APPLICATION OF AMMONIUMCYANOFERRATE IN REMEDIAL MEASURES IN AN AREA OF ELEVATED Cs-CONTAMINATION IN MILK

Konrad Mück[1], J. Zechner[2]

[1]Austrian Research Centre Seibersdorf
[2]Austrian Federal Ministery of Health and Consumer Protection

INTRODUCTION

Due to their relative long half-life and the high transfer factor into foodstuff Cs-isotopes may cause a substantial contribution to the exposure of the population after a fallout from a reactor accident or nuclear weapons' detonations. The reduction of Cs-concentration in foodstuff, therefore, is one of the major goals in proper countermeasures after such a contamination. Ammonium-Ferric-Hexacyano-Ferrate (AFCF), commonly referred to as "Giese-salt", is known to result in a substantial reduction of the Cs-activity concentration in milk and meat when applied with the fodder to the animal. The theoretically achievable reduction in activity concentration under experimental conditions and under scientific surveillance is known and amounts to about 80 - 90 % (1,2,3). Such experiments have been performed up to now on only a few individual animals. The actual reduction, however, which is achievable under realistic conditions is not well known. This reduction depends on such factors as acceptance of application by farmers, knowledge and ability to apply it in the prescribed way, but also on the variability between different animals, etc.

To investigate this question under realistic terms incorporating a large number of animals was enabled by the fact that ammoniumcyanoferrate was applied in an area of elevated ^{137}Cs-activity concentrations in milk in a part of the province of Lower Austria in the first half of 1988 (4). In contrast to the ideal situation of a research farm under controlled and optimized circumstances, the application was performed by several hundred farmers not acquainted to the application before. These had been only trained by members of the local agricultural associations and colleges before application and applied the salt according to their ability and understanding.

Since cyanides are allegedly carcinogenic, prior to application the AFCF was tested for possible long-term effects and in particular for carcinogeneses. The results of these tests shall also be discussed in short.

METHOD

The application of the Giese-salt was performed in an area of about 1000 km^2 involving several thousand cows and some 100 farmers. The region was chosen because of generally higher activity concentrations in milk than elsewhere although samples above EC intervention levels were very rare at the time of intervention. The application lasted for 6 months ensuring that equilibrium conditions were reached (4). The application started only 1.5 years after the Chernobyl event. The reason for this delay compared to ordinary practices after a nuclear fallout was that in the initial phase activity levels in that region were not significantly higher than elsewhere. But, because of the partially semi-natural habitat the long-term decrease was slightly slower than elsewhere in the country. Furthermore, after having realized the higher activity levels in that region, it took some time to get results on the epigenetic and carcinogenic tests providing the basis for approval of the application by the authorities. The application was performed by feeding 2 - 3 g of AFCF per cow each day. It lasted from 1 February to 10 July 1988.

The measurements of the activity concentration in the milk produced were not performed in the milk of individual cows in order to ensure optimum results. Rather the activity concentrations were determined in the milk powder produced in the milk drying plant of that region. By taking milk powder samples each sample averaged about 10000 - 30000 l of milk (5) since the milk is collected in milk collecting tours imploying tanks of about 3000 - 10000 l the contents of which are combined in the drying plant in even larger tanks. Thus, by taking powder samples a good averaging over large feeding areas and a great number of several 1000 individual cows was achieved.

The milk powder samples were measured in 1 l Marinelli-beakers on HPGe-detectors of 20 - 30 % relative efficiency which had been calibrated by intercomparison tests and milk powder standards supplied by the Austrian Research Centre Seibersdorf. Thereby, all measurements, even if they had been from different milk drying plants, are comparable. With a measurement time of 1000 s a detection limit of 5 Bq/kg and an average error of 5 - 7 % with individual samples of typical activity concentrations was achieved.

TESTS ON HARMLESSNESS OF APPLICATION

Ammonium-Ferric-Hexacyano-Ferrate is known to have an extremely low resorption in the body of animals (6). Also practically no transfer into body liquids and particularly milk occurs. Thus no effects in foodstuff should be observed. Despite this low resorption particular attention was paid to possible epigenetic - carcinogenic effects of the substance. Therefore, on behalf of the Austrian Federal Ministry of Health mutagenity tests were performed by the Austrian Research Centre Seibersdorf, Toxicology Departement. They comprised the reverse mutation assay with salmonella typhimurium (AMES) and the micronucleus test (7,8). The micronucleus test was performed on 15 male and 15 female mice in combination with a positive and a negative control group at 3000 mg/kg body weight (7). In the AMES assay AFCF was tested on five different salmonella breeds in two separate tests per breed and at 5 different concentration levels of AFCF with positive and negative controls (8).

AFCF did not show any mutagenity according to AMES nor in the Micronucleus test on NMRI-mice. For health reasons there were thus no obstacles for an application as an additive to fodder in a contaminated area for a limited period of time under restricted conditions (application of 2 - 3 g at most per animal per day)

RESULTS AND DISCUSSION

The weekly average activity concentration in milk powder is given in figure 1 as sum of ^{134}Cs and ^{137}Cs for the period August 1987 (before application) to August 1988 (after application). Obviously, from August to November 1987 the activity concentration decreases only slowly. This is expected due to fixation of caesium in soil and penetration into deeper layers. The effective half-life of this process which was determined for this site in another study over 7 years, amounted to 634 d (9) for ^{137}Cs. This is equivalent to a biological half-life of 672 d. Taking into account that in figure 1 the activity concentration is given as sum of ^{134}Cs and ^{137}Cs, the effective half-life for the sum activity of 540 d is displayed in figure 1. The activity concentration obviously follows this half-life very well during the period August to November.

In December an increase is observed which is mainly caused by feeding of hay which was harvested in summer 1987. The activity concentration, therefore, shows an average value comparable to the hay concentration of that period. This value was also predicted in a study by the Horak et al. (7) in which a large number of grass and hay samples of that region had been measured in summer 1987 and by deploying a transfer factor of 0,004 $d.kg^{-1}$ an average activity concentration of 25.5 Bq/l in milk had been predicted. This activity concentration which should last as long as this hay is fed to the cows (May 1988) is also displayed in fig.1.

With the start of the feeding of AFCF in February the activity concentration in milk rapidly drops to approximately 15 Bq/l followed by a further slow decrease to values around 7 - 9 Bq/l in May and June. The average activity concentration in that period is about 30 % of the value resulting from feeding the hay alone (without application of Giese-salt). Thus, the application of the caesium-binder reduced the activity concentration by about 70 %.

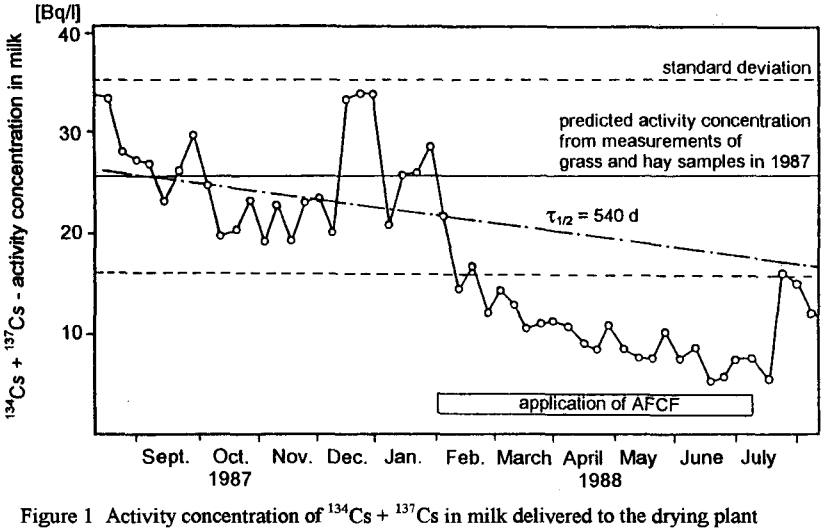

Figure 1 Activity concentration of ^{134}Cs + ^{137}Cs in milk delivered to the drying plant

At the end of the application period (10 July 1988), the activity concentration rises again to about 16 Bq/l. This value would be expected since without application of AFCF and after the winter hay feeding period the activity concentration in milk should be given by the broken-dotted line in fig.1 (resulting from the effective half-life of 540 d as described before). This is well demonstrated by the observed activity levels in figure 1 after mid-July.

Another proof of the reduction effect of activity concentration in milk is given in figure 2 which shows the activity concentration of $^{134}Cs + ^{137}Cs$ in milk in the four major drying plants in Austria in the period January to July 1988. These plants are situated in different provinces in Austria, separated by more than 80 km distance. During the very period all other drying plants show a rather constant value of the activity concentration with plant No.2 and 3 having average milk values of about 10 Bq/l and slowly decreasing to 8 Bq/l on average. Plant No.4 starts off with an average concentration of about 35 Bq/l and stays at this value till about May when the activity levels decrease to about 20 Bq/l. Only in plant No.1 where the AFCF had been applied to the cows, the decrease in activity concentration appears already in February with the beginning of AFCF feeding and stays at this level until June.

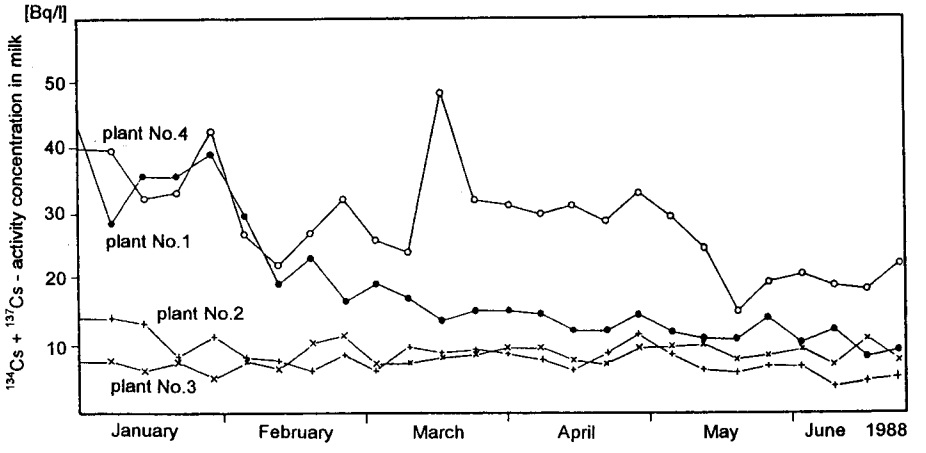

Figure 2 Activity concentration of $^{134}Cs + ^{137}Cs$ in milk in the four major drying plants

CONCLUSIONS

The application of Ammonium-ferric-cyano-ferrate (AFCF), also called "Giese-salt" with reference to its discoverer, may result in a substantial reduction of the activity concentration of ^{134}Cs and ^{137}Cs in milk, even if the drug is applied by farmers not familiar to its application before. Farmers who had been only quickly advised in its application and who had applied the drug for six months, achieved an average reduction in activity concentration by about 70 % during the period of application. This takes into account the variability of Cs-clearance in different individual animals as well as the varying ability of each farmer to cope with the problem.

The decrease in activity concentration is comparable to other efforts to reduce the activity concentration. A reduction by 30 - 80 % had been achieved after the Chernobyl accident in Austria for example by selection of low activity milk in the dairies. The application of AFCF is toxicologically safe and may be satisfactorily performed even by farmers not acquainted with ist application before. It, therefore, is a very valid tool in protecting the public in case of serious ^{137}Cs-contaminations after major nuclear accidents.

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