

Human hair radioactivity in the Chernobyl area

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Abstract

Studies of recent decades have shown that the elemental composition of human hair can be considered as an indicator of both internal and external human body status. However, there are only a few studies on human hair radioactivity. The Chernobyl accident necessitated the study of the hair of various groups of inhabitants of the contaminated area. Data on hair radioactivity and elemental composition allowed us to draw the following conclusions.

(1) When account is taken of the simplicity of sampling and measurements of human hair activity in polluted areas can be used as a monitor for fast detection of the scale and area of pollution.

(2) Measurement of hair α and β activity is useful for preliminary body burden determination while the whole body counter is inapplicable.

(3) Study of activity along the hair strand allows dating of contamination with an accuracy of about 10 days and gives information about the removal of radioactivity from the body.

(4) Determination of uranium could give additional information about the distance of nuclear fuel transport from the damaged reactor.

(5) Elemental composition of hair and profiles along the strand reflect the influence of medical treatment and can be used to study post-accident health status.

1. Introduction

For some decades the elemental composition of human hair has been a subject of interest in various fields of science. A number of investigations have shown that hair elemental composition reflects both the internal elemental status of the human body and the environment. Radionuclides, being chemical elements, ought to behave in an organism like the trace elements. Hair radioactivity is commonly measured in order to determine the surface contamination during and after various accidents. Many investigators conclude that hair could function as a minor excretory organ especially for toxic elements. Actually the amount of mercury found in hair in cases of acute poisoning is quite close to the toxic dose. So, why not consider hair also as an excretory organ for radionuclides? In this case hair radioactivity (or radionuclide composition) can be used for body burden determination, especially for α and β emitters, when the whole body counter is unusable.

There are only a few studies of hair radioactivity. The most-studied radionuclides are ^{134}Cs and ^{137}Cs [1, 2]. They deposit in hair throughout the time of hair life. Specific activity rapidly reaches dynamic equilibrium with the whole body. For example, with Alaskan Eskimos it was shown that whole body ^{137}Cs activity equal to

37 kBq corresponds to hair activity of 0.05–0.13 Bq g^{-1} for males and 0.02–0.33 Bq g^{-1} for females [2].

Analyses of samples taken from reindeer breeders of the northern part of the former USSR gave values from 0.08 to 0.3 Bq g^{-1} . Comparison of ^{137}Cs concentration in hair and muscle of inhabitants of 49 cities of the former USSR allowed the following correlation to be found:

$$Q_m = 405.4 + 16.2Q_h \quad (1)$$

where Q_m and Q_h are activities of ^{137}Cs in muscle and hair, both referred to 1 g of potassium.

We did not find any studies of such a kind for other nuclides.

An urgent necessity for the study of hair activity was connected with the Chernobyl accident. The present study was focused on the development of a method for the routine screening of population contamination and monitoring of post-accident changes.

2. Experimental details

2.1. Sample collection and treatment

Hair samples were taken from inhabitants of the cities Minsk and Kiev who did not visit the accident area (June, 1986), participants from Moscow in after-accident work after they returned (October, 1986), and

inhabitants of the Chernobyl area (Chernobyl, Prip'yat, Bucha, Borodyanka), staff of the power station, and inhabitants of the cities Kharkov and Gomel who did not visit the accident area (January, 1987). The samples were collected in accordance with IAEA recommendations for element analysis [3]. Samples were washed to remove surface contamination in accordance with the same recommendations.

2.2. Measurements and analysis technique

Measurements of α activity were carried out by two methods.

In the first method a silicon semiconducting detector was used. A thin layer of hair (thickness of about a single hair) was placed in a small bag made of Macrofol foil (thickness of 3 μm). The samples were placed in a vacuum chamber and pressed against the surface of the detector. The efficient area of the detector was $10 \times 40 \text{ mm}^2$. The vacuum was 10^{-2} mmHg . Exposure was 2–4 h. Detector efficiency was determined with a standard ^{238}Pu source.

The second method was the track detector technique. Foil (LR-115) $2 \times 10 \text{ cm}^2$ in size was contacted with a thin layer of the sample for 15–20 days. Then the detector was etched by 20% aqueous NaOH solution at 60 °C for 2 h. The length of the tracks shows that α emitters in most cases were incorporated into the internal layer of a single hair.

The efficiency of registration of the track detector was a little less than that of the Si detector owing to the role of the critical angle of registration. When samples were ashed the efficiency of registration by the track detector increased up to 5 times. However, in this case the losses of radionuclides during ashing were unknown and this method of sample preparation was not used in the present work. Data obtained by the two described techniques corresponded to each other satisfactorily.

To determine the background levels of hair α radioactivity the samples from a remote area (Tashkent) were measured. The measured values were from 7×10^{-5} to $2 \times 10^{-4} \text{ Bq g}^{-1}$. The data for samples from the Chernobyl area were from 7×10^{-4} to $2 \times 10^{-2} \text{ Bq g}^{-1}$.

Measurements of β activity were carried out with a β counter with gas flow proportional counter. The background was less than 10^{-2} Bq or $10^{-3} \text{ Bq g}^{-1}$ for a 100 mg sample. Reproducibility of measurements was about 0.05. Usually β radioactivity of hair is caused by the presence of ^{40}K and can reach 0.03 Bq g^{-1} [1]. Average β radioactivity of samples taken from inhabitants of a remote area (Tashkent) was 0.01 Bq g^{-1} . Average β activity of samples taken from inhabitants or visitors of an accident area reached 2.3 Bq g^{-1} , and the maximal value for some hair sections reached 24 Bq g^{-1} . In some of the samples radiochemical separation

of ^{90}Y was carried out. The samples were ashed. Ash was dissolved in hydrochloric acid. After 10 days yttrium was extracted by monoisooctylmethylphosphonic acid. The β activity of ^{90}Y was measured and identified by decay curve analysis.

γ Spectra were measured with a Ge(Li) detector and computer analyser. The exposure was 3 h. The background spectra (Tashkent) contained peaks of ^{40}K , $^{212,214}\text{Pb}$, ^{214}Bi , ^{228}Ac , and ^{208}Tl . Spectra for hair taken from exposed people contained in addition peaks of ^{60}Co , ^{95}Nb , ^{95}Zr , $^{134,137}\text{Cs}$, ^{140}Ba , ^{140}La , and $^{141,144}\text{Ce}$.

For elemental analysis instrumental activation analysis was used [4]. Samples of 20–70 mg were sealed into bags made from thin foil of pure polyethylene. Samples together with standards were irradiated by a flux of $10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ for 15 s. Then the spectra were measured to determine short-lived isotopes (^{37}Cl and ^{124}I) and again after 1 h (^{24}Na , ^{56}Mn , and ^{64}Cu). After one week samples were packed also in foil of high purity aluminium and irradiated for 15 h. After one week of "cooling" the spectra were measured to determine middle-lived isotopes (^{47}Sc from Ca, ^{82}Br , ^{141}La , ^{198}Au , ^{239}Np from U), and one month later for determination of long-lived isotopes (^{46}Sc , ^{51}Cr , ^{56}Fe , ^{60}Co , ^{65}Zn , ^{75}Se , ^{85}Sr , ^{95}Zr , $^{110}\text{Ag}^m$, ^{124}Sb , ^{134}Cs , and ^{203}Hg). Reproducibility of the analyses was 0.05–0.2. For accurate examination the standard reference material HH-1, human hair (IAEA) [5], was used.

Activity measurements and analyses were carried out both for strands 0.5–1.0 cm in length and for small sections (up to 5 mm in length). Study of smaller sections allowed us to detect changes in time, while, as is known, the changes in body status are "recorded" along the hair. On the graphs of these data the x axis is given in months of 1986–1987 with the assumption that the hair growth rate is about 1 cm per month.

3. Data obtained and discussion

As a result of the accident about 3%–5% of the fuel was ejected into the atmosphere in the form of so-called "hot particles" containing α emitters such as $^{238,239,240,241}\text{Pu}$, ^{241}Am , and $^{242,243}\text{Cm}$. These particles have sedimented mainly within a distance of 60 km from the power plant. Soils in the vicinity of the plant contain about 7.3 Bq g^{-1} of ^{238}Pu , 6.9 Bq g^{-1} of ^{242}Cm , $0.12\text{--}17.4 \text{ Bq g}^{-1}$ of ^{134}Cs , and $0.49\text{--}47 \text{ Bq g}^{-1}$ of ^{137}Cs [6, 7]. The material ejected contained *inter alia* ^{90}Sr , mainly in insoluble or hardly soluble forms [8], and strontium was also distributed in the vicinity of the plant. Nuclides of caesium were in more soluble forms and can migrate to longer distances. This as well as our data allows us to conclude that elevated hair radioactivity is connected with the accident.

3.1. Average data

More than 80 samples were collected and analysed. The intervals of data obtained for α and β activity are given in Table 1. The spread in values (wide intervals) could be explained by differences in patients' life and work conditions and durations of their stays in the accident area.

3.2. Remote areas

Remote areas include the following cities: Kiev (about 90 km to the south), Gomel (about 140 km to the north-east), Minsk (about 310 km to the north-east), Kharkov (about 440 km to the south-east).

α Activity for this group was $0.02\text{--}0.06\text{ Bq g}^{-1}$: considerably higher than background levels ($7 \times 10^{-5}\text{--}2 \times 10^{-4}\text{ Bq g}^{-1}$). The distribution of α emitters along the hair was very irregular. As an example Fig. 1 gives profiles of α activity along a single hair of an inhabitant of Kiev who did not visit the Chernobyl area after the accident. The total activity (curve a), the activity of the surface (curve b), and the activity

TABLE 1. Intervals of α and β activity of hair

Group	α activity (Bq g^{-1})	β activity (Bq g^{-1})
Plant staff	0.0045–0.14	0.01–1.9
Inhabitants of 30 km zone	0.006–0.19	0.01–2.3
Inhabitants of remote cities	0.02–0.03	0.01–0.24
Post-accident workers (Moscow)	0.0007–0.018	0.048–12.3

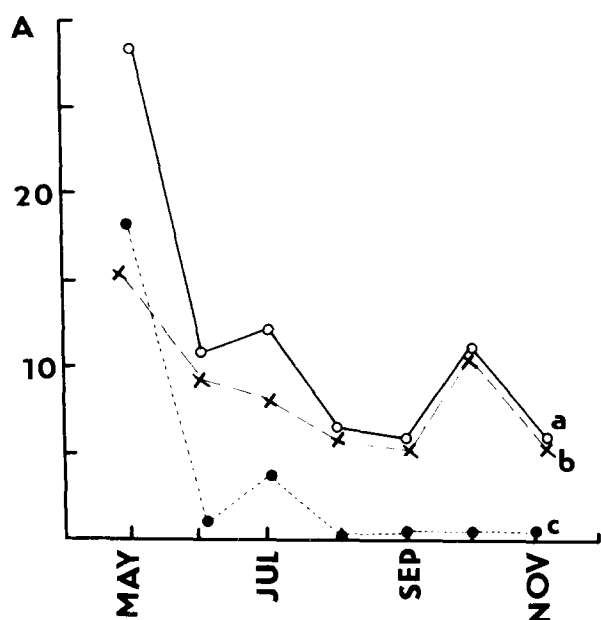


Fig. 1. Distribution of α activity along the hair strand of an inhabitant of Kiev: curve a, total activity; curve b, internal activity; curve c, surface activity.

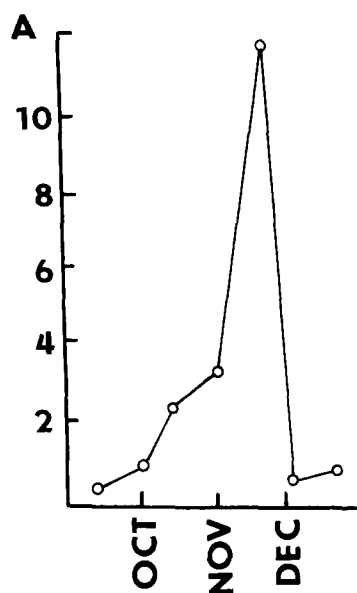


Fig. 2. Distribution of β activity along the hair strand of one individual who once visited the accident area.

of the internal layer (curve c) are shown. Elevated activity of the surface was detected only for the initial months after the accident and could be more or less caused by surface contamination. However, incorporated activity remains elevated for the whole length of the hair and could reflect the internal body status. Additional peaks probably correlate with additional ejections from the damaged reactor. The activity of the proximal end of the hair can be explained by the long excretion period of plutonium from the human organism. The α activity profile for a more remote city (Minsk) also contains additional peaks which can be explained by additional ejections of various nuclide compositions [8].

Maximum β activity for this group reached 0.24 Bq g^{-1} .

In γ spectra of samples taken from inhabitants of remote cities the peaks of ^{60}Co , ^{95}Nb , ^{95}Zr , $^{134,137}\text{Cs}$, ^{140}Ba , ^{140}La , and $^{212,214}\text{Pb}$ were found. Individual activities of these nuclides were $0.012\text{--}0.047\text{ Bq g}^{-1}$. The total γ activity reached 0.2 Bq g^{-1} . It is of interest that the activities of nuclides (^{60}Co and ^{95}Zr) which can be more or less connected with construction materials were comparable or higher than the activities of fission products. Can this serve as evidence of an elevated migration ability of these nuclides?

3.3. Post-accident workers

This group includes the persons who visited the Chernobyl area and were living in Moscow at the moment of sample collection. For this group different levels of contamination could be assumed. However, this group was not directly contaminated by the first explosion.

This is why the variations for α activity (presumably mainly ^{239}Pu) were higher than for β activity (as shown by results of radiochemical analysis, usually about 50% of β activity was caused by ^{90}Sr).

Intervals of β activity are given in Table 1. Figure 2 gives the profile of β activity of a person who spent, in November, 1987, one week in the Chernobyl area. It is characteristic for this group that the peak of activity corresponds to the time of stay in Chernobyl. The activity of the distal end of hair is much less than that of the proximal. This can serve as evidence that the surface contamination was removed by the sample pretreatment procedure. The higher activity of the proximal end could be connected with the excretion of incorporated activity from the organism.

The γ activity of samples for this group was significantly elevated. The γ spectrum of one of the most contaminated persons (a volunteer from Moscow who took part in post-accident work from the very first days) contained peaks of ^{208}Tl , $^{134,137}\text{Cs}$, ^{228}Ac , ^{60}Co , and other nuclides. Activity was 0.04 Bq g^{-1} for ^{134}Cs , 0.024 Bq g^{-1} for ^{137}Cs , and 0.07 Bq g^{-1} for ^{60}Co . The concentration of ^{137}Cs in muscle calculated in accordance with eqn. (1) reached 40 Bq g^{-1} or 1.1 MBq of ^{137}Cs in total skeletal muscle (supposing the muscle mass of a "standard man" to be 28 kg [9]).

3.4. Inhabitants of the 30 km zone and power station staff

This group included inhabitants of Pripjat who moved after the accident to Chernobyl who did not work at the plant, inhabitants of Chernobyl, Bucha, and Borodyanka who did not work at the plant, the plant employees (including those who arrived after the accident), and participants in post-accident work who were in the Chernobyl area at the time of samples collection.

Intervals of measured α and β activity are given in Table 1. Figure 3 gives the profile of α activity of one of the employees of the plant. The surface contamination was probably not completely removed. From this assumption the elevated activity of the distal end corresponded to the period before the accident. The peak of May is probably caused by the accident. The elevated activity of the proximal end indicates continual excretion of activity from the body.

The wide interval of the measured β activity is probably due to very different life and work conditions. Figure 4 gives profiles of the β activity of a female who lived in Pripjat and was evacuated after the accident to Chernobyl (curve a), a child who lived in Pripjat and was evacuated after the accident to a remote part of the country (curve b), and an employee who worked at the plant before the accident and stayed there until the date of sampling (curve c). The high activity of

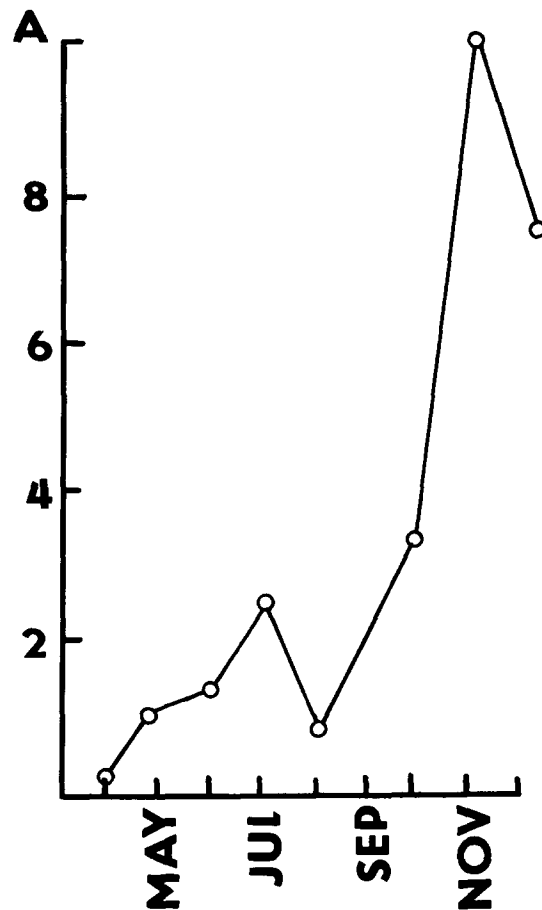


Fig. 3. Distribution of α activity along the hair strand of an employee of the Chernobyl plant.

the proximal parts of the hair indicates overloading of the body by β emitters and continual excretion from the organism.

γ Spectra contained peaks of ^{60}Co , $^{134,137}\text{Cs}$, ^{212}Pb , ^{214}Pb , and ^{228}Ac . The ratio of ^{134}Cs to ^{137}Cs was more than 1 for plant employees and less than 1 for inhabitants not working at the plant.

3.5. Elemental composition

Elemental analysis was focused on the search for elevated uranium concentrations (^{238}U from fuel of low enrichment) and detection of specific changes in hair composition.

The uranium concentration was less than 0.001 ppm in inhabitants of Minsk, 0.079–0.16 ppm in inhabitants of Kiev, and reached 0.31–0.52 ppm in the hair of plant staff and inhabitants of the 30 km zone. Elevated uranium as a rule in all samples was found only in the parts corresponding to May 1986. Other parts of hair strands contained background levels of uranium.

Specific changes in element concentrations were found for Ca, Br, Sc, Co, Fe, and Zr. Analysis of the changes after the accident showed that there are no

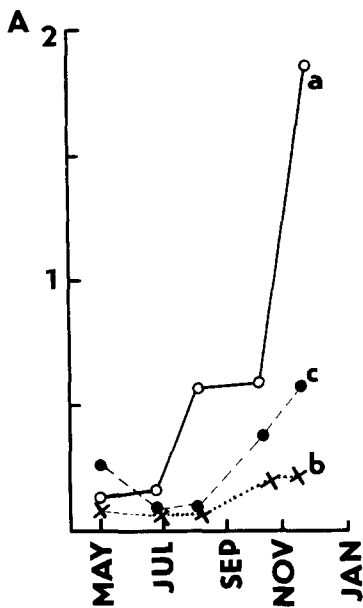


Fig. 4. Distribution of β activity along the hair strand of an inhabitant of Pripyat who moved after the accident to Chernobyl (curve a), an inhabitant who left the Chernobyl area within 1 month (curve b), and an employee who stayed at the plant (curve c).

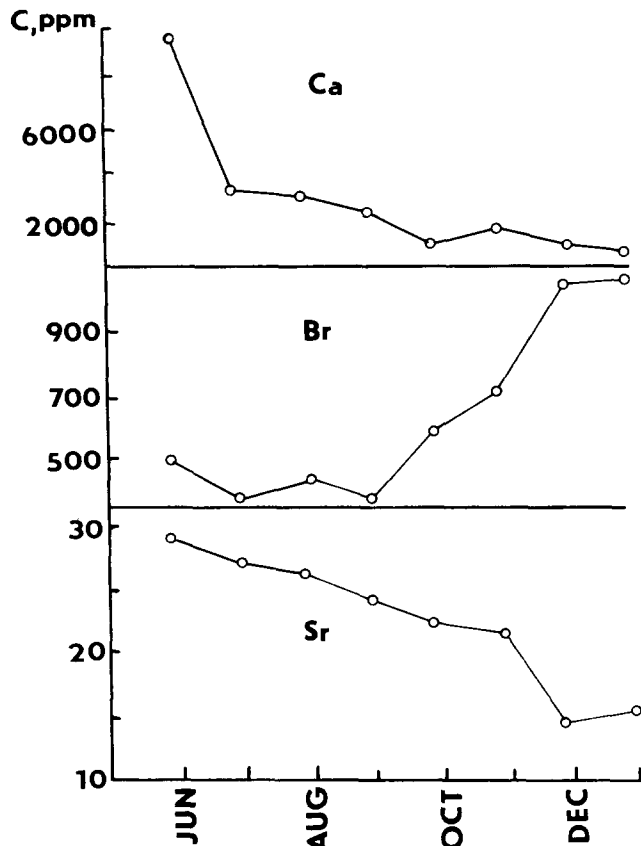


Fig. 5. Distribution of some elements along the hair strand of one employee of the plant.

signs of a return to the normal composition. Changes specific for cardiovascular and cancerous diseases were often detected.

As an example some curves for a female from the plant staff are given in Fig. 5. The distribution of β activity probably indicates a continuous input of nuclides. In this sample ^{212}Pb , ^{134}Cs (0.022 Bq g^{-1}), ^{137}Cs (0.013 Bq g^{-1}), ^{208}Tl , and ^{228}Ac were found. The concentration of lanthanum is about 5 times higher than in the normal population. Similarly, cerium is 10 times, and zinc 5 times, higher. Decreased levels of selenium could be evidence of gastrointestinal dysfunction, and the decreased level of calcium indicates possible cardiovascular dysfunction.

4. Conclusion

The data obtained were proposed as a background for a wide programme, which had included radiochemical analysis, population and geographical screening, drawing maps of hair radioactivity analogously to the maps for hair elemental composition [10] etc. Unfortunately this programme was not accepted. Nevertheless, the preliminary data given in the present paper allow us to consider this approach as an important, inexpensive and simple additional method for the determination of the scale of various radiation accidents, the study of correlations of radionuclide (including actinide) accumulation in human body tissues, fluids, and hair in order to develop a simple method to determine whole body burden, the study of actinide behaviour in organisms etc.

References

- 1 A.A. Moiseev, Cesium-137, *Environment*, Energoatomizdat, Moscow, 1985 (in Russian).
- 2 T.M. Beasley and H.F. Palmer, *Health Phys.*, 11 (1965) 454.
- 3 Yu.S. Ryabukhin, *J. Radioanal. Chem.*, 60 (1) (1980) 7-31.
- 4 L.I. Zhuk, N.S. Osinskaya, T.Y. Kutyakova, I.N. Mikholskaya and S.V. Agzamova, *Lab. Delo*, (1) (1987) 53-55 (in Russian).
- 5 S.B. M'Baku and R. Parr, *J. Radioanal. Chem.*, 69 (1982) 171-180.
- 6 Yu.V. Dubasov, A.S. Krivokhatskiy and N.N. Khramov, *Radiokhimiya*, (4) (1992) 119-124 (in Russian).
- 7 N.A. Loschinov, V.A. Kashparov, V.D. Polyakov, E.B. Yudin, V.P. Protsak, M.A. Zhurba and A.E. Parshakov, *Radiokhimiya*, (4) (1992) 113-119 (in Russian).
- 8 I.I. Bobovnikova, K.P. Makhonko, A.A. Siverina, P.A. Rabotnova, V.P. Gutoreva and A.A. Volokitin, *At. Energ.*, 71 (5) (1991) 449-454 (in Russian).
- 9 W.S. Snyder, M.J. Cook, E.S. Nasset, L.R. Karhausen, G. Parry Howells and I.H. Tipton, *Report of the Task Group on Reference Man*, Pergamon, Oxford, 1974.
- 10 L.I. Zhuk and A.A. Kist, *Biol. Trace Elem. Res.*, 26-27 (1990) 307-320.