

Zeolite and bentonite as caesium binders in reindeer feed

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Abstract: The effects of zeolite and bentonite on the accumulation and excretion of radiocaesium (Cs-137) in reindeer were studied in two feeding experiments. Six animals in each experiment were given lichens contaminated with radiocaesium from fallout after the Chernobyl nuclear power plant accident. In addition, they were fed pellets containing bentonite (Experiment I) or zeolite (Experiment II). Two animals, controls, in each experiment received no caesium-binder. The activity concentration of radiocaesium in blood was used to evaluate the radiocaesium level in the body. Faeces and urine were collected to measure the excretion of radiocaesium.

The animals in Experiment I were depleted of radiocaesium before the start of the experiment. After three weeks, with an intake of 17 - 18 kBq Cs-137/day, the controls had reached activity concentrations of radiocaesium in blood corresponding to 4 - 4.5 kBq Cs-137/kg in muscle. Reindeer fed 23 or 46 g of bentonite per day stabilized at values below 0.8 kBq/kg in muscle. In Experiment II, the reindeer started with radiocaesium activity concentrations in blood corresponding to 2 - 4.5 kBq Cs-137/kg in muscle. After four weeks of feeding, with an intake at about 8.5 kBq Cs-137/day, controls had increased their radiocaesium values by an average of 40%. Reindeer receiving 25 or 50 g zeolite per day decreased with 18 and 45%, respectively. Net absorption of radiocaesium from the gastro-intestinal tract was calculated at 50 - 70% in animals receiving no caesium-binder. Reindeer fed bentonite had an absorption below 10% while those fed zeolite absorbed around 35%.

Key words: radiocaesium, radioactivity, fallout, Chernobyl

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Introduction

In large parts of the reindeer herding area of Scandinavia, lichens - the main winter feed for reindeer - are highly contaminated with radiocaesium (Cs-134 and Cs-137) from the Chernobyl fall-out (Figure 1). In contaminated areas reindeer have a high intake of radiocaesium

from September to April. Radiocaesium is easily absorbed from the gastro-intestinal tract and accumulates in soft tissues, resulting in high activity concentrations of radiocaesium in the muscles. Reindeer meat that is sold for human consumption in Sweden is not allowed to contain more than 1500 Bq Cs-137/kg. In many

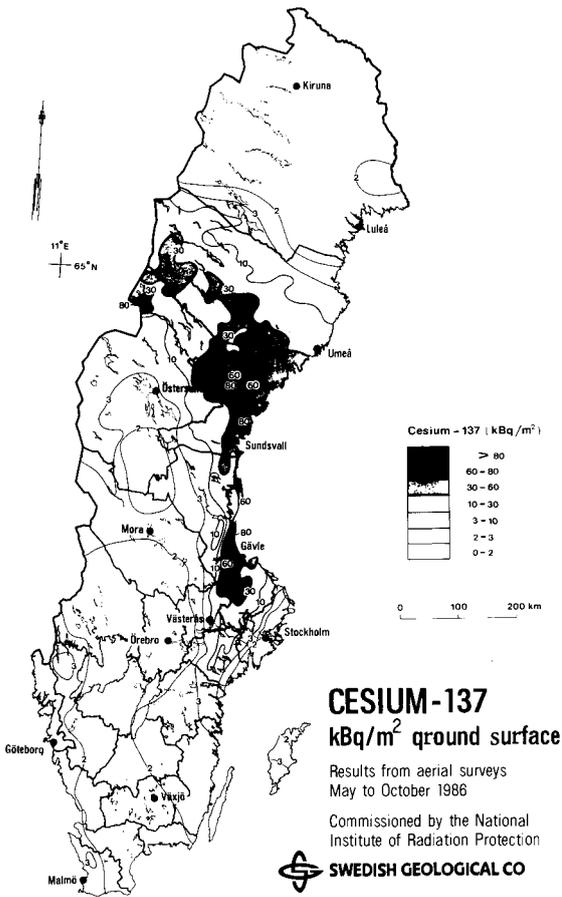


Figure 1. Ground cover of Cs-137, kBq/m² over Sweden. The amount of Cs-137 per m² corresponds roughly to the amount of Cs-137 per kg of dry lichens.

areas reindeer meat can reach values 10 to 30 times this during the winter.

The biological half-life of radiocaesium in reindeer is relatively short. With no radiocaesium intake the body content of radiocaesium is halved in 1 - 4 weeks (Holleman *et al.* 1971, Blix 1988, Åhman 1988b). A high intake of potassium has been shown to increase excretion of radiocaesium in sheep (Mraz 1959) and reindeer (Hove *et al.* 1987).

Holleman *et al.* 1971 found the shortest half-lives (6.7 days) in June - July when metabolic rate was high and potassium intake was about 10 times higher than in the winter.

During the last two winters feeding has been

used in Swedish reindeer management as a means to reduce radiocaesium in reindeer before slaughter. For practical and economical reasons, and also not to risk the health and well-being of the animals, it is important to keep the time of feeding as short as possible. One way of doing this is to add a caesium-binder to the feed given to the reindeer. When feeding reindeer outdoors, in corrals, there will always be some vegetation available and there is also the possibility of soil-eating. An efficient caesium-binder can prevent most of the ingested radiocaesium from being absorbed. Radiocaesium from the body pool secreted into the gastrointestinal tract may also be restricted from reabsorption.

Since the 1960's caesium-binders have been used in animal studies. Hexacyanoferrate (Giese-salt, Prussian blue) were used as caesium-binder to rats by Nigrović (1965) and to rat, pig and cattle by Giese (1971). In Norway hexacyanoferrate has been given to reindeer with good results (Blix 1988, Hove *et al.* 1988). Hexacyanoferrate is not, however, allowed in animal feed by the Swedish authorities. Van den Hoek (1976 and 1980) used bentonite, a clay consisting mainly of montmorillonite, as caesium-binder to sheep and cattle. With 10% of bentonite in the concentrate ration, absorption of radiocaesium from the gastro-intestinal tract was less than 5%. Pellets containing bentonite (2.5%) have also been used in practical reindeer feeding in Sweden during the winter of 1987/88. Bentonite, however, increases water consumption considerably (Åhman 1988a), which is a serious draw-back in reindeer feeding. More recently zeolites, a group of tectosilicate minerals, have been used in animal feed. Of the zeolites, chabazite, mordenite and also clinoptilolite have high affinity to caesium. Mordenite has been shown to accelerate radiocaesium excretion in sheep and goats (Forberg *et al.* 1988). Clinoptilolite significantly reduced absorption of radiocaesium in sheep (Philippo *et al.* 1988). Zeolites have also been used as dietary supplements to improve health and pro-

ductivity in many domestic animals (Mumpton & Fishman 1977).

The aim of this study was to investigate the effect of bentonite and zeolite on radiocaesium absorption and excretion in reindeer fed lichen contaminated with radiocaesium from the Chernobyl fallout.

Material and methods

One feeding experiment with 6 male reindeer calves was performed during January and February 1987 (Experiment I) and another during April and May 1988 (Experiment II). The animals were fed lichens contaminated with radiocaesium (30 - 45 kBq Cs-137/kg DM) and pellets with bentonite, Experiment I, or zeolite (mordenite), Experiment II. In each experiment two reindeer were fed the same pellets without any caseium-binder added. Both experiments were made as parts of more comprehensive studies (Åhman 1988b and Åhman 1988c).

The animals were kept individually in out-

door pens. They were fed twice a day. Daily food and water consumption was registered and the animals were weighed once a week.

Feed consumption and intake of Cs-137 in the two experiments are shown in Tables 1 and 2. Daily radiocaesium intake varied somewhat depending on varying radiocaesium content and ray matter in the lichens. Samples of lichens were taken continuously for the analyses of dry matter and radiocaesium content and the average daily amount of radiocaesium provided to the reindeer was calculated.

In Experiment I the animals were given 17 - 20 kBq of C-137 daily. During the first three weeks two animals (Group 1) got no bentonite, two (Group 2) got 23 g of bentonite/day and two (Group 3) got 46 g/day. During the following two weeks all animals got 27 g of bentonite/day.

In Experiment II the amount of Cs-137 given to the reindeer was 8 - 9 kBq/day during the first four weeks, followed by 1 kBq/day during 10 days. Two reindeer (Group 1) got no zeolite,

Table 1. Daily consumption of lichens and pellets in Experiment I. Intake of Cs-137 and bentonite per day.

Period (1987)	Lichens (DM) per day	Cs-137 per day	Pellets (91% DM) per day	Supplements of bentonite per day		
				Group 1	Group 2	Group 3
Jan. 16-29	500 g	17-20 kBq	750 g	—	23 g	46 g
Jan. 30 - Feb. 6	500 g	17-20 kBq	850 g	—	23 g	46 g
Feb. 7-20	500 g	17-20 kBq	900 g	27 g	27 g	27 g

Table 2. Daily consumption of lichens, hay and pellets in Experiment II. Intake of Cs-137 and zeolite per day.

Period (1988)	Lichens (DM) per day	Hay (87% DM) per day	Cs-137 per day	Pellets (91% DM) per day	Supplements of zeolite per day		
					Group 1	Group 2	Group 3
April 22-							
May 19	280 g	110 g	8.9 kBq	980 g	—	25 g	50 g
May 20-29	35 g	180 g	1 kBq	1410 g	—	25 g	50 g

two (Group 2) got 25 g/day and two (Group 3) got 50g of zeolite/day during both periods.

Blood samples were taken in order to monitor radiocaesium content of the body. Faeces and urine were collected for determination of the daily excretion of Cs-137. In Experiment I, collection was made on four occasions, each time during two days from one of the reindeer in each of the three groups and during the following two days from the other reindeer in each group. During collection the reindeer were kept indoors in cages. Faeces were collected in a bag connected to a harness put on the animal. Urine was collected in a vessel beneath the cage.

In Experiment II collection of faeces was made during two or three days on two occasions. Unfortunately the reindeer had begun to grow large antlers which made the cages too small for them. However, faeces could be collected with the reindeer in an indoor pen. Urine could only be collected from three animals during the first collection period and from one during the second. During the period with low radiocaesium intake collection of faeces was made from three animals during one day. From one animal also urine was collected.

In Experiment I Cs-137 + 134 in whole blood and in urine (20 ml samples) was measured in a well scintillation (NaI) detector at the Department of Radioecology, Swedish University of Agricultural Sciences. The amount of Cs-137 was calculated as total amount of Cs-134 and Cs-137 times a factor 0.69. Cs-137 in lichens, hay, and faeces was measured by means of Ge hyperpure detectors at the same department. In Experiment II the corresponding samples were measured either with a NaI(Tl) or a Ge detector at the Department of Nuclear Chemistry, The Royal Institute of Technology, Stockholm.

Results

The reindeer in Experiment I maintained their

body weight, 49 kg on average, throughout the experiment. In Experiment II, body weights increased by 8 kg, from a mean of 44 to 52 kg, during the experiment.

Animals given bentonite in Experiment I had higher water consumption than controls, especially during collection periods when the reindeer were kept indoors in cages (cf. Åhman 1988a). We found no such effect of zeolite.

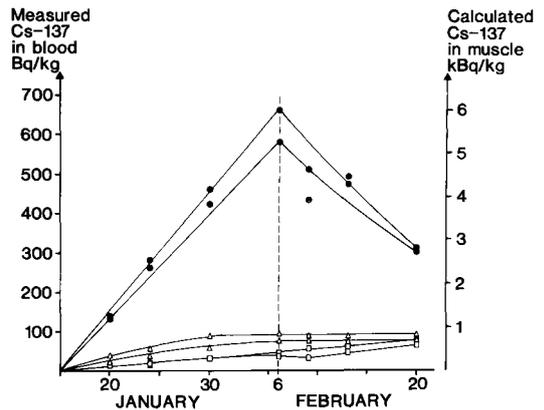


Figure 2. Cs-137 in blood and muscle (calculated: 9 x Cs-137 in blood) from reindeer in Experiment I. The reindeer were fed lichens giving 17-20 kBq Cs-137/day. During the first period, January 15 to February 5, Group 1 (●) got no caesium-binder in their feed, Group 2 (△) got 23 g of bentonite per day and Group 3 (□) got 46 g per day. From February 6 all reindeer got 27 g of bentonite per day.

The changes of Cs-137 activity in blood are shown in Figures 2 and 3. In Experiment I the animals started with a Cs-137 activity at zero level. In Group I (controls) Cs-137 activity in blood increased rapidly to about 600 Bq/kg without levelling in three weeks. Those getting bentonite reached 70 - 90 Bq/kg (Group 2) or about 40 Bq/kg (Group 3).

When the feeding of bentonite continued with 27 g bentonite per day to all animals, the level of Cs-137 in blood of controls decreased to about half the maximum value (Figure 2). In

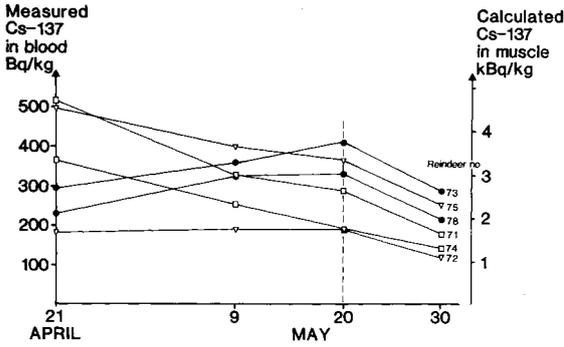


Figure 3. Cs-137 in blood and muscle (calculated: 9 x Cs-137 in blood) from reindeer in Experiment II. Group 1 (●) got no caesium-binder in their feed, Group 2 (▽) got 25 g of zeolite per day and Group 3 (□) got 50 g of zeolite per day. During the first month, April 21 to May 19, the reindeer were fed lichens giving: 8-9 kBq Cs-137/day. From May 20 the reindeer were given 1 kBq/day.

Group 2 radiocaesium activity concentration in blood did not change while for animals in Group 3, that got less bentonite than before, the values increased to the level of Group 2 (70 Bq/kg).

In Experiment II the animals started at higher levels of radiocaesium in blood (200 - 500 Bq Cs-137/kg, Figure 3). In controls the levels increased by about 100 Bq/kg (40% on average) in four weeks. One animal in Group 2 remained on 200 Bq/kg throughout the four weeks, while in the other radiocaesium activity decreased slightly. The average reduction in Group 2 was 18%. Group 3, getting the higher dose of zeolite, decreased by 45% on average. During the ten last days of the experiment, when the reindeer got only 1 kBq of Cs-137/day, blood-Cs decreased by 30 - 40%. No effect of the zeolite was discernible during this phase of the experiment.

Excretion of Cs-137 with faeces and urine in the two experiments is shown in Figures 4 and 5. In Experiment I (Figure 4), total excretion in Group 1 (controls) increased from zero to

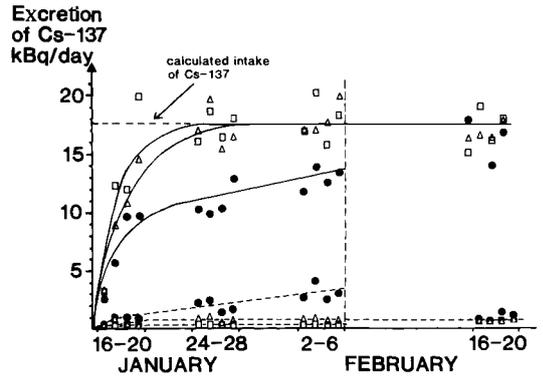


Figure 4. Daily excretion of Cs-137 in Experiment I. Excretion in urine (---) and total excretion, urine + faeces (—), in Group 1, controls (●) and in reindeer receiving bentonite; Group 2, 23 g/day (△) and Group 3, 46 g/day (□). From February 6 all reindeer got 27 g of bentonite per day.

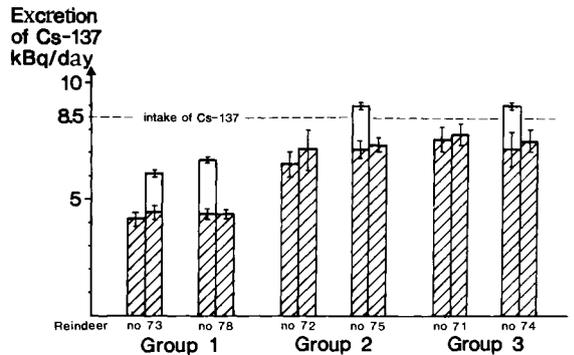


Figure 5. Excretion of Cs-137 in Experiment II. Daily excretion in faeces (hatched) and urine (white) during two periods, April 25-28 and May 16-20, (mean \pm S.D.) in controls (Group 1) and in reindeer receiving 25 g zeolite/day (Group 2) or 50 g zeolite/day (Group 3).

about 11 kBq/day in one week. After three weeks the excretion was about 14 kBq/day with 20 - 35% of Cs-137 being excreted with urine. In reindeer receiving bentonite (Groups 2 and 3), excretion of Cs-137 reached a maximum level at about 18 kBq/day in less than two weeks. The amount of Cs-137 excreted with urine in these groups was 0.3 - 1.0 kBq/day which is 2 - 5% of total excretion of Cs-137.

When all animals were given 27 g/day of bentonite, the total excretion of Cs-137 with faeces and urine in Group 1 (controls) increased. After 10 days it was at the same level as in Groups 2 and 3 (18 kBq/day). The share of Cs-137 excreted with urine decreased. In Groups 2 and 3 the total excretion did not change significantly. The fraction excreted with urine increased somewhat in Group 3.

Figure 5 shows the excretion of Cs-137 in Experiment II. Excretion with urine was measured on one occasion in four of the animals. For the remaining two, only excretion with faeces was measured. For controls (Group 1), excretion with faeces was 4 - 4.5 kBq/day and with urine about 3.5 kBq/day (about 36% of total excreted Cs-137). For reindeer fed zeolite (Groups 2 and 3) the excretion with faeces was 6.5 - 7.8 kBq/day. Excretion with urine was 1.9 kBq/day (21% of total excreted Cs-137). When intake of radiocaesium was reduced to 1 kBq Cs-137/day the excretion with faeces (measured in one animal of each group) was 1.5 - 1.8 kBq/day. There were no significant differences between the groups during this period. Excretion with urine, measured in one of the controls (no 73), was 1.6 kBq/day (50% of total excretion).

Discussion

In the experiment there were fluctuations in the daily intake of radiocaesium due to variations in radiocaesium content of the lichens. The exact lichen intake was also difficult to estimate, since leftover lichens were spilled out in the pen and trampled down.

An estimation of the daily intake could be made by measuring daily excretion of radiocaesium when the animals were at steady state. In Experiment I, the average excretion of Cs-137 in Group 2 during the periods February 2-6 and 17-20, when the animals in this group had constant radiocaesium levels in their blood (Figure 2), was 17.4 ± 1.4 kBq/day (Figure 4). This value, we believe, gives a good estimate of

the daily intake. In Experiment II one animal in Group 2 (no 72) had constant radiocaesium activity concentration in the blood during the first phase of the experiment (Figure 3). In this animal, only excretion of radiocaesium with faeces was measured (Figure 5). Assuming that the part excreted with urine is the same as for the other animals in Group 2 and 3 (21%), the total excreted Cs-137 will be 8.5 kBq/day, which thus should be equivalent to the daily radiocaesium intake.

In these experiments, the radiocaesium activity concentration of blood was used to measure the level of radiocaesium in the body. It has been shown that the activity concentration of radiocaesium in muscle is 7 to 10 times the concentration in blood (Åhman, B 1986, Åhman, G. 1986, Blix 1988, Eikemann 1988). The lower factor applies to animals that are increasing their levels of radiocaesium (Åhman, B. 1986). This seems logical since blood transporting radiocaesium to the tissues should have a higher concentration relative to tissue than blood draining radiocaesium from the tissues. In this report we have used the factor 9 when calculating radiocaesium in muscle. For animals at or near steady state this factor should give a good estimate of radiocaesium in muscle. The factor 9 is probably too high for controls (Group 1) in the beginning of Experiment I, when these animals were increasing their radiocaesium levels rapidly. The factor 7 might give a better estimate of radiocaesium in muscle in this group.

In Experiment I, the controls (Group 1) had, after three weeks of estimated daily intake at 17.4 kBq Cs-137/day, reached blood values corresponding to 4.0 - 4.6 kBq/kg in muscle (7 x Cs-137 in blood). By feeding these animals bentonite (27 g/day), we were able to revert the rapid increase of radiocaesium in the body into a decrease, although the animals had the same radiocaesium intake as before. Reindeer given bentonite throughout the experiment (Groups 2 and 3) seemed to approach radiocaesium le-

vels corresponding to concentrations in muscles at 0.5 - 0.8 kBq/kg. This is well below the current limit for reindeer meat sold in Sweden (1500 Bq/kg).

The conditions of Experiment II differed from those in Experiment I in various respects. The animals were in a growth period, also with some practical problems as a consequence. They were fed less lichens and more hay and pellets. The estimated radiocaesium intake was 8.5 kBq Cs-137/day. The starting level of radiocaesium in the body was much higher than in Experiment I. Also the individual variations

were larger. On average, however, the radiocaesium levels in blood of the controls increased by 40% from April 21 to May 20, while reindeer fed zeolite, Groups 2 and 3, decreased with 18% and 45%, respectively.

Excretion of Cs-137 was measured also in another part of Experiment I, when the reindeer had no radiocaesium intake (cf. Åhman 1988b). The relation between the activity concentration of radiocaesium in blood and the excretion per day was calculated. In reindeer receiving no caesium binder the amount of Cs-137 excreted with faeces was about 5 times

Table 3. Calculated net absorption of Cs-137 from the gastro-intestinal tract in Experiment I. Estimated radiocaesium intake was 17.4 kBq Cs-137/day.

Rein-deer no.	Day	Cs-137 in blood Bq/kg	Estimated excretion in faeces from body deposits kBq/day	Measured excretion in faeces kBq/day	Not absorbed Cs-137 from the feed kBq/day	% of intake	Absorbed % of daily intake
Group 1 - controls			(5 x blood)				
19	Jan 26-28	361	1.80	10.1	8.3	48 %	52 %
	Feb 4-6	572	2.86	10.3	7.4	43 %	57 %
21	Jan 24-26	328	1.64	7.8	6.2	36 %	64 %
	Feb 2-4	602	3.01	9.4	6.4	37 %	63 %
							59 ± 6 %
Group 2 - 23 g bentonite/day			(10 x blood)				
27	Jan 26-28	48	0.48	15.5	15.0	86 %	14 %
	Feb 4-6	72	0.72	18.3	17.6	101 %	-1 %
	Feb 16-18	72	0.72	16.0	15.3	88 %	12 %
28	Jan 24-26	65	0.65	17.3	16.6	96 %	4 %
	Feb 2-4	89	0.89	16.2	15.3	88 %	12 %
	Feb 18-20	89	0.89	16.4	15.5	89 %	11 %
							9 ± 6 %
Group 3 - 46 g bentonite /day			(10 x blood)				
23	Jan 26-28	28	0.28	17.0	16.7	96 %	4 %
	Feb 4-6	38	0.38	16.7	16.3	94 %	6 %
26	Jan 24-26	22	0.22	17.2	17.0	98 %	2 %
	Feb 2-4	41	0.41	18.3	17.9	103 %	-3 %
							2 ± 4 %

the amount of Cs-137 in one kg of blood. For reindeer fed bentonite, the factor was 10. To calculate the absorption of radiocaesium from the feed, we estimated faecal excretion of radiocaesium from body deposits and compared it to the measured daily excretion in Experiments I and II. The results of these calculations are shown in Tables 3 and 4. (Blood values for the actual day are taken from the graphs shown in Figures 2 and 3.) These calculations are based on factors that are somewhat uncertain. The estimated radiocaesium intake greatly affects the calculation, especially if the absorption is small.

According to the calculations, controls, (Group 1) in Experiment I had an average net absorption from the gastro-intestinal tract of 59% (Table 3), while the absorption in Groups 2 and 3, receiving bentonite, was 9 and 2%, respectively. The relation between Group 1 and Group 2, with about 6 - 7 times more radiocaesium absorbed in controls than in those fed bentonite, is in accordance with the results discussed previously. The true absorption in Group 3 is most likely larger than the calculated 2%. With a radiocaesium intake at 18 kBq/day (instead of the estimated 17.4 kBq) the absorption will be 5%.

Table 4. Calculated net absorption of Cs-137 from the gastro-intestinal tract in Experiment II. Estimated radiocaesium intake was 8.5% kBq Cs-137/day.

Reindeer no.	Day	Cs-137 in blood Bq/kg	Estimated excretion in faeces from body deposits kBq/day	Measured excretion in faeces kBq/day	Not absorbed from the feed kBq/day	Cs-137 % of intake	Absorbed % of daily intake
			(5 x blood)				
Group 1 - controls							
73	Apr 26-28	313	1.56	4.2	2.6	31 %	69 %
	May 17-20	414	2.07	4.5	2.4	28 %	72 %
78	Apr 26-29	277	1.38	4.4	3.0	35 %	65 %
	May 17-19	257	1.28	4.4	3.1	37 %	63 %
							67±4 %
Group 2 - 25 g zeolite/day							
72	Apr 26-29	182	0.91	6.5	5.6	66 %	34 %
	May 1-20	187	0.94	7.1	6.2	73 %	27 %
75	Apr 26-28	462	2.31	7.1	4.8	56 %	44 %
	May 17-20	284	1.42	7.3	5.9	69 %	31 %
Group 3 - 50 g zeolite/day							
71	Apr 26-29	431	2.16	7.6	5.4	64 %	36 %
	May 17-20	290	1.45	7.8	6.4	75 %	25 %
74	Apr 26-29	319	1.60	6.2	4.6	54 %	46 %
	May 17-20	203	1.02	7.4	6.4	75 %	25 %
							34±8 %

In Experiment II (Table 4) the average absorption in Group 1, controls, was 67%. The calculated absorption in reindeer fed zeolite, Group 2 and 3, was about half of this, 34% at an average.

The caesium binding characteristics of the zeolite and bentonite were studied *in vitro*, in solutions of KCl and NH₄Cl and in rumen fluid from reindeer (Forberg 1989). Caesium had a higher affinity to the actual quality of zeolite than to the bentonite, indicating that some factor had reduced the efficiency of the zeolite when it was given to reindeer. Artificial mordenite had a somewhat higher affinity to cesium. Artificial mordenite was also shown to be very efficient in experiments with goats and sheep (Forberg *et al.* 1988). It was, however, judged too expensive for practical application on reindeer.

The natural zeolite was ground, before it was mixed into the pellets, until 73% by weight passed through a 32 μ m sieve when wet. Particles of this size are not expected to settle in the gastro-intestinal tract. However, agglomeration might reduce the efficiency of zeolite and may be an explanation for the inferior effect.

Conclusions

Bentonite is a caesium-binder efficient enough to be used in reindeer feeding in heavily contaminated areas of Sweden. With 2% of bentonite in the total food, absorption seems to decrease to about 15% of the absorption when no caesium-binder is added. At an intake of about 20 kBq Cs-137/day, 2% of bentonite in the food should be sufficient to keep radiocaesium levels in muscles below the limit 1500 Bq/kg. Since bentonite increases water requirements, it is important that the reindeer have access to drinking water. This could be a problem especially at low temperatures. Also, due to its unspecific binding of minerals and electrolytes, it is important to keep the bentonite dose at a moderate level.

The zeolite used in our study binds caesium

less efficiently than bentonite. When the reindeer were given 2 or 4% of zeolite in their feed, absorption decreased to about 50% of the absorption without caesium-binder. Zeolite, however, does not have the negative effects of bentonite on water balance. Further studies are required to find a more efficient form of zeolite than the one we have used.

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