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DETERMINATION OF Cs, Rb AND Ba IN NEUTRON-ACTIVATED BIOLOGICAL MATERIALS

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In the present paper, the determination of Cs, Rb and Ba in biological materials, including human bones, eggs, milk, meat and vegetables by neutron activation analysis is described. The proposed technique is based on the measurement of the long-lived radioisotopes ^{131}Ba, ^{134}Cs and ^{86}Rb with a Ge(Li) detector, after a simple radiochemical separation based on selective retention of these nuclides on HAP in 6M HF. Detection limits of 0.001 ppm can be achieved with 5% precision. Data are given for replicate determinations in six different types of materials.

Introduction

There is interest, mainly for radioecological studies, in the determination of cesium, rubidium and barium in biological materials, in order to know "normal" values of those elements in the human body and in the components of the average human diet.

Contaminant radionuclides are generally diluted in their stable isotopes. Neglecting isotopic effects, in the absorption of radionuclides by living organism an equilibrium is reached between the specific activity of the element in the organism and that of the environment. Thus, a knowledge of the environmental concentrations of stable elements should indicate the maximum possible concentrations of the radionuclides for equilibrium conditions.

The determination of cesium, rubidium and barium in biological samples by neutron activation analysis cannot always be accomplished by non-destructive instrumental analysis. The relatively high contents of phosphorus, bromine, scandium and zinc usually interfere, especially if good accuracy is required and if at the same time low levels of Cs, Rb and Ba are involved.

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Different solutions have been proposed to perform simultaneous analyses of Cs, Rb and Ba in biological materials.

Fourcy, et al.[1,2] described a general semi-automatized method for the determination of Cs, Rb and Ba in plants, based on ion-exchange, but the technique is rather time-consuming. Sowden, et al.[3] have determined Ba and Sr in bones by neutron activation analysis based on short-lived radioisotopes and a long chemical processing. Koch,[4] Haller,[5] Ewing[6] and Heinonen[7] have determined Cs, Rb and Ba in different types of matrices of radioecological interest, but unfortunately the radiochemical separation techniques used are not quite satisfactory for application to routine analysis.

Merlini, et al.[8] have studied some elements associated with reactor effluents and radioactive wastes in the aquatic ecosystem. However, data for Cs and Rb are limited.

Grummitt, et al.[9] have performed a complete study on Cs, Ba and Ca contents of some components of the human diet. The data presented by Grummitt, and those of Bowen[10] on determinations in bone and milk were compared with our results and good agreement was found in all data.

In the present paper, the determination of Cs, Rb and Ba in biological materials, including human bones, eggs, milk, meat and vegetables by neutron activation analysis is described. The proposed technique is based on the measurement of the long-lived radioisotopes ^{131}Ba, ^{134}Cs and ^{86}Rb after a simple radiochemical separation based on selective retention of these nuclides on HAP in 6M HF.

Experimental

Standards

Standard solutions were prepared from the following Specpure chemicals: $Ba(NO_3)_2$, RbCl and CsCl. Aliquots of solutions containing 5 – 10 μg of each element were weighed in fused silica containers, dried at 90 °C and sealed.

Samples

Samples of eggs, milk, meat and vegetables prepared from freeze-dried fresh material were carefully powdered in a ball mill. Sample collection was carried out from daily consumer products.

Bone samples, corresponding to normal children and adults, were carefully cleaned, dried and powdered.

Bone samples weighing about 40 mg, and all other samples weighing about 300 mg, were put in silica containers and sealed.

Irradiations

Irradiations were performed in the RA-3 reactor at an integrated neutron flux of 10^{18} n . cm^{-2}. Each run consisted of four samples with its corresponding standards.

Chemical processing

After a period of about 8 – 10 days, short-lived activities, mainly ^{24}Na, ^{82}Br and ^{42}K, have decayed and allow treatment of the samples with low exposure dose.

At this stage the gamma-ray spectra of most samples show the presence of ^{65}Zn, ^{60}Co, ^{59}Fe, ^{124}Sb, ^{46}Sr, ^{47}Sr, ^{51}Cr and bremsstrahlung radiation due to ^{32}P. Exceptionally, in some samples the 1,078 keV peak of ^{86}Rb and even the 124 keV peak of ^{131}Ba could be identified.

Samples with carriers of cesium, rubidium and barium and ^{133}Ba, ^{137}Cs, ^{84}Rb or ^{83}Rb as tracers were put in a flask for their mineralization. A phosphorus hold-back carrier was added to the bone samples.

Mineralization was carried out by adding several portions of fuming HNO_3 and heating. The solution was next concentrated, cold 30% H_2O_2 was added once, and the solution was again heated until colourless. Finally it was heated to dryness. The residue was taken up in 6 – 8 ml of 6M HF and heated until dissolution. At this stage, a fine precipitate of CaF_2 may appear, but it does not interfere in the subsequent separation or in the final determination. The radiochemical separation of Cs, Rb and Ba was carried out by selective retention in an inorganic absorber. Hydrated antimony pentoxide in a 50 x 8 mm plastic column previously washed with 10 ml of 6M HF was used as absorber, as recommended by Girardi, et al.[11] The mineralized sample was passed through the column at a flux of 2 ml/min and washed with 25 ml of 6M HF and finally all the absorber material was transferred into a measuring vial.

Standards and tracers were also retained in HAP with 6M HF in batch operation.

Measurements

Measurements of activities were performed in a 40 cm^3 Ge(Li) detector coupled to a 4096 M.C.A.

The detector-preamplifier system used had a resolution of 3.5 keV for the 1,332.5 keV gamma-ray of ^{60}Co.

Samples were counted for 120 min each and peak areas computation were evaluated by the method proposed by Wasson.[12] When possible for Cs and Ba, peak area ratios were used as a purity criteria.

Some improvements have lately been made for routine analysis. Measuring time per sample has been reduced to 60 min (with better statistical precision) using a larger Ge(Li) detector (14% efficiency) coupled to a twelve-position sample changer allowing overnight counting.

Results and discussion

Final results and chemical yields for six different types of samples are given in Table 1.

Precision in all cases was better than 10% and detection limits are low, especially for Cs (0.001 μg) which may be even lower by increasing the decay time before counting.

Table 1

Cs, Rb and Ba contents of biological samples

Sample	Cs, ppm	Yield,%	Rb, ppm	Yield,%	Ba, ppm	Yield,%
Human bones	0.104±0.019	96.1	1.98±0.13	82.0	4.09±0.32	66.0
	0.092±0.017	95.6	1.87±0.13	83.4	4.16±0.36	67.2
Lettuce No. 1	0.019±0.002	97.9	31.05±1.88	44.0	18.98±1.53	66.7
	0.019±0.002	94.0	31.74±1.90	42.7	24.41±1.65	69.2
	0.018±0.002	93.2	32.08±1.87	44.0	22.17±1.51	68.7
Lettuce No. 2	0.016±0.002	96.2	34.22±1.67	46.7	23.02±1.89	69.0
	0.018±0.003	95.7	30.91±1.56	41.6	21.11±1.43	64.7
Salt wort No. 1	0.036±0.004	57.1	16.36±1.10	41.3	37.07±2.44	86.0
	0.044±0.005	56.4	15.90±1.39	42.4	36.06±2.35	89.1
	0.039±0.004	55.2	15.99±1.07	41.7	37.01±2.36	87.6
Salt wort No. 2	0.036±0.004	54.7	15.67±1.21	42.0	35.80±2.06	82.1
	0.039±0.005	58.2	16.96±1.46	44.2	36.54±2.07	84.6
	0.003±0.0007	67.6	7.00±0.41	67.2	2.56±0.13	65.9
Eggs	0.004±0.0005	66.2	7.10±0.39	66.9	2.44±0.11	66.7
	0.003±0.0005	65.1	7.20±0.41	67.4	2.77±0.13	68.2
	0.008±0.0009	98.4	4.78±0.26	87.5	4.14±0.37	67.5
Milk	0.008±0.001	97.6	5.80±0.33	86.1	3.92±0.58	66.0
	0.009±0.001	98.0	5.07±0.33	88.6	4.11±0.55	65.0
Meat No. 1	0.010±0.003	96.3	4.89±0.34	86.8	0.26±0.02	72.0
	0.011±0.003	95.6	4.74±0.31	88.3	0.20±0.02	73.4
Meat No. 2	0.012±0.004	97.9	4.55±0.36	85.9	0.18±0.02	75.4
	0.014±0.004	96.1	4.67±0.34	81.0	0.17±0.02	76.2

On the other hand, it has to be pointed out that retention yields of Cs, Rb and Ba varied with the nature of the matrix, but were constant for every nuclide determined in each type of matrix.

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