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The Use of Modified Clay in Reduction of ^{137}Cs Deposition in Artificially Contaminated Broilers

Gordana VITOROVIĆ[1)], Mitrović BRANISLAVA[1)], Janaćković ĐORĐE[2)], Andrić VELIBOR[3)], Stojanović MIRJANA[4)], Vitorović DUŠKO[4)], Adamović IVANA[5)], Grdović SVETLANA[6)], Petrujkić BRANKO[6)]

[1)]Department of Radiology and Radiation Hygiene, Faculty of Veterinary Medicine, University of Belgrade, 18 Oslobodjenja Bv., 11000 Belgrade, Serbia: radijacija@vet.bg.ac.rs
[2)]Faculty of Technology and Metallurgy, University of Belgrade, 4 Karnegijeva Street, 11000 Belgrade, Serbia
[3)]Institute of Nuclear Sciences Vinča, 11001 Belgrade, Serbia
[4)]Institute for Technology of Nuclear and other Mineral Raw Materials, 86 Franchet d' Esperey Street, 11000 Belgrade, Serbia
[5)]Department of Animal Science, Faculty of Agriculture Zemun, University of Belgrade, 6 Nemanjina Street, 11080 Belgrade, Serbia
[6)]Department of Nutrition and Botany, Faculty of Veterinary Medicine, University of Belgrade, 18 Oslobodjenja Bv., 11000 Belgrade, Serbia

Abstract. Animals, twenty Hubbard broiler chicks, were six weeks old and fed with diets of standard composition that have completely satisfied their nutritional needs. The chicks were randomly allotted into one of the four groups (five chicks per group) and kept in confinement (battery cage). At the beginning of the trial, each chick was orally contaminated with ^{137}Cs of total activity of 3750 Bq. All groups of animals (control and experimental) received radiocaesium, while experimental groups in addition to contaminant, simultaneously received by 5 ml of various caesium binders. I experimental group was given organo-zeolite; II experimental group organo-bentonite solution and III experimental group of chicks received acid-activated sepiolite. The animals were sacrificed 24 hours after artificial contamination and administration of the binders. At that time samples of meat, liver and gizzard for gamma spectrometry determination of ^{137}Cs concentration activity were taken. Administration of organo-zeolite lowered radiocaesium activity from 62.0 to 72.0%. Similar effects were recorded in organo-bentonite administration (59.8 to 67.2%), while efficiency of acid-activated sepiolite in decreasing radiocesium activity was lowest (24.4 to 36.0%). It can be said that the process of modification failed to show beneficial effects to improve radiocaesium binding and eliminating from digestive tract and body of broilers. Efficiency of protection was similar or the same as in the use of natural, unmodified clay minerals.

Keywords: broilers, modified clay minerals, radiocaesium, protection

INTRODUCTION

Radioactive contamination of the environment as a consequence of nuclear accident can lead to contamination of feedstuffs with biologically significant radionuclides: ^{90}Sr, ^{131}I and ^{137}Cs. Prevention of ingestion of these radionuclides is a priority in radioprotection. In

farm producing animals it is necessary to ensure that the contamination of animal products is below intervention levels.

Cessation of feeding contaminated feed and subsequent feeding with uncontaminated feed should be an effective measure in decontaminating the chicken's meat. Second line of countermeasures in radioprotection is the use of caesium binders, which have the ability to bind caesium (in digestive tract) and reduce its accretion in animal soft tissues.[3] Because of mentioned, combination of those two countermeasures is recommended in the cases of environment and feed radioactive contamination.

During past 20 years ammonium ferric hexacyanoferrate (AFCF) is considered the most effective radiocaesium binder, and it is widely used in contaminated animals for inhibition of ^{137}Cs transfer to animal's tissues and organs. Reduction of about 90% ^{37}Cs deposition was achieved in cows supplemented with 3 g AFCF/day, calves and pigs 2g AFCF/day[2] and in broiler chickens with 0.2 g AFCF/day. [14,15,16]

Practical experience gained after the Chernobyl accident, has proven efficiency of natural clay minerals: bentonite and zeolite in preventing high radiocesium levels in animal products. Bentonite was widely used as radiocaesium binder in reindeer[1], rats[13] and broiler chickens.[12] Natural zeolite–clinoptilolite, as one of the most important ion exchangers found in nature, has also been used for adsorption of radioactive caesium in rats[6], rabbits[9] and sheep.[4,10] In our previous studies[15] we have determined that ^{137}Cs binding efficiency of natural zeolite in broiler chickens ranges between 50.0 % and 70.0 %. The main advantage of natural zeolite is high selectivity for radiocaesium beside its low cost and availability in many parts of the world. Serbia has several deposits of high quality clinoptilolite. Modified clinoptilolite has also shown significant efficiency in reducing ^{137}Cs transfer from feed to chicken meat.[11]

Sepiolite is fibrous clay mineral with high ability to absorb inorganic as well as organic compounds. The information about sepiolite efficiency in radiocaesium absorption in domestic animals are scarce. The presence of micro pores and channels together with the fine particle size and its fibrous nature contribute to its mickle surface area. Acid treatments have been used in order to increase the surface area and obtain solids with high porosity and radionuclide ion adsorption power.[5]

The objective of the present study was to investigate the efficiency of three modified clay minerals: organo-zeolite, organo-bentonite and acid-activated sepiolite in ^{137}Cs binding (decreasing ^{137}Cs transfer and deposition in the meat, liver and gizzard) in the single alimentary contaminated broiler chickens.

MATERIALS AND METHODS

Preparation of the binders -the three mineral origin binders used in this trial were: zeolite, bentonite and sepiolite. Zeolite (clinoptilolite) –is natural zeolite tuff from the Beočin deposit (Fruška Gora, Serbia). The cation exchange capacity (CEC), of the clinoptilolite rich tuff was measured with 1M NH_4Cl. The surfactant hexadecyltrimethylammonium (HDTMA), as the Cl-salt (Hoechst AG-Germany) was used for the preparation of organo-zeolite. The zeolite tuff was treated with 5meq/100g surfactant. Zeolite tuff (5 g) was then mixed with 100 ml of the HDTM solution in a turbo mixer at 9000 r.p.m for 3 min. at 50º C. After mixing, the suspension was filtered and the concentrations of exchanged Ca^{2+}, Mg^{2+}, Na^{+} and K^{+} in supernatant were determined using atomic absorption spectrophotometry (AAS). The amount of unreacted amine in supernatant was also determined. The organo-zeolite was then rinsed with distilled water until Cl-ions were no longer detected and then dried at 60º C. This

product was denoted as OA-5. Bentonite used in this experiment was natural bentonite tuff from Sipovo (Republika Srpska). Organic modified bentonite was created by the activation of natural bentonite with a mixture of alkilammonium salts with the alkyl chain length of 14 carbon atoms (Dodigen 1828®, Hoechst AG, Germany). Its grindness was less than 100 μm. Ograno-bentonite was then dried at 105º C and grinned so its granulation was 90% < 50 μm. Sepiolite used as the starting material originated from Andrici (Serbia). The sepiolite was dried at 110º C for 2 h, then powdered and sieved. The fraction between 250 and 800 μm was used in experiment. The acid-activated sample was prepared by dispersion of 10 g of sepiolite in 100 cm^3 of 4M HC solution and suspension was stirred for 10 h at room temperature. The solid was then separated from the solution by centrifugation and rinsed with distilled water until Cl^- ion-free. The obtained acid-activated sample was dried at 110º C for 2 h.

Experimental design -Animals, 42 days old male Hubbard broiler chickens were used. The birds were divided into four groups (5 birds per group) and kept in confinement (cage system). A body weight of chicks was uniform (2.0–2.2 kg). The chicks were fed with standard finishing diet containing 18% of proteins which have completely satisfied their needs (NRC, 1994).[8]) Food and fresh water were given *ad libidum.* The birds of all groups were orally contaminated (gastric tube) receiving a single dose of 3 ml CsCl solution with total activity of 3750 Bq (1250 $Bqml^{-1}$ ^{137}Cs). The first group was control and received no binders. The other three groups, in addition to ^{137}Cs, received organo-bentonite, organo-zeolite and acid activated sepiolite as caesium binders respectively. The chicks in the second group (I experimental) received 2 g of organo-zeolite in 5 ml water suspension. The chicks of the third group (II experimental) received 2 g organo-bentonite in 5 ml water suspension and chicks of fourth group (III experimental) received 2 g of acid activated sepiolite in 5ml water suspension. The application of binders to the chicks was performed immediately after the application of ^{137}Cs and was done by gastric tube. The animals from each group were stunned by electricity and sacrificed by cervical dislocation 24 hours after contamination with CsCl solution and binder administration. The samples of breast meat (220–230g), liver (35-45g) and gizzard (31-43g) were taken from each animal at the time of sacrifice. The samples were homogenized and placed in the counting vessels of defined geometry and kept 2 months frozen at the temperature of -18º C. The ^{137}Cs activity concentration (Bqkg-1 fresh weight) was determined in thawed samples using gamma spectrometry system (HPGe, ORTEC) with pure germanium vertical gamma detector with 30.3% efficiency. The measuring time was 12000 s.

The experimental data's were subjected to the analysis of variance (ANOVA) using the STATISTICA for Windows Software (Stat Soft Inc. version 6). The significance of differences between means was tested using Tuckey´s LSD Test. The protective efficiency (E_{pr}) of modified clay minerals in ^{137}Cs accumulation reduction in the breast meat, liver and gizzard of broiler chicks was calculated in relation to the first (control) group which received no binders and was calculated by the formula [6,7)]

$$E_{pr} = \left[1 - \frac{A_{exp}}{A_{contr}}\right] \cdot 100$$

where A_{exp} is the ^{137}Cs activity concentration in breast meat, liver and gizzard of chickens in the experimental groups and A_{contr} ^{137}Cs activity concentration in breast meat, liver and gizzard of chickens in the control group.

RESULTS AND DISCUSSION

The effects of the administration of different caesium binders on the ^{137}Cs deposition in the breast meat, liver and gizzard in contaminated broiler chicks are presented in *Table 1*.

The broiler chickens of all experimental groups, which were given modified clay minerals as a caesium binders, showed significant reduction in the deposition of ^{137}Cs in breast meat, liver and gizzard, compared to the control group, which did not receive the binders.

Efficiency of organo-zeolite, organo-bentonite and acid-activated sepiolite in ^{137}Cs binding in digestive tract of chickens and inhibition in its transfer and deposition in the breast meat, liver and gizzard was expressed in percents (decreasing percentage relative to control). The result is shown in *Figure 1*.

Tab. 1

^{137}Cs activity concentration (Bqkg^{-1} fresh weight) in breast meat, liver and gizzard of broiler chickens after a single contamination and binders application.

Group	BREAST MEAT (Bqkg^{-1}) Mean ± SD	LIVER (Bqkg^{-1}) Mean ± SD	GIZZARD (Bqkg^{-1}) Mean ± SD
Group 1 - control (^{137}Cs)	$2613 \pm 160^{a,A,B}$	$1038 \pm 80^{b,C,D}$	$2190 \pm 130^{c,E,F}$
Group 2 (^{137}Cs + organo-zeolite)	988 ± 55^{A}	295 ± 20^{C}	631 ± 51^{E}
Group 3 (^{137}Cs + organo-bentonite)	1060 ± 61^{B}	360 ± 26^{D}	728 ± 52^{F}
Group 4 (^{137}Cs + acid-sepiolite)	1984 ± 97^{a}	785 ± 51^{b}	1614 ± 91^{c}

Table legend: a-c $p < 0.05$; A-F $p < 0.01$

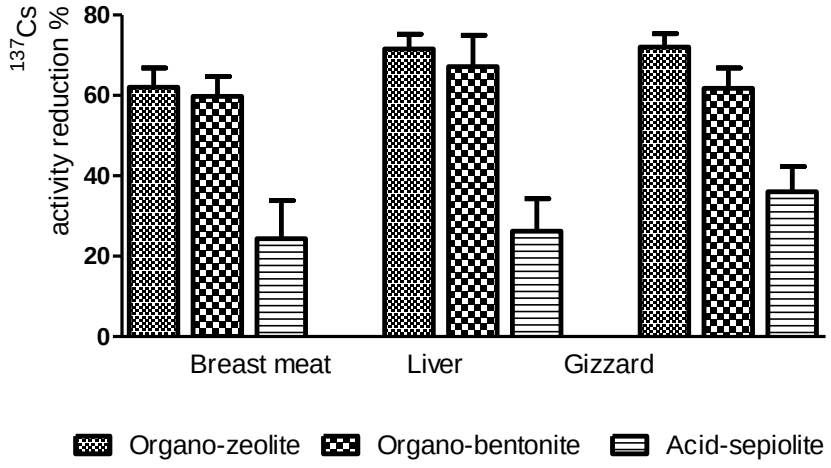

Fig. 1. Percent of decrease in ^{137}Cs accumulation in the breast meat, liver and gizzard of broiler chickens of experimental groups compared to the control group.

Compared to the control group, decrease of ^{137}Cs accumulation in the breast meat of chickens given organo-zeolite was 62.0%, in the liver 71.6% and in the gizzard 72.0%. Similar but slightly lower efficiency was established by administration of organo-bentonite, reduction of ^{137}Cs deposition in breast meat was 59.80%, in liver 67.2% and in gizzard 61.8%.

Administration of acid-activated sepiolite demonstrated lowest protective effects in radiocaesium decreasing, 24.4% in meat, 26.2% in liver and 36.0% in the gizzard.

The obtained results show that in a single ^{137}Cs orally contaminated broiler chickens, simultaneous administration of modified clay minerals, significantly reduces the ^{137}Cs transfer and deposition in breast meat, liver and gizzard. The efficiency of organo-zeolite in reducing ^{137}Cs deposition in muscle, liver and gizzard of chickens, ranged between 62.0-72.0%. This finding is in accordance to the findings of other authors[4,11] who also found that modified clinoptilolite has a significant efficiency in reducing ^{137}Cs transfer from feed to chicken meat. Nevertheless these results are in agreement with our previous study[7] where also efficiency of natural zeolite (clinoptilolite) in ^{137}Cs accumulation reduction was tested. It was then determined that ^{137}Cs binding efficiency of natural zeolite ranged between 50.0% and 70.0%. Modification of zeolite had no effect on increase of radocaesium binding on the digestive tract of broilers, efficiency of protection was similar or the same as in the use of natural, unmodified zeolite.

Similar effects were recorded in organo-bentonite administration (59.8-67.2% ^{137}Cs accumulation reduction) and are in agreement with opinion[12,13] that bentonite could be an effective agent used in case of the environmental radioactive pollution, when radionuclide caesium is released.

According to the researchers from Ukraine[6] based on the radioactivity protective efficiency all substances can be separated into three categories. The first category (insufficient protective action) consists of substances with protective efficiency of 0-30%; the second category (efficient) consists of substances with protective efficiency of 30-60% and the third category (highly-efficient) consists a substance with protective efficiency of 60-100 %.

On the basis of these divisions, organo-zeolite and organo-bentonite (in a dose of 1g/kg body weight) can be classified as substances of the second or even third category, with efficient or highly efficient ^{137}Cs protective action and can be applied as preventive measure in cases of radioactive contamination risk. Acid-activated sepiolite, with low radiocaesium decreasing percentage (24.4-36.0%) proves insufficient protective action.

CONCLUSIONS

It can be seen that the process of modification failed to show beneficial effects to improve radocaesium binding and eliminating from digestive tract and body of broilers. Efficiency of modified clays in radioactivity protection was similar or the same as in the use of natural, unmodified clay minerals.

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REFERENCES

1. Ahman, B. (1996). Effects of Bentonite and Ammonium-feric (III)-hexacyanoferrate (II) on uptake and elimination of radiocaesium in reindeer. J Environ Radioactivity, 31:1,29-50.

2. Giese, W. (1988). Ammonium-ferric-cyano-ferrate (II) (AFCF) as an effective antidote against radiocesium burdens in domestic animals and animal derived foods. Br. Vet. J., 144:363-369.
3. Howard, B., Beresford, N. and Voight, G. (2001). Counermeasures for animal products: a review of effectiveness and potential usefulness after an accident. Journal of Environmental Radioactivity, 56:115-137.
4. Jandl, J. and Novosad, L. (1995). In vivo reduction of radiocesium by modified clinoptilolite in sheep. Veterinarni medicina, 40:237-241.
5. Lazarevic, S., Jankovic-Castvan, I., Jovanovic, D., Milonjic, S., Janackovic, DJ. and Petrovic, R. (2007). Adsorption of Pb^{2+}, Cd^{2+} and Sr^{2+} ions onto natural and acid-activated sepiolites. Applied Clay Science, 37:47-57.
6. Lonin, A. (2009_. Clinoptilolite influence on the radionuclide ^{137}Cs removal from the animal organism. Problems of Atomic Science and technology, 52:5,46-49.
7. Mitrovic, B., Vitorovic, G., Vitorovic, D., Dakovic, A. and Stojanovic, M. (2007). AFCF and clinoptilolite use in reduction of ^{137}Cs deposition in several days contaminated broiler chicks. Journal of Environmental Radioactivity, 95:2-3,171-177.
8. NRC. (1994). Nutrient Requirements of Poultry. 9th ed. Natl. Acad. Press, Washington, DC.
9. Pethes, G., Rudas, P., Bartha ,T. and Frenyo, L. (1988). Accumulation and elimination of radiocesium consumed in fooder from different tissues of broiler chickens and rabbits. Acta veterinaria, 38:299-306.
10. Philippo, M., Gvozdenovic, S., Gvozdenovic, D., Chesters, K., Paterson, E. and Mills, F. (1988). Reduction of radiocesium absorption by sheep consuming feed contaminated with fallout from Chernobyl. Vet. Rec.,122:560-563.
11. Pschl, M. and Øezáè P. (2003). Comparison of three caesium binders affecting the ^{137}Cs transfer from feed to meat of broiler chicken. Acta. Univ. Agric. et Silvic. Mendel. Brno,1:127-134.
12. Rachubik, J. (2000). Effectiveness assessment of oral administration of bentonite or alginate in reducing the transfer of radiostrontium to tissues and organs of rats. Bull. Vet.,44:253-258.
13. Rachubik, J. and Kowalski, B. (2001). Ammonium ferric hexacyanoferrate (AFCF) as a counter measure for reducing the radiocaesium transfer to muscles and inner edible organs of broiler chickens. Bull. Vet. Inst. Pulawy., 45:57-61.
14. Rachubik, J., Dziura, A. and Kowalski, B. (2001). Bentonite from Polish geological deposits as a countermeasure for reducing the radiocaesium transfer to muscle and inner edible organs of broiler chickens. In Mineralstoffe: Mengen-Spuren-und Ultraspurenelemente in der Prävention, hrsg. von Manfred Anke (Stutgart: Wissenschaftliche Verlagsgesellschaft mbH), 361-366.
15. Vitorovic, G., Slavata B., Stosic K., Mladenovic V., Vitorovic D. (2002). The effect of clinoptilolite on ^{137}Cs binding in brolire chickens. Agricultural and Food Science in Finland, 11:137-141.
16. Vitorovic, G. (1993). The application of ammonium-ferric (III)-hexacyanoferrate (II) in the protection of chicken meat from contamination with radicesium. Acta Veterinaria.m 43:351-358.