caesium, but 3–5 d for strontium. On the other hand, for iodine and caesium very similar values for the biological half-life were observed. In particular, when the same experimental conditions are used, almost identical weathering constants are derived. This is particularly demonstrated by different sites observed by MÜCK et al. (1994) in Austria. The explanation for this may be found in the rather quick uptake of both radionuclides by the plant which considerably reduces the weathering effects in a later phase.

The influence of cutting the grass on the activity concentrations is not clear. Most experiments concentrated on grassland which was undisturbed during

Table 1

Biological half-lives for iodine, strontium and caesium in grass

Biological half-life [d]	radio-nuclide	surveil-lance period [d]	author	time in the year	remarks
13.9	Cs, Sr	60	Agr.Res.Counc. (Arcrl 1960)	summer	sites in UK
13	I	40	Martin (1964)	summer	arid area in Nevada
26	Sr				
9	Sr		Milbourn and Taylor (1965)	summer	
8.3±1.3	I	16	Cline et al. (1965)	grow.season	7 plots near Hanford
30	Cs, Sr		Kirchmann et al. (1966)		
14			Chamberlain (1970)	grow.season	
19	Sr		Chadwick and	grow.season	
50			Chamberlain (1970)	winter	
7–10	Cs, Sr	75	Krieger and Burmann (1969)	May-July	pasture in Ohio
7.5±0.5	I	22	Heinemann and Vogt (1980)	May-Septemb.	sites near Jülich
8	I	14	Fulker (1987)	After Chernobyl	1 site at Sellafield
5–6	Cs				
21.9	Cs	90	Martin et al. (1988)	After Chern.	1 site in NE Scotland
19	I	50	Ishida et al. (1988)	After Chern.	1 site in Japan
14	I	50	Clooth and Aumann (1990)	After Chern.	single site
18	Cs				Bonn (FRG)
8.3±1.5	I	20	Spezzano and Giacomelli (1990)	After Chern.	pasture in NW Italy,
5.6	Cs				single site
9.9 (4–50)	I	–	Monte (1991)	After Chern.	15 sites in Northern
7.9 (2.3–17)	Cs				and Central Italy
22±2	Cs	98	Martin et al. (1989)	After Chern.	pasture, NE Scotland
12–13	Cs	70	De Meijer et al. (1990)	After Chern.	pasture in N. Holland
5.7	Cs		Eriksson (1991)	After Chern.	pasture in Sweden
10.5±1.4	Cs	83	Mück et al. (1994)	After Chern.	9 sites in Austria

the surveillance period. Furthermore, before the Chernobyl accident this question was virtually not addressed, probably because the amount of contaminated grass was not sufficient to answer this question. After the accident this question seems to be only addressed in our work (Mück et al. 1994). We found no change in the exponential decrease in activity concentration even during the period of grass cutting which in our opinion is to be expected if the major activity loss in grass is due to plant growth. From the decay curves observed by De Meijer, however, a stagnation after cut might be observed.

Also large discrepancies in the trend of the effective half-life curves were observed. While the majority of authors report an exponential decay by a single decay constant (Arcrl 1960, Martin 1964, Milbourn and Taylor 1965, Cline et al. 1965, Nielsen 1981, Clooth and Aumann 1990, Spezzano and Giacomelli 1990, Monte 1991, Mück et al. 1994), others suggest a decay by two half-lives (Oberländer et al. 1986, Fulker 1987, Martin et al. 1989), a shorter one immediately after fallout and a longer one in the subsequent 2–3 months. Also the retention observed by De Meijer (1990) suggests two decay constants. Results from some of the nine sites in Austria also suggest the existence of two effective half-lives. However, if we take the average over all nine sites only a single exponential decay curve is obtained. Possibly the derivation of two decay constants by some authors is a consequence of a too small number of sites investigated.

It should be emphasized that most of the half-lives investigated up to now were obtained during a period of intensive plant growth. In such a period dilution due to plant growth is dominant among activity reduction effects and, thus, these values might be only valid for this period of the year. As observed by KRIEGER and BURMANN (1969), a significantly longer biological half-life would be expected for autumn and winter.

2.3 Comparison to milk activity concentration

Applying an effective half-life of 10.5 days, a plant-animal transfer (ratio of activity concentration in milk to daily activity intake) of 0.004 $d \cdot kg^{-1}$ (HORAK et al. 1990) and a daily feeding of 60 $kg \cdot d^{-1}$ to the model proposed by COUGHTREY and THORNE (1983) for a single intake of fodder, we obtain after integrating the activity concentration in milk $U(t_a)$ at day t_a:

$$U(t_a) = 0.00551 \cdot A_0 \cdot \left(0.16 \cdot e^{-\ln 2 \cdot t_a} \cdot \left(-1 + e^{\ln 2 \cdot \frac{t_a}{1.13}} \right) + 19.34 \cdot e^{-\ln 2 \cdot \frac{t_a}{33.4}} \cdot \left(1 - e^{-\left(\ln 2 \cdot \frac{t_a}{15.3} \right)} \right) \right)$$

where A_0 ....activity concentration in fodder [$Bq \cdot kg^{-1}$] (1)

Averaged over the nine sites in Austria (fig. 1), an average ^{137}Cs-activity content in grass of 30.4±11.8 kBq/kg d.m. on 1 May is derived. With a dry matter content of 15 % in grass this is equivalent to 4.6 kBq/kg f.m. If we assume that by 2 May all cows were feeding on fresh grass (average activity concentration 4.3 kBq/kg f.m. on that date), we would obtain curve A in fig. 2 from this equation. This significant overestimation of the activity concentration in milk compared to the actually observed values may be explained by the fact that only a fraction of cows had been feeding on fresh grass and, additionally, the feeding had been delayed by the countermeasure to feed cows on old hay. Assuming a delay of feeding by nine days and a complete feeding of all cows on fresh grass by 11 May, curve B in figure 2 is obtained.

Both curves clearly indicate that a transfer factor of 0.007 $d \cdot kg^{-1}$ as reported earlier (NG et al. 1978) overestimates the activity concentration in milk in the conditions after the Chernobyl fallout. A value of 0.004 $d \cdot kg^{-1}$ fits much better although it still seems to overestimate. Many authors reported transfer factors of 0.002–0.004 $d \cdot kg^{-1}$ after the Chernobyl accident (STEINWENDER et al. 1988, VOIGT et al. 1989, WARD et al. 1989, TRACY et al. 1989) which are significantly lower than reported before the Chernobyl accident.

From equation 1 and from fig. 2 it is evident that the long-term decline in activity concentration is determined by the effective half-life of ^{137}Cs in the cow rather than that in the fodder. For $t_a > 40$ d, $U(t_a)$ is virtually totally determined by the last term in which 1-exp(-ln2-t_a/15.3) approaches 1 so that the term practically follows a decline according to a half-life of 33.4 d. This is also well observed in the measured values where the long-term decay follows a decline according to a half-life of 33 d.

However, the overall match of theoretical curve to observed values is not optimal. A possible explanation would be that the delay of feeding contaminated grass was shorter which would result in an earlier increase of activity

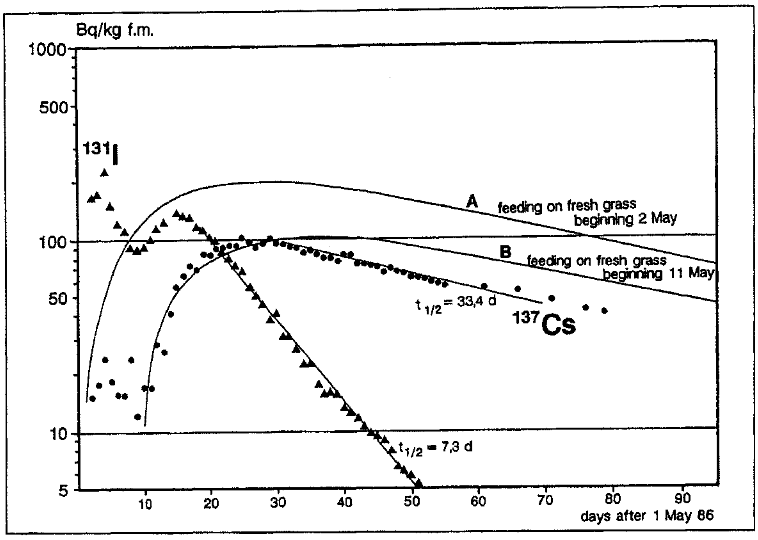

Fig. 2: Average activity concentrations of ^{131}I and ^{137}Cs in milk in Austria compared to theoretical curves calculated from grass activity concentrations.

concentration and subsequently in an earlier decline. In this case not only the first section of the curve would not match, but the transfer factor should be substantially lower than 0.004 $d \cdot kg^{-1}$ to fit the values.

2.4 Discussion of short-term behaviour

The short-term decline in ^{137}Cs-activity concentration in grass is given by an exponential decay with an effective half-life of approximately ten days in the period begin of May to end of July. Slightly shorter half-lives as observed in Nordic countries after the Chernobyl accident may be attributable to faster grass growth at an earlier growth stage after winter than in warmer regions. On the other hand, in areas with slow plant growth as in natural pastures longer half-lives are observed. Also values obtained in the desert of Nevada are considerably longer, but not well applicable to Central or Northern European climates. Comparably, values obtained for Central and Northern European countries may not readily be transferred to subtropical or tropical countries since the biomass growth may significantly differ from countries with temperate climate. Further research in these countries is required.

The effective half-life in the initial phase after fallout obviously depends strongly on plant growth and dilution effects caused by plant growth. As the biomass increase is significantly depending on the season of the year, effective half-lives derived for the period May to July may not be applied for other periods of the year. For these periods a determination of half-lives is required before application. Since the accuracy of modern predicition models (ECOSYS, FARMLAND) seriously depends on the thorough knowledge of effective half-lives over the whole year, further research in this field is urgently required to improve the prediction quality of such codes to be used in case of accidents.

The model of Coughtrey and Thorne (1983) seems to give best agreement between predicted values for ^{137}Cs-activity concentration in milk and those observed in grass if a transfer factor of 0.004 $d \cdot kg^{-1}$ and an effective half-life of 10.5 d is applied. The intermediate decline of activity concentration in milk is determined by the metabolism in the cow resulting in a decrease by an effective half-life of 7.3 d for ^{131}I and 33.4 d for ^{137}Cs, once the concentration in grass substantially decreased.

3. Long-term behaviour of radiocaesium on pastures and grassland

In order to be able to predict the long-term exposure of the population, a sufficient knowledge of the long-term retention of Cs-isotopes in relevant environments is essential and, in particular, their variation among the major food production areas is of great interest. Also the long-term retention in special habitats is of concern which may contribute little to the collective dose as their production size is usually small, but may cause substantial doses to exposed groups and should be known if proper countermeasures should be required. To assist in these efforts, the grasslands were investigated with regard to their long-term Cs-availability.

Milk may be chosen as a sensitive and qualified indicator of activity concentrations in grass in large scale radioactive contaminations for two reasons:

- The transfer fodder – milk is considered to be of low variability within a given fodder type (WARD et al. 1989).
- The measured milk activity values average over a large area if the milk is sampled from several cows. This significantly reduces the problem of appropriate sample taking which would be of importance with grass samples due to possible inhomogenities in deposition.

3.1 Method

One liter of milk averages over approximately 50 m² of intensively cultivated grass and 100–200 m² of extensively cultivated grassland (MÜCK et al. 1990b). If the milk sample is taken from a typical milk collection tour which covers the daily milk production of 50 to 300 cows, i.e. a milk tank of 1000–5000 l, an average over 5–25 ha of grassland is obtained.

As an example, the decrease in ^{137}Cs-activity concentration in grassland with time was observed in a series of different areas in Austria by observing the activity concentration in milk collected in these areas. They comprise typical grasslands in low-lying, intensively cultivated areas (Seibersdorf area, Leoben), pastures in more elevated, but still intensively cultivates areas (Salzburg town, Klagenfurt, Graz, Stainz) and in elevated, extensively cultivated areas (alpine pastures). All three area types are typical for grassland utilized for milk and meat production in Austria. The ^{137}Cs soil contamination was measured by means of in-situ gamma spectroscopy.

3.2 Observations in Austria

The average concentrations in milk in 1986 (peak value), in winter 1986/87, in 1988 and in 1993 as well as the ratios between the average concentration in 1988 and the peak-value 1986 as well as between the average concentration in 1988 and the peak-value 1986 are given in table 2 for these investigated areas.

Obviously, independantly from the total amount of fallout and the original contamination, the activity concentration decreases in different regions by different rates. While in some areas the decrease after the first two years amounted to 1–2 % of first year concentration levels, a second group of areas can be identified where the activity concentrations decreased to 4–8 % of first year levels. Besides these two, a third group is identifiable in which there is virtually no decrease in activity concentration observable in the first two years. In this third

Table 2

Decrease in ¹³⁷Cs-activity concentration in milk in Austria after the Chernobyl accident (Mück et al. 1990b)

Site	^{137}Cs-activity concentration in milk [Bq kg^{-1}]				ratio of activity concentration at various times after fallout	
	peak value 1986	winter average 1986/87	average value 1988	average value 1993	ratio 1988 / peak 1986	ratio 1993 / peak 1986
Seibersdorf area	37.0±5	13.0	0.1±0.07	0.03±0.01	0.003±0.002	0.001±0.0004
Salzburg town	233.0±40	51.8	4.9±1.7	1.16±0.67	0.021±0.008	0.005±0.003
Klagenfurt	92.5±23	45.1	4.1±5.5	0.62±0.22	0.044±0.024	0.035±0.024
Leoben	80.3±19	13.7±0.05	1.8±1.1	0.40±0.15	0.022±0.014	0.005±0.002
Graz	120.0±17	23.4	4.8±2.6	0.57±0.28	0.040±0.022	0.005±0.002
Stainz	172.0±27	44.4	14.1±3.7	2.47±1.60	0.082±0.025	0.014±0.010
alpine pasture 1	444±47	-	337±37	147±30	0.76±0.12	0.33±0.08
alpine pasture 2	196±56	-	279±106	148±70	1.4±0.7	0.76±0.42
alpine pasture 3	635±61	-	476±69	188±46	0.75±0.13	0.30±0.26

group the initial contamination levels were not higher than in other areas. Quite contrary, contamination levels in other areas (e.g. Salzburg town) were higher than in these habitats. Similar effects, but not as pronounced, were also observed in other countries (Wiechen 1991).

After seven years (summer 1993) the first group shows a decrease to activity levels of about 0.1–0.5 % of first year maximum levels, while the second group shows a decrease to 1.4–3.5 % of first-year values. The third group (alpine pastures) decreased only to about 30–70 %.

3.3 Classification of Cs-availability in different areas

The variation of the decrease of ^{137}Cs in milk and thus in caesium-availability in different areas suggests a classification approach to assist in adopting appropriate countermeasures if required. To this purpose the aggregated transfer coefficients for milk (T_{ag}) defined as the ratio of ^{137}Cs-activity concentration in milk in $Bq \cdot l^{-1}s$ to the total ^{137}Cs-deposition on soil in kBq/m^2 were calculated (table 3).

Generally, three categories of radiocaesium availability could be distinguished according to the T_{ag}-levels two years after deposition date:

- group I with T_{ag}-values of 0,04–0,16 $Bq \cdot m^2/l \cdot kBq$
- group II with T_{ag}-values 0,24–0,50 $Bq \cdot m^2/l \cdot kBq$
- group III with T_{ag}-values of >1,8 $Bq \cdot m^2/l \cdot kBq$

T_{ag}-values in groups I and II can be attributed to soil characteristics, neutral and acid soils, respectively. A clear break was observed between the second and third category. The third category, comprising alpine pastures, exhibited T_{ag}'s of an about ten times higher level, in one case about 100 times higher than those of group I or II.

In Austria, however, only a small fraction of milk (<1 %) is produced in these areas, of which only 5 % showed higher contamination levels. The contribution to the collective dose of the Austrian population thus is less than 0.1 % and therefore negligible (Mück et al. 1990b). The fraction of these pastures is further decreasing since there is a significant trend against milk production at high alpine pastures for economic reasons. More and more, only young animals before lactating age feed on high alpine pastures.

Table 3

Aggregated transfer coefficients (T_{ag}) for milk two years after deposition date

Site	Average ^{137}Cs-deposition [kBq m^{-2}]	^{137}Cs-activity concentration in milk [Bq l^{-1}] 1988	1993	Aggregated transfer coefficients T_{ag} [Bq l^{-1}/kBq m^{-2}] 1988	1993	soil characteristics
Stotzing	7.1±1.2	0.22	0.14±0.02	0.04±0.01	0.020±0.004	I:
Amstetten	26.4±20.3	1.5	0.27±0.04	0.06±0.04	0.010±0.008	neutral
Garsten/Linz	29.5±8.5	3.1	0.80±0.29	0.11±0.03	0.027±0.013	cambisols,
Ried	24.0±14	3.0	0.58±0.06	0.13±0.08	0.024±0.015	fluvisols
Innsbruck	12.0	1.5	–	0.13±0.09	–	(± neutral)
Enns	53.0	12.5	1.7±1.5	0.24	0.032±0.028	II:
Horn	3.5	0.85	0.24±0.03	0.24	0.069±0.009	dystric
Hartberg	18.0	4.9	0.41±0.18	0.27	0.023±0.010	cambisols,
Stainz	52.5±14.9	14.1±3.7	2.5±1.6	0.27±0.10	0.048±0.022	podzols
Stainach	54.5	13.8±4.2	2.1±1.6	0.25±0.08	0.039±0.013	(acid, sandy)
Gmünd	28.3±14.8	10.0	2.3±0.9	0.35±0.18	0.082±0.054	
Weitra	27.9±13.1	13.7	–	0.49±0.23	–	
alpine pastures in						III:
East-Tyrol	50	231±130	101±45	4.6±2.6	2.0±0.9	alpine dystric
Northern Styria	250	481	–	1.9	–	cambisols,
Southern Salzburg	27.1	476±69	193±44	17.6±2.6	7.1±1.6	podzols (acid, sandy)

4. Long-term behaviour of radiocaesium at an alpine pasture in Salzburg/Austria

As we have seen in 3.3, at certain sites the ^{137}Cs-bioavailability remained high after the Chernobyl fallout even years after deposition (Kirchner 1989, Kirchner et al. 1992). These alpine sites give rise to significant medium- to long-term ^{137}Cs-activity concentrations in milk as consequence of the distinctly higher mobility of radiocaesium in the ecosystem compared to agricultural sites. Figure 3 shows the time-dependancy of T_{ag}'s at an alpine site in Salzburg. Seven years after fallout the T_{ag} decreased only to 30 % of the value observed in 1986. Assuming only minor variations of the ^{137}Cs-fodder-milk transfer factor, an effective half-life for ^{137}Cs in plant biomass of 4.05 y can be deduced from the values given in figure 3.

This half-life value cannot be compared to the short-time half-life value as described in chapter 2 since the deposition occurred on the pasture when it was still mostly covered with snow. Thus, the activity was deposited by melting water in a later phase in contrast to the agricultural pastures. The activity decrease, therefore, even in the early phase, is not primarily caused by biomass growth. This is also demonstrated in figure 3 by the fact that the first year value fits very well the long-term decay while in agricultural ecosystems the first-year value after Chernobyl was substantially higher and would not fit the long-term decay (Mück 1994).

The half-life value observed is about two times higher than average long-term values observed in agricultural grasslands which range between 1.7–2.0 years (Mück 1994). Obviously, radiocaesium shows a completely different behaviour at some alpine sites with the effect of growth dilution and weathering being masked by other site specific factors.

A first study at this respective alpine pasture ("Naßfeld") in southern Salzburg, 1650 m above sea-level, focussed on mineralogical properties of the soils (Kirchner 1989). However, no clear relation was observed which could

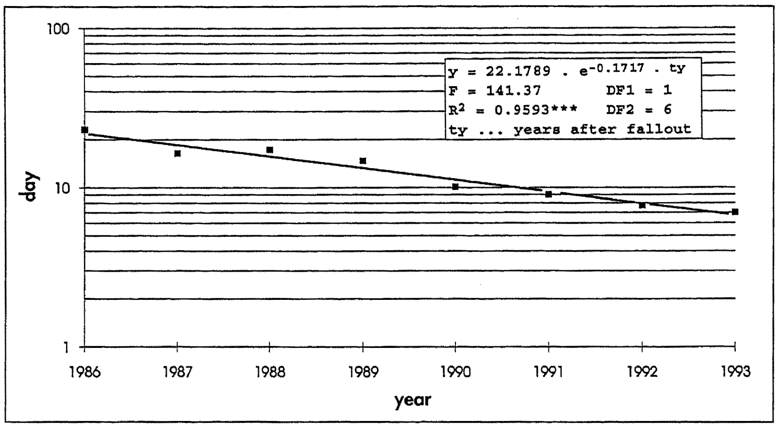

Fig. 3: Time-dependency of T_{ag}'s ($Bq \cdot l^{-1}$/ $kBq \cdot m^{-2}$) for radiocaesium into milk on an alpine pasture (Naßfeld) in Southern Salzburg.

explain the high Cs-availability. Thus, a second study tried to elucidate the effect of several parameters like soil properties, plant sociology and other site specific parameters (GERZABEK and MOHAMAD 1993, KIRCHNER et al. 1992).

4.1 Materials and methods

Four sites (I-IV) were chosen at the alpine pasture "Naßfeld" and, for comparison purposes, a fifth site (V) established in "Ursprung", in the lowlands near the city of Salzburg. Sites I to IV are characterized by stagnant water during snow melt in spring. Typically these alpine soils (I: Gleyi-Umbric Leptosol, II: Gleyic Cambisol, III: Epi-Gleyic Cambisol, IV: Gleyi-Dystric Fluvisol) have an organic horizon with high root densities and a loose layer of plant litter. Sampling was conducted in 1990. Site five was classified as Gleyic Cambisol (NESTROY 1993).

The pH-values of all five sites ranged from 3.8 (site II) to 5.5 (site III) without showing significant differences within the profile. In all cases the extractable potassium content was sufficient for pastures (>11 mg K_2O_{DL} / 100 g soil). The bulk density of the soil layers was 1.0 $g \cdot cm^{-3}$ or lower (mean: 0.91 $g \cdot cm^{-3}$). Soil organic matter contents of the first three cm were between 12.6 % (site V) and 44.7 % (site II).

The more frequent plant species were collected and the litter and sward mass determined. The results from this study are given elsewhere (KIRCHNER et al. 1992). Roots were isolated by carefully washing off adherent soil. Four 25×25 cm soil columns were taken from each site and subdivided into three layers (0–3 cm, 3–6 cm, 6–9 cm). Four different kind of samples were evaluated: soil with roots, soil without roots, shoots and roots. ^{137}Cs-activity concentrations were measured by gamma spectroscopy on a HPGe-detector. Soil samples additionally were extracted with 1 M NH_4-acetate and 0.1 M EDTA (pH = 10) to characterize the mobile Cs-fraction.

4.2 Observations at Naßfeld and Ursprung

^{137}Cs-concentrations in shoot tissue of plants of sites I–IV ranged from $3 \cdot 10^3$ to $9.1 \cdot 10^3$ Bq/kg (d.m.) and were 38 to 115 times higher than in Ursprung (V, table 4). At sites I to IV the second soil layer (3–6 cm) exhibited on average a 12 times lower Cs-concentration in the year 1990 than the first one. In 1988 the respective ratio was 11 (KIRCHNER et al. 1992). This may be explained either with low migration from the first to the second layer or with comparatively high leaching from the second layer. At site V, which – as the other sites – has not been disturbed by tillage after fallout, the respective value was 1.8. Due to the fact that extractability and, therefore, mobility of radiocaesium was lower at the latter site (table 4), differences can only be attributed to site specific migration rates (SCHIMMACK et al. 1989) during or shortly after fallout before equilibrium in soil. Table 5 shows the total amount of ^{137}Cs present in 1990. In Naßfeld the mean ^{137}Cs-contamination reached 22.8 kBq/m^2, which matches quite well the earlier estimate of 27.2 kBq/m^2 obtained by in-situ measurements (MÜCK et al. 1990b). At sites I to IV 12.6 to 24.6 % of the total ^{137}Cs were bound to living or dead plant material (roots, shoots and litter), which was at least a factor of 6 higher than in Ursprung (V).

Table 4

Total ^{137}Cs-concentrations in shoots and soil layers and extractable ^{137}Cs-fractions in soil samples from Naßfeld (I–IV) and Ursprung (V) in 1990 (KIRCHNER et al. 1992 and calculated from GERZABEK and MOHAMAD 1993)

Site	activity concentration [$kBq \cdot kg^{-1}$ d.m.] in				extractable (% of total)			
	shoot		soil		CH_3COONH_4		EDTA	
	mean	range	mean	range	mean	range	mean	range
I–IV shoot	4.6	3–9.1						
soil 0–3 cm			3.4	2.5–3.8	0.028	0.010–0.066	0.0085	0.0040–0.015
3–6 cm			0.28	0.25–0.32	0.042	0.009–0.120	0.0073	0.0019–0.019
V shoot	0.079							
soil 0–3 cm			0.66	–	0.004	–	0.001	–
3–6 cm			0.037	–	0.007	–	0.001	–

Table 5

Total ^{137}Cs-contamination levels in Naßfeld (I–IV) and Ursprung (V) (GERZABEK and MOHAMAD 1993, KIRCHNER et al. 1992)

Site	^{137}Cs-activity concentration [kBq/m^2] (% of total)				
	total	roots	soil with roots	shoots	plant bases[a] and/or litter
I	19.5 (100)	16.8 (86)	1.06 (5.4)	1.17 (6.0)	1.52 (7.8)
II	22.7 (100)	19.7 (87)	2.55 (11.2)	1.55 (6.8)	1.49 (6.6)
III	25.3 (100)	23.2 (92)	2.24 (8.9)	1.80 (7.1)	0.36 (1.4)
IV	23.8 (100)	21.3 (89)	0.50 (2.1)	1.86 (7.8)	0.64 (2.7)
V	13.0 (100)	13.0 (100)	0.27[b] (2.1)	0.003 (0.02)	–

[a] plant bases: bottom part of shoots [b] rough estimate, root mass not measured

Radiocaesium extractability was distinctly higher in "Naßfeld" than in "Ursprung" (table 4). Shoot/soil concentration ratios increased with an increasing exchangeable Cs-fraction at the Naßfeld sites

$y = 3.334 \cdot e^{26.981 \cdot x}$ (r = 0.92)

y = shoot/soil concentration ratio

x = % ^{137}Cs exchangeable (1 M NH_4-acetate)

Extrapolating the equation to site V and using the respective exchangeable Cs-fraction in soil a shoot-to-soil concentration ratio of 3.86 was derived. The measured average at site V was 0.165. Thus, it can be deduced that the quantity of mobile soil caesium explains only partly the shoot contamination of the alpine sites I–IV.

Generally herbs showed higher radiocaesium contents than gramineae (Kirchner et al. 1992). At site IV heather plants were highest in ^{137}Cs-content. These general findings correspond to the literatur (Jackson and Smith 1989, Colgan et al. 1990). However, the variations of ^{137}Cs-contaminations between plant species and the different plant species frequency distribution of sites I–IV and V could not explain the overall ^{137}Cs plant contamination level of the alpine sites. Root/shoot concentration ratios ranged from 0.99 to 1.51 at sites I to IV. At site V the respective values were 2.44 and 5.47 for herbs and gramineae.

Gramineae showed higher values in bottom parts of the shoots ("plant bases") than all other plant parts. Plant bases in general are known to have a high persistence of radiocaesium (Caput et al. 1990).

The following general differences between alpine and lowland permanent pastures could be observed:

- At alpine sites ^{137}Cs is mainly bound to the uppermost organic layer.
- Radiocaesium concentrations in shoot tissue were 38 to 115 times higher at the alpine sites, which can only partly be explained by total deposition, plant species distribution and the mobile ^{137}Cs-fraction in the soil.
- At the alpine sites 12.6 to 24.6 % of the total radiocaesium contamination was bound to living or dead plant biomass in 1990. At the lowland site the respective value was 2.1 %. The reasons for these findings are not known in detail, yet. However, some explanations may be suggested:

The main ^{137}Cs-deposition in Austria after the Chernobyl accident occured between 29th April and 1st May, during or shortly after snow melt at the alpine pasture (Naßfeld). Thus, stagnant water in the soil profiles may be assumed to result in a low migration velocity of radiocaesium which gave enough time for fixing of radiocaesium in plant tissue and litter. A low dilution effect due to a low growth rate of the plants and cycling of radiocaesium within living and dead plant biomass (Livens et al. 1991) may be further important reasons for the long effective half-life observed at this and some other alpine pastures. Above all, there is a chance for repeated contamination of plant shoots at Naßfeld due to the runoff of contaminated particles from the surrounding slopes (Maubert et al. 1990).

5. State of knowledge and requirement for further research

The effective half-life controlling the short-term decay of long-lived contaminants in plants and foodstuff seems to be well established in Central and Northern European countries. Data collected after the Chernobyl accident improved the knowledge considerably and are reliable enough to be used in prediction models on fallout consequences to result in sufficiently accurate predictions. However, these data apply only for the period of May to July which may be considered as the most critical period with regard to possible consequences after a serious fallout. For other periods in the year the effective half-life derived for this period may not be readily transferable. Further research is required to establish experimentally verified effective half-lives for any period in the year as the time of nuclear accidents or nuclear weapons detonations is unpredictable.

Effective half-lives derived for central and northern European countries may not be readily transferable to subtropical or tropical conditions. Further research in these countries on the comparitivity of the presented values is required if predicition models are used in accident mitigation.

Effective half-lives derived for intensively cultivated agricultural areas may differ significantly from those in less-intensively cultivated or semi-natural habitats. A classification scheme on categorisation of grassland was proposed and may prove a useful tool for governmental decision makers in mitigating the effects of large-scale nuclear fallouts. This classification scheme needs further input of observations in other countries to be fully applicable. Such observations, unfortunately, are rather sparsely up to now.

At certain semi-natural habitats, e.g. alpine pasture or highland sites, the ^{137}Cs-mobility in the ecosystem and thus the concentrations in milk remained much higher than in other areas, even for years. The reasons for this behaviour are not known in detail, possible influences are waterlogging, lack of dilution due to slow plant growth, cycling of radiocaesium within living and dead plant biomass, high, ammonium concentrations in soil as a result of water-logging which may increase ^{137}Cs-availability due to ion-exchange (HAUNOLD et al. 1987) and runoff effects. Although these agricultural areas usually are not of great importance to milk or food production of a country nor to the collective dose of the population, this question should be addressed in future research as it tends to remain a question of prolonged interest to the public because of the elevated values. Also it seems a matter of scientific interest to investigate the factors which lead to reduced Cs-binding features in these areas.

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(Manuskript eingelangt am 29. Juli 95, angenommen am 21. September 95)

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