Acta vet. scand. 1964, *5*, 331—346.

From Norwegian Defence Research Establishment, Kjeller, Norway.

UPTAKE AND EXCRETION OF CESIUM 137, POTASSIUM AND ZIRCON/NIOBIUM 95 IN CATTLE

By

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It has been shown by earlier measurements made in Norway that the presence of Cs 137 in agricultural products is significant.

It was therefore considered of interest to perform gamma spectrographic measurements in order to study the uptake and excretion of Cs 137, K and Zr/Nb 95 in cattle.

The purpose of the study was to determine the ratio between Cs 137 intake and the amount which passes into milk. It was also of interest to determine which kinds of fodder give the highest contribution to the total daily intake of Cs 137. Potassium was also measured in order to find the ratio of K to Cs 137, and the content of Zr/Nb 95 was calculated because of the high concentration of these substances in the samples.

The study was intended in the first place to be a preliminary one, in that one particular cow with a fixed feeding roster was chosen. The experiment lasted for 6 weeks, commencing 22 January 1963. At the end of this period the cow was slaughtered and samples of different parenchymatous organs were taken and measured spectrographically for Cs 137, Zr/Nb 95 and I 131.

METHODS

Collection of samples

Recent routine measurements of I 131 and Cs 137 in thyroid and meat at the Norwegian Defence Research Establishment have shown that the Cs 137 content has been relatively high in the county of Trøndelag.

Enquiries were made at a number of farms in the vicinity of Trondheim. Retgjerdet Farm was found to offer the best facilities for our purpose, both with regard to regular sampling and despatching of the samples.

The choice of cow fell on Ylis 136 because it could be slaughtered at the end of the experimental period. It had the following data: Born 14. May 1959, last calved 4. December 1962, daily milk production 12 kg, breast measurement 178 cm and calculated live weight (Brinch-Lassen method) 490 kg.

The cow consumed the following fodder:

6	kg	hay
14	„	silage
3	„	processed cattle food (cattle food mixture C)
0.5	„	molasses
6	„	lyed straw*)
2	„	straw (2.—9. Feb.)
1	„	barley meal
55	„	water

All fodder ingredients were tested, in addition to milk, urine and fæces. The fodder ingredients were dried and pulverized at the State Agricultural Laboratory for Chemical Testing in Trondheim, whereafter they were immediately despatched to Kjeller. Samples were taken once a week for 6 weeks. The cow was slaughtered on 11. March 1963 and samples of the following parenchymatous organs were taken: Liver, spleen, kidneys, heart, brain, throat muscles, muscles of the foreleg, bone marrow, bone (metacarpus), thyroid and blood.

The samples were despatched from Trondheim in plastic containers holding 2.5 litres. Hay, lyed straw and silage were dehydrated and pulverised, and the percentage dry weight calculated.

Measurement of radioactivity

For the measurements a 50 channel gamma spectrograph with 4″ diam × 3″ NaI (TI) crystal as detector, was used.

As an initial test, portions of all samples were incinerated at 320°C for 24 hours and the ash put into the kind of receptacles used for fallout.

Using this method, it was found that the ash from only one-hundredth part of the contens of a 2½ litre container could be held in the fallout receptacle. Even though the measuring geometry is better when using a fallout receptacle, the count rate obtained is only one-fiftieth of that which would have been obtained had the contents of the 2½ litre container been measured directly.

Samples of hay, cattle food, silage, lyed straw, barley meal, molasses, urine, fæces, milk and blood were measured in plastic containers, the lids of which were made with a deep indentation, into which the gamma spectrograph crystal fitted exactly. The crystal was thus

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*) Straw steeped in an alkaline solution, then rinsed.

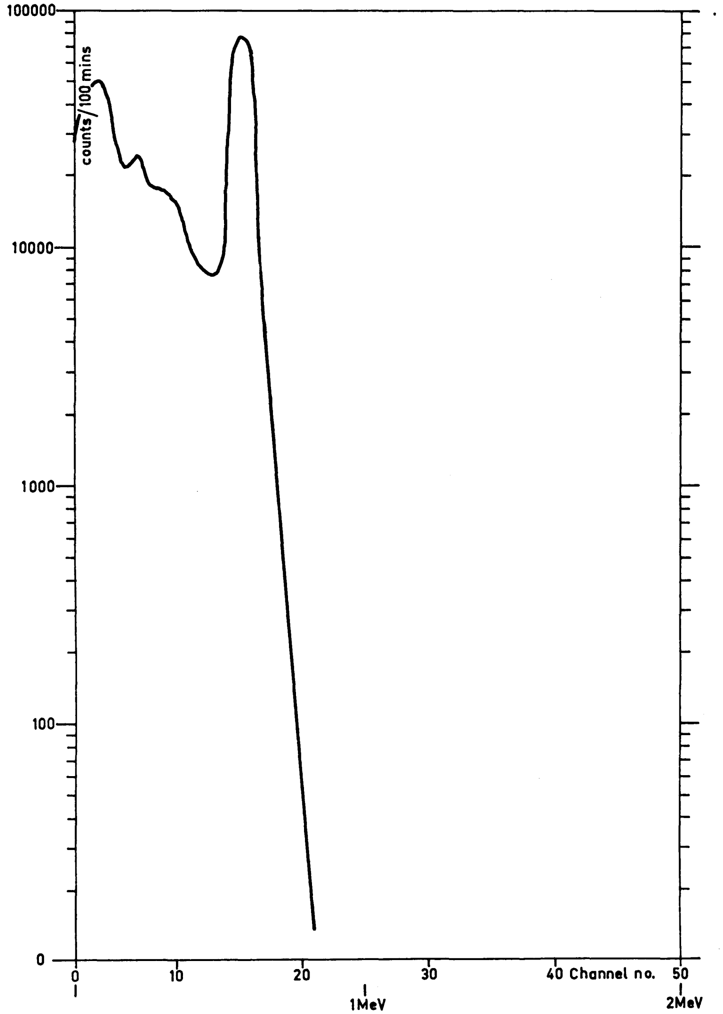

Fig. 1. Standard spectrum Cs-137.

"dipped" into the sample and a very good geometry for measuring purposes was obtained. From the fodder samples, a suitable quantity was extracted, depending on the sample's specific weight and consistence. Distilled water was added to each sample to bring its weight and self-absorption up to that of a container filled with distilled water only. The container of distilled water was used to calculate the background radiation. The samples were measured for 100 minutes, using the same procedure as for routine measurements of milk, described in detail by *Berg et al.* (1960).

For calibration purposes, known quantities of Cs 137, KCl and Zr/Nb 95 were used. These three substances were dissolved in 2200 g

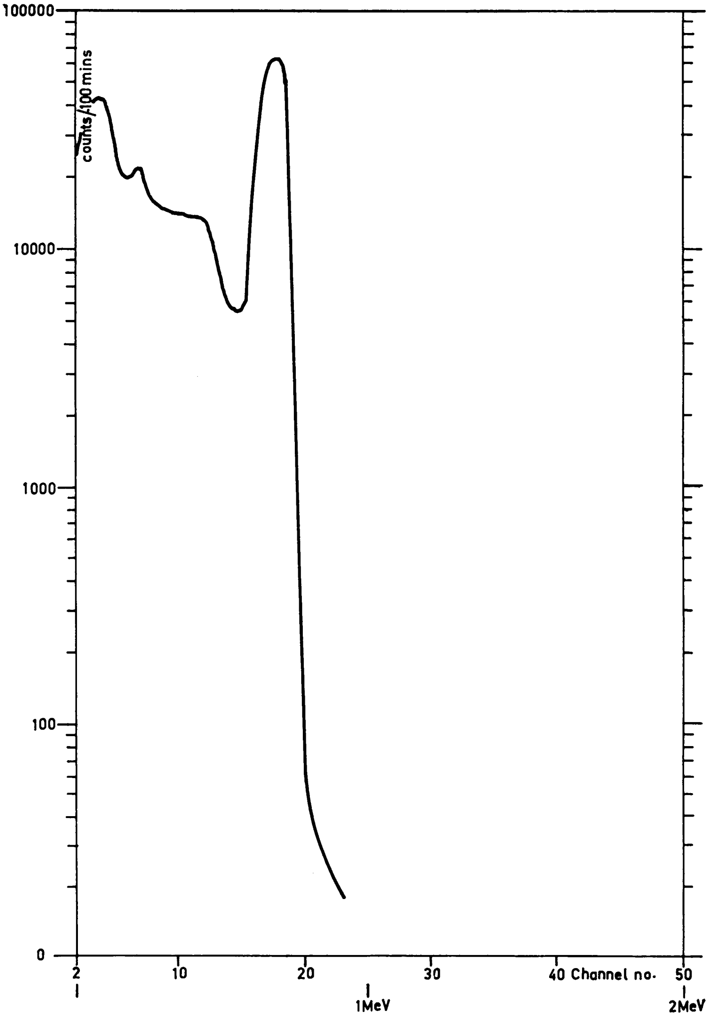

Fig. 2. Standard spectrum Zr/Nb 95.

distilled water in each their container, the same kind as used for sample measurement. The calibration curves for Cs 137, Zr/Nb 95 and K 40 are shown in figures 1—3.

The method of spectrum analysis is identical to that described by *Lillegraven* (1959 a, 1959 b) and *Thoresen* (1962), except that the spectra for Zr/Nb 95 and argon 41 must be deducted. Argon is present occasionally on account of sporadic leaks from the nuclear reactor at IFA.

Zr/Nb 95 has a photopeak in the region of 0.72—0.77 MeV, corresponding to channels 17, 18 and 19 on the gamma spectrograph used. Argon has a photopeak at 1.29 MeV, i.e. channels 29, 30, 31 and 32.

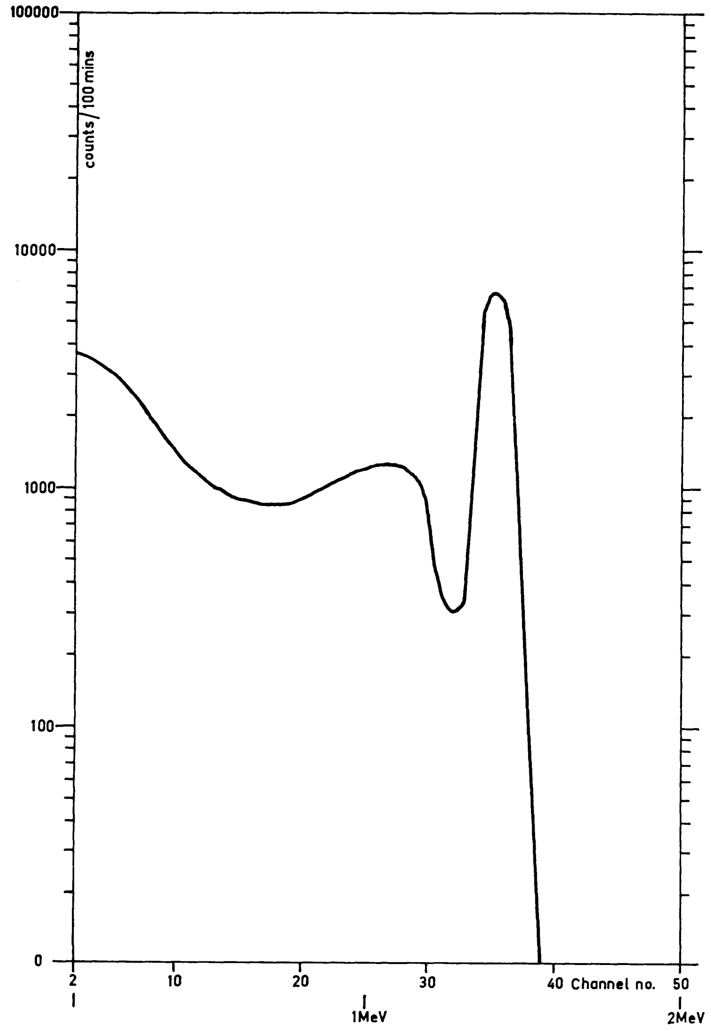

Fig. 3. Standard spectrum K-40.

The spectrum for argon is calculated from the background spectrum with and without argon. The calculations are made by the "strip-off" method, by which the contributions from the radioactive materials to the measured spectrum are deducted, one at a time, beginning with the one which gives the highest gamma energy. First of all, the background radiation is deducted, followed by K 40, then argon and thereafter Zr/Nb 95. The radioactivity of the materials is found by comparison with the standard spectra.

When measuring spectrographically muscle samples from foreleg, neck and heart, brain, liver, spleen, kidney, lung, bone marrow, bone and thyroid, the samples were placed in stainless steel containers, as described for measurement of meat by *Lillegraven* (1960). Also in this case standard spectra measurements for Cs 137, Zr/Nb 95 and K 40 were taken, using steel receptacles so that calculations of Cs 137, Zr/Nb 95 and K 40 could be included in the strip-off program.

Measurement of drinking water was carried out along the same lines as used previously (7). Drinking water samples were evaporated and total beta activity measured, as for fallout. Al six samples in the fallout receptacles were finally measured together in the gamma spectrograph.

RESULTS

The results and calculations therefrom are shown in tables 1—14. In addition to measuring errors, there are also considerable physiological uncertainties in connection with the number of tests taken and the dispersal of the radioisotopes within the cow and their excretion.

Firstly, it would have been better had the sampling been done daily instead of weekly. This is particularly true in the case of fæces, which is evident from the uneven results. The total quantities of fæces and urine per day were not measured, but were assessed from the amounts of fodder consumed and milk produced. Fæces may vary between 20 and 40 kg, and in the experiment was set at 30 kg per day. Urine may vary between 15 and 20 litres and was set at 18 litres per day.

By increasing the number of samples and collecting all fæces and urine, the total error could have been reduced considerably. The total error has been expressed as the sum of the measuring errors and those due to physiological uncertainties. When considering the individual results and calculations in this report, a total margin of error of ± 25 must be taken into account.

Table 1. Cesium 137, nC/kg fodder type.

Date of sampling	Hay	Silage	Lyed straw	Straw	Barley meal	Cattle food	Molasses	Drinking water
22/1 1963	2.10	0.45	0.20		0.40	0.25	0.22	0.002
29/1 1963	2.10	0.46		1.50	0.42	0.25	0.24	
7/2 1963	1.80	0.50		1.30	0.33	0.20	0.25	
12/2 1963	1.90	0.59	0.17		0.37	0.18	0.25	
21/2 1963	1.50	0.49	0.19		0.27	0.19	0.26	
26/2 1963	1.50	0.53	0.21		0.67	0.19	0.25	
Average	1.80	0.50	0.19	1.40	0.41	0.21	0.25	0.002

Table 2. Cesium 137, nC/kg milk, urine, fæces.

Date of sampling	Milk	Urine	Fæces
22/1 1963	0.05	0.13	0.30
29/1 1963	0.07	0.15	0.63
7/2 1963	0.06	0.11	0.52
12/2 1963	0.07	0.13	0.53
21/2 1963	0.07	0.10	0.48
26/2 1963	0.06	0.14	0.45
Average	0.06	0.13	0.49

Table 3. Zr/Nb 95, nC/kg fodder type.

Date of sampling	Hay	Silage	Lyed straw	Straw	Barley meal	Cattle food	Molasses
22/1 1963	3.10	1.60	10.90		2.90	0.34	0
29/1 1963	3.70	1.10		34.50	3.60	0.27	0
7/2 1963	2.70	1.10		31.00	2.80	0.16	0
12/2 1963	3.00	1.00	8.80		2.20	0.12	0
21/2 1963	1.90	1.00	6.00		2.70	0.17	0
26/2 1963	1.80	0.90	4.40		2.30	0.10	0
Average	2.70	1.10	7.50	32.70	2.70	0.19	0

Table 4. Zr/Nb 95, nC/kg milk, urine, fæces.

Date of sampling	Milk	Urine	Fæces
22/1 1963	0	0	3.90
29/1 1963	0	0	3.80
7/2 1963	0	0	2.80
12/2 1963	0	0	4.70
21/2 1963	0	0	7.90
26/2 1963	0	0	4.50
Average	0	0	4.60

Table 5. Potassium, g/kg fodder type.

Date of sampling	Hay	Silage	Lyed straw	Straw	Barley meal	Cattle food	Molasses
22/1 1963	20.6	2.3	0.9		5.7	11.5	16.4
29/1 1963	19.7	2.3		14.3	5.3	10.3	20.5
7/2 1963	19.2	2.7		14.0	4.8	13.2	22.0
12/2 1963	21.9	3.0	0.7		4.8	12.2	20.9
21/2 1963	10.3	2.8	1.1		4.1	12.7	22.2
26/2 1963	10.4	2.6	0.8		7.4	12.1	21.4
Average	17.0	2.6	0.9	14.2	5.4	12.0	20.6

Table 6. Potassium, g/kg milk, urine, fæces.

Date of sampling	Milk	Urine	Fæces
22/1 1963	1.5	7.7	1.3
29/1 1963	1.1	8.1	2.6
7/2 1963	1.3	7.6	1.6
12/2 1963	1.4	6.7	2.5
21/2 1963	1.7	3.7	1.3
26/2 1963	1.5	7.5	1.0
Average	1.4	6.9	1.7

Table 7. Intake of Cs 137 per day in nC.

Type of fodder	Mean concentration × daily fodder consumption	Intake per day 22/1-26/2	% of total Cs 137 intake per day	Intake per day 2/2-9/2	% of total Cs 137 intake per day
Hay	1.80 × 6	10.8	53.3	10.8	49.4
Silage	0.50 ×14	7.0	34.7	7.0	32.2
Lyed straw	0.19 × 6	1.14	5.7		
Straw	1.40 × 2			2.8	12.6
Barley meal	0.41 × 1	0.41	2.0	0.41	1.9
Cattle food	0.21 × 3	0.63	3.1	0.63	2.9
Molasses	0.25 × 0.5	0.13	0.6	0.13	0.5
Drinking water	0.002×55	0.11	0.6	0.11	0.5
Total		20	100	22	100

Table 8. Excretion of Cs 137 per day in nC.

	Mean concentration × daily excretion	nC Cs 137 excreted daily	% of total daily Cs 137 excretion
Milk	0.06×12	0.72	4.4
Urine	0.13×18	2.34	12.9
Fæces	0.49×30	14.7	82.7
Total		18	100

Table 9. Intake of Zr/Nb 95 per day in nC.

Type of fodder	Mean concentration × daily fodder consumption	Intake per day 22/1-26/2	% of total Zr/Nb 95 intake per day	Intake per day 2/2-9/2	% of total Zr/Nb 95 intake per day
Hay	2.7× 6	16.2	20.3	16.2	16.1
Silage	1.1×14	15.4	19.3	15.4	15.4
Lyed straw	7.5× 6	45.0	56.3		
Straw	32.7× 2			65.4	65.3
Barley meal	2.7× 1	2.7	3.4	2.7	2.7
Cattle food	0.2× 3	0.6	0.7	0.6	0.5
Molasses		0	0	0	0
Total		80	100	100	100

Table 10. Excretion of Zr/Nb 95 per day in nC.

	Mean concentration × daily excretion	nC Zr/Nb 95 excreted daily	% of total daily Zr/Nb 95 excretion
Milk		0	0
Urine		0	0
Fæces	4.60×30	138	100
Total		138	100

Table 11. Intake of K per day in gram.

Type of fodder	Mean concentration × daily fodder consumption	Intake 22/1-26/2	% of total K intake per day	Intake 2/2-9/2	% of total K intake per day
Hay	17.0×6	102.0	52.2	102.0	46.7
Silage	2.6×14	36.4	18.6	36.4	16.7
Cattle food	12.0× 3	36.0	18.4	36.0	16.5
Molasses	20.6×0.5	10.3	5.3	10.3	4.7
Lyed straw	0.9× 6	5.4	2.7		
Straw	14.2× 2			28.4	13.0
Barley meal	5.4× 1	5.4	2.7	5.4	2.4
Total		196	100	219	100

Table 12. Excretion of K per day in gram.

	Mean concentration × daily excretion	nC K excreted daily	% of total daily K excretion
Milk	1.4×12	16.8	8.8
Urine	6.9×18	124.2	64.7
Fæces	1.7×30	51.0	26.5
Total		192	100

Table 13. Results of tests of parenchymatous organs from cow Ylis 136.

Organ	Date of slaughtering	Date of measurement	I 131	Cs 137 nC/kg	Zr/Nb 95 nC/kg	K g/kg
Foreleg muscle	11/3 1963	20/3 1963	0	0.30	0.10	5.1
Neck muscle	„	20/3 1963	0	0.23	0.06	4.9
Heart muscle	„	22/3 1963	0	0.20	0.12	4.6
Kidney	„	18/3 1963	0	0.36	0.12	1.1
Brain	„	18/3 1963	0	0.27	0.12	5.9
Liver	„	19/3 1963	0	0.13	0.12	3.5
Lung	„	19/3 1963	0	0.18	0.70	3.0
Spleen	„	22/3 1963	0	0.11	0.09	4.5
Bone	„	22/3 1963	0	0.03	0.04	0.6
Bone marrow	„	19/3 1963	0	0	0.27	3.1
Thyroid	„	18/3 1963	0	0	0.41	1.9
Blood	„	14/3 1963	0	0.03	0	0.6

Table 14. Content of Cs 137 and Zr/Nb 95 in different types of fodder on the basis of crop per areal unit.

Type of fodder	Crop kg/areal unit	Daily consump. as areal unit m^2	Cs 137/m^2 nC	Zr/Nb 95/m^2 nC
Hay	830	7.2	1.5	2.3
Straw	200	10.0	0.3	6.5
Silage	2250	6.2	1.1	2.5

DISCUSSION

It will be seen from tables 1 and 7 that the highest concentration of Cs 137 was found in hay. The average value is 1.8 nC/kg, i. e. 53 % of the total daily intake of Cs 137. Silage gives 0.5 nC/kg, or 35 % of the total intake. Forraging with straw gives 13 %, as against 6 % for lyed straw. The concentration per kg was 1.4 nC for straw compared to 0.2 nC for lyed straw. Dry straw was fed for 7 days only (2.—9. February 1963). Taking the percentage dry weight of lyed straw, it is seen that approximately 0.5 nC, i. e. about 35 %, is removed per kg dry straw during the lyeing process.

Manufactured cattle food and barley meal give a considerably lower Cs 137 contribution compared to the fodder sorts already mentioned. Cattle food contributes 3 % and barley meal 2 % to the total intake. Molasses and drinking water are of little interest in the present connection, since they each contribute only 0.6 % to the total intake.

From table 8 it is seen that of the total Cs 137 excreted per day, 4 % is in milk, while the corresponding values for urine and fæces are 13 % and 83 %, respectively. The total daily intake of Cs 137 is approximately 20 nC, while the amount excreted is about 18 nC per day. The difference between intake and excreted quantity is in this case about 10 %.

Cs 137 in milk is calculated to 3.8 % of the total intake per day. Cs 137 in 1 litre milk represents 0.3 % of the total daily intake. This agrees well with the values obtained by *Ilin & Moskalev* (1957) and *Cragle* (1961).

When calculating the total intake and excretion, the amount of Cs 137 absorbed through the respiratory organs hat not been taken into consideration. Estimating that a cow inhales about 200 m^3 of air during 24 hours and that the Cs 137 content is about 0.0001 nC/m^3, this gives a contribution of only 0.02 nC per day, which is negligible in our calculations.

From table 13 it is seen that the average amount of Cs 137 in meat from the foreleg and neck is 0.26 nC/kg. This corresponds to 1.3 % of the daily intake per kg meat.

From table 2 it is seen that the ratio of Cs 137 in milk to that in urine is 1:2. Comparing this with the values for K given in table 6, a ratio of 1:5 is obtained, and thus it seems that Cs 137 does not follow the same route as potassium. Potassium tends to be excreted readily in urine (about 65 % of the total amount excreted) whilst only 13 % Cs 137 is excreted through urine. For fæces the figures are 83 % and 27 % of the total amounts excreted, for Cs 137 and K respectively.

In table 14 is shown the daily consumption of different kinds of fodder expressed in terms of crop per areal unit. The cow consumed hay corresponding to an area of 7.2 m^2, straw corresponding to about 10 m^2 and silage corresponding to 6.2 m^2. For these three sorts of fodder Cs 137 and Zr/Nb 95 per m^2 have been calculated. For hay and silage the amounts were 1.5 nC and 1.13 nC Cs 137 per m^2, respectively, which agrees well when seen in relation to the daily fodder consumption expressed in m^2. In the case of straw, an amount of 0.28 nC/m^2 was obtained, which is one-fifth of that found in hay. The reason for this can be explained as follows. Straw will be spread over a wider area per unit weight than hay (about 4:1). Further, straw has a less effective surface than hay. Hay is comprised of finer stalks and intermixed leaves, which makes it more liable to surface contamination from fallout than straw. Cs 137 in the various fodder discussed here must have originated from the nuclear bomb explosions in the summer of 1961. The hay was harvested during the period 16. July—17. August 1962 and was therefore not contaminated by fallout from the series of explosions which started on 5. August 1962. The silage was put into silos on 14. July 1962. The straw, on the other hand, was out in the fields much longer and was not gathered in until 28. September 1962. In addition to being exposed to the fallout from 1961, therefore, the straw was also exposed to the fallout which originated from the test series which recommenced on 5. August 1962. It will be realised, however, after consulting a fallout decay chart, that the latter test series has not, after 4—5 weeks, contributed to any great extent to increasing the amount of Cs 137 in straw. It must thus be assumed that the Cs 137 measured in straw was from fallout originating from the explosions in the autumn of 1961, and that the straw is therefore directly comparable with the hay and silage.

For Zr/Nb 95 the situation is somewhat different. In addition to the existing large quantities in fallout from the 1961 test series, large amounts were deposited after the renewed test series commenced on 5. August 1962. The fresh fallout had no significance for hay and silage, but straw was exposed for 5 weeks. Therefore, as seen from table 14, straw contains 6.5 nC/m^2, whilst hay contains 2.3 nC/m^2 and silage 2.5 nC/m^2. Whilst the Cs 137 ratio between hay and straw was 5:1, for Zr/Nb 95 it is found to be almost reversed, i. e. 1:3.

From table 3 it is seen that straw and lyed straw contain most Zr/Nb 95, 32.7 nC and 7.5 nC/kg, respectively. Hay and barley meal contain on the average high concentrations of Zr/Nb 95, namely 2.7 nC/kg. Silage contained 1.1 nC/kg, cattle food 0.2 nC/kg and molasses none. In terms of percentage of total Zr/Nb intake, straw, hay and silage are the most important contributors (see table 9). Table 3 also shows that the amount of Zr/Nb 95 decreased between the date of sampling and the end of the experimental period, on an average 35 %. This agrees well with the half-life of Zr/Nb 95.

Zr/Nb 95 could not be traced in milk and urine. Measurements of fæces showed wide variations. Had the samples been taken more often, the results should have shown a decreasing tendency from the date of the first sampling to the end of the experiment.

From table 13 it is seen that only the lungs contained significant quantities of Zr/Nb 95, i. e. 0.7 nC/kg. This is entirely due to inhalation of the isotopes in question. The daily intake of Zr/Nb 95 in the period 2.—9. February was 100 nC, while in the remainder of the experimental period, when lyed straw was used, it was 80 nC per day. Zr/Nb 95 passed out with fæces was calculated as 138 nC per day. In this connection reference is made to the uncertainties already mentioned. It follows that Zr/Nb 95 it not absorbed into the organism, but merely passes through the digestive canals. Zr/Nb 95 has therefore not the same biological interest as Cs 137. During its passage through the digestive canals, which will normally only take 2—3 days, Zr/Nb 95 irradiate the intestinal walls locally. During this period, Zr/Nb 95 will lose only about 3 % of its radioactivity. It is not known whether this local irradiation has any pathological significance, but large quantities daily for a long period could represent a certain hazard.

Measurements of parenchymatous organs did not reveal any ostensible quantities of I 131. Neither was this expected in view of its short halflife.

CONCLUSIONS

Measurements of various kinds of fodder show that hay has the highest concentration of Cs 137, followed by straw. Expressed as percentage of total intake per day, hay contributes over 50 %, silage 35 %, followed by straw and lyed straw. Calculations of the total Cs 137 intake and excretion show that the quantity consumed and the quantity passed out are the same. Of the total daily intake of Cs 137, 3.8 % passes into milk. This corresponds to 0.3 % per liter milk. Meat from foreleg and neck contained an average of 0.26 nC Cs 137 per kg. This is equivalent to 1.3 % of the daily intake per kg meat.

The results show that Cs 137 does not follow the same route as potassium.

A comparison has been made between the daily consumption of hay, straw and silage on the basis of crop per areal unit. The daily consumption of hay was equivalent to 7.2 m^2, of straw 10 m^2 and of silage 6.2 m^2. Per m^2 the amount of Cs 137 in hay was 1.5 nC, in straw 0.3 nC and in silage 1.1 nC. The Cs 137 in these three sorts of fodder originated from the nuclear test explosions in the autumn of 1961. Per m^2 the amount of Zr/Nb 95 in hay was found to be 2.3 nC, in straw 6.5 nC and in silage 2.5 nC.

The relatively higher concentration of Zr/Nb 95 in straw compared to Cs 137 was by reason of the last test explosions which commenced 5. August 1962. Measured quantities of Zr/Nb 95 showed a falling tendency between the beginning of the experimental period and the end, on an average 35 %. This is in good agreement with the half-life of these isotopes. Straw and lyed straw contained most Zr/Nb 95, i. e. 32.7 nC/kg and 7.5 nC/kg, respectively. Hay and barley meal contained 2.7 nC/kg each, silage 1.1 nC/kg, and cattle food 0.2 nC/kg. Molasses contained negligible amounts.

The lungs were the only parenchymatous organs which contained significant amounts of Zr/Nb 95, i. e. 0.7 nC/kg. This was due to inhalation of these isotopes. The total intake of Zr/Nb 95 during the period 2.—9. February was 100 nC/day. When lyed straw was used, it was 80 nC/day. The total excretion of Zr/Nb 95 was calculated to 138 nC/day. Zr/Nb 95 does not pass into the organism, and will merely pass through the digestive canals.

I 131 could not be traced in the parenchymatous organs.

Acknowledgements

This study has been undertaken in cooperation with and under the leadership of the following: Norwegian Defence Research Establishment, Mr. T. Hvinden; Norwegian Army Veterinary Corps, Lt. Col. P. Gjestvang; Institute for Food Control and Hygiene, Norwegian Veterinary College, Prof. S. Hauge.

The authors wish to express their thanks to the following in particular: Mr. Ivar Jullumstrø, Reitgjerdet Farm, for kind assistance during the test; Mr. Lars Beseth, Town Veterinary Laboratory, Trondheim, for taking and despatching samples; Adolf

Moen, Agricultural Chemical Testing Station, Trondheim, for undertaking dehydration and pulverization of the fodder; Mr. Per Torboe for the statistical processing.

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SUMMARY

Gammaspectrographic examinations have been carried out of uptake and excretion of Cs 137, K and Zr/Nb 95 in a dairy cow.

At the end of the experiment the cow was slaughtered and measurements of parenchymatous organs were made.

Cs 137 and Zr/Nb 95 in hay, straw and silage are discussed in connection with crop per areal unit.

ZUSAMMENFASSUNG

Aufnahme und Ausscheidung von Cesium 137, Kalium und Zirconium/ Niobium 95 im Rind.

Bei einer laktierenden Kuh sind gammaspektrographische Untersuchungen über Aufnahme und Ausscheidung von Cs 137, K und Zr/Nb 95 gemacht worden.

Am Ende des Versuches wurde die Kuh geschlachtet, und Aktivitätsmessungen der parenchymatösen Organe vorgenommen.

Cs 137 und Zr/Nb 95 in Heu, Stroh und Silage wurden mit Hinblick auf den Ertrag pro Flächeneinheit diskutiert.

SAMMENDRAG

Opptak og utskillelse av Cesium 137, Kalium og Zircon/Niobium 95 hos storfe.

Det har blitt foretatt gammaspektrografiske undersøkelser over opptak og utskillelse av Cs 137, K og Zr/Nb 95 hos en enkelt ku.

Ved forsøkets slutt ble kua slaktet, og det ble foretatt målinger av parenchymatøse organer.

Cs 137 og Zr/Nb 95 i høy, halm og silo er diskutert i forbindelse med avling pr. arealenhet.

(Received December 26. 1963).