



Alpha and gamma spectrometry for the radiological characterization of animal feed



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ABSTRACT

Consumption of food is the most common pathway of radionuclides into the human body. Foods of animal origin are important in the European human diet. Therefore, radiation control of animal feeds and animal products will contribute to producing food for human nutrition without, or with low risk, for human health. This paper presents data obtained by alpha and gamma spectrometric analysis for natural radionuclides and ¹³⁷Cs in animal feed. ⁴⁰K, ²²⁸Ac, ²¹²Pb, ²⁰⁸Tl, ²¹⁴Pb, ²¹⁴Bi and ¹³⁷Cs were determined by gamma spectrometry, ²¹⁰Po by alpha spectrometry. In all samples examined, ¹³⁷Cs activity concentration was below the detection limit (0.25 Bq kg⁻¹) except for one sample. The mean activity concentration was 2.26 ± 2.27 Bq kg⁻¹ for ²¹⁴Pb; 2.42 ± 2.24 Bq kg⁻¹ for ²¹⁴Bi; 1.76 ± 1.17 Bq kg⁻¹ for ²¹²Pb; 1.88 ± 1.42 Bq kg⁻¹ for ²⁰⁸Tl; 2.29 ± 1.93 Bq kg⁻¹ for ²²⁸Ac; 3.48 ± 3.48 Bq kg⁻¹ for ²¹⁰Po, and 322.2 ± 115.6 Bq kg⁻¹ for ⁴⁰K. The principal component analysis showed differences in the radioactivity content between samples with dicalcium phosphate and without dicalcium phosphate explaining 76.5% of the total variance. A significant difference for ²¹⁰Po and ⁴⁰K activity concentration was detected between the samples with and without dicalcium phosphate.

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1. Introduction

Consumption of food is usually the most important route by which natural and artificial radionuclides can enter the human body. The foods of animal origin are largely represented in the human diet; the key transfer pathways to animals are ingestion of contaminated feed and in some cases of soil [1]. Therefore control (and measurement) of radioactive content in animal feed will contribute to producing food for human nutrition without or with low risk for health.

Radionuclides are present in the environment either naturally or artificially; the transfer of artificial radionuclides along terrestrial food chains has been studied extensively during the last 40 years. Natural radionuclides have received less research attention than many artificial radionuclides, despite being naturally ubiquitous in the environment. There has been increasing interest in radiological assessments of the discharges of naturally occurring radionuclides in the terrestrial environment, both in terms of current releases from industrial sites and from the legacy sites with historical contamination.

Amongst the alpha emitters, ²¹⁰Po is estimated to contribute about 7% of the effective dose to humans from ingested natural radionuclides

[2]. ²¹⁰Po is relatively important regarding doses due to its high linear energy transfer (LET), and its toxicity is comparable to ²³⁹Pu and about 5 times greater than ²²⁶Ra [3].

²¹⁰Po, and its precursor ²¹⁰Pb, belong to the ²³⁸U series. Their presence in the terrestrial environment arises from ²²²Rn which, once produced, may remain in soil air spaces, decay within the mineral matrix of soil or be released to the atmosphere. ²¹⁰Pb and ²¹⁰Po return to the earth's surface via both wet and dry deposition [4–7], and their presence in vegetation is ubiquitous. Natural concentrations of ²¹⁰Pb and ²¹⁰Po in the environment can be increased locally by anthropogenic activities such as phosphate ore processing, coal-fired power stations, coal mining, metal smelting, etc. [8].

The environment is also contaminated by the presence of the biologically significant radionuclides (¹³¹I, ¹³⁴Cs, ¹³⁷Cs) due to atmospheric nuclear weapon testing (1945–1963) and to a series of nuclear accidents, Windscale 1957, Kyštym 1957, and Chernobyl, 1986 [9,10], or as consequences of natural disasters (Fukushima 2011). The most important long-lived radionuclide is ¹³⁷Cs with long half-life (30.17 years), which distributed in all soft tissues in animals including muscle and milk of animals. Its activity concentration in samples of animal feed and human food decreases with time after deposition according to its biological, ecological and physical half life [11].

Recently, there has been a growing concern about the effect of low level radioactivity on human health. There is a current lack of information on activity concentrations in animal feed for some radionuclides,

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especially for naturally occurring isotopes and for feeds used for mono-gastric agricultural animals; more information should be collected for a range of possible feed sources. Radiometric control of products involved in the food chain is an important part of ongoing quality control of products related to food and feed. These controls contribute to ensuring the safety of animal feed and, ultimately, the health of animals and people and the European Community has determined intervention limits for radionuclides in feeds following a nuclear accident and any other cases of radiological emergency [12].

Animal feed is given routinely to domestic animals in the course of animal husbandry; it is blended from various raw materials and additives and is manufactured by feed compounders as meal type or pellets. Additives are composed of microingredients such as vitamins, minerals, chemical preservatives, antibiotics, fermentation products, and other essential ingredients that are purchased from premix companies for blending into commercial rations. In particular monocalcium (MCP) and dicalcium phosphate (DCP) are used as a feed supplement for farm and domestic animals, as a source of phosphorus and calcium. These two elements are often deficient in the practical diets of livestock [13,14]. Monocalcium phosphate and dicalcium phosphate are derived from either sulphuric acid or hydrochloric acid digestion of inorganic phosphate rock, a raw material often rich in uranium and its decay chain products which are usually in equilibrium with precursors [15]. During the industrial manufacturing processes used for monocalcium and dicalcium phosphate, deviations from secular equilibrium occur, as the various radionuclides are partitioned into various phases according to their solubility. Thus, these products are enriched in significant quantities of some natural radionuclides; in particular, they can be enhanced in ^{210}Po and ^{210}Pb , two radionuclides which are of great interest because their contribution to the dose received by ingestion is large compared with other natural radionuclides [16–18].

Therefore, it is necessary to pay special attention to systematic radiation hygienic control of feedstuffs of vegetable, mineral and animal

origin. Natural and artificial radioactivity has been measured in foodstuffs for human consumption from different countries [17,19–22], but few data have been published regarding natural radionuclide activity concentrations in commercial animal feed. In this study the determination of natural radionuclides such as, ^{214}Pb and ^{214}Bi (two radionuclides coming from the ^{226}Ra decay), ^{228}Ac (from ^{228}Ra), ^{212}Pb and ^{208}Tl (from ^{228}Th), ^{210}Po and ^{40}K , and of the artificial ^{137}Cs was carried out in various manufactured animal feed. The purpose was a) to provide information on the activity concentration of some natural radionuclides to obtain useful baseline values and contribute to the creation of databases on natural radioactivity; b) to evaluate the correlation between the phosphate feed supplements and the natural radioactivity with a particular emphasis on ^{210}Po because of its potentially high contribution to adsorbed doses for internal radiation through ingestion; and c) to evaluate the residual impact of nuclear weapon testing and the Chernobyl accident on the Italy, by the determination of the ^{137}Cs content.

2. Material and methods

2.1. Samples

Analyses were conducted on animal feed samples of Italian origin purchased in Italian stores where the local farmers normally buy these products. Table 1 shows the list of the animal feed samples and their analytical composition. The twenty five analysed samples were: poultry (8), turkeys (1), rabbits (3), pigeons (2), horses (2), pigs (4), bovine (2), mixed feed for ovine and caprine (2), and mixed feed for bovine, horse and ovine (1).

2.2. Analytical method

The samples (weight = 1 kg) were weighed individually, dried in an oven at 105 °C for 24 h and the dried samples were weighed and

Table 1
List of species analysed and their analytical composition (%).

Sample	Dicalcium phosphate	Protein	Cellulose	Fats	Ash	Lysin	Methionine	Calcium	Phosphorus	Sodium
Poultry										
1	With	22	4.0	5.5	6.3	1.0	0.38	1.0	0.58	0.18
2	With	25	3.0	5.0	7.0	1.4	0.55	1.25	0.70	0.12
3	Without	8.0	2.0	2.5	8.5	0.2	0.10	-	-	0.01
4	With	18.5	4.5	4.0	7.0	0.8	0.30	1.2	0.83	0.20
5	With	20.3	4.3	3.0	6.8	1.1	0.43	1.07	0.17	0.72
6	Without	8.4	2.7	1.9	14.0	0.3	0.17	-	-	0.20
7	Without	13	5.5	2.9	3.5	-	-	-	-	0.03
8	With	16.5	3.0	5.5	14.0	0.76	0.35	4.0	0.57	0.15
Turkey										
9	With	30.0	3.4	5.5	7.5	1.57	0.53	1.20	0.90	0.06
Rabbit										
10	with	20.0	6.0	3.0	7.0					0.10
11	With	15.0	17.0	3.5	7.5			0.8	0.70	0.20
12	With	14.7	18.5	3.0	9.5			1.2	0.60	0.20
Pigeon										
13	Without	Analytical	composition	is	not		reported			
14	Without	Analytical	composition	is	not		reported			
Horse										
15	With	14.0	14.0	2.8	8.5					0.40
16	With	10.4	4.2	2.2	2.0	0.47	0.17	0.09	0.30	0.03
Pig										
17	With	33.0	3.8	4.2	13.0	2.1	0.52			0.45
18	With					0.01	0.003	24.5	1.75	5.30
19	With	15.8	5.2	3.0	5.8	0.91	0.48	0.77	0.37	0.16
20	With	17.0	4.5	4.5	6.0	1.0	0.33	0.65	0.65	0.19
Bovine										
21	Without	16.5	9.3	2.3	9.2					0.59
22	Without	16.0	13.0	4.5	8.5					0.40
Ovine-caprine										
23	With	18.0	8.7	4.2	8.6					0.50
24	Without	16.0	7.2	4.0	8.5					0.50
Bovine horse ovine										
25	Without	11.61	4.77	3.02	2.37	0.50	0.17	0.09	0.34	0.02

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