SIMULTANEOUS DETERMINATION OF $^{232}$Th AND $^{238}$U IN BIOLOGICAL SAMPLES. APPLICATION TO THE ESTIMATION OF THEIR DAILY INTAKE THROUGH DIET

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A simple method employing neutron activation and radiochemical separation was developed for simultaneous determination of the concentrations of $^{232}$Th(Th) and $^{238}$U(U) in biological materials. Using this method, it is possible to detect 0.05 and 0.2 ng of Th and U, respectively, in the samples. This method was applied to determine the daily dietary intake of these two nuclides by the population living in the high background areas of India (Monazite area), where the soil contains very high levels of these two nuclides. The comparison of the daily intakes by the population living in high and normal background areas showed significantly higher intake of these two nuclides by the high background population.

Introduction

$^{232}$Th and $^{238}$U are two important naturally occurring radionuclides, which are used in the production of nuclear energy. Certain areas in India (in the states of Tamilnadu and Kerala), Brazil and Egypt have very high concentrations of these two radionuclides (especially $^{232}$Th) in soil. The population gets exposed to these nuclides by virtue of living in these areas, and also during mining, milling and processing of radioactive mineral monazite. Both of these radionuclides are radio-toxic as well as chemically toxic to the human system, when present even at low levels. It is important to study their concentrations in different biological materials, such as food, tissues, etc., to understand their metabolic behavior and also to assess the exposure of occupational subjects to these radionuclides.

There are large variations in the reported concentration values for similar kinds of biological samples. The variations could be because techniques such as α-spectrometry, fluorimetry and spectrophotometry employed for analysis of biological materials are not sufficiently sensitive to perform reliable analyses at low levels of concentration (< 5 ng·g$^{-1}$) as are generally found in biological materials. Large reagent blanks are also reported to be associated with these analytical procedures.

WRENN et al.$^2$ have reported the detection limit of 46 ng (this value was converted from activity) for $^{232}$Th using α-spectrometry. FISHER et al.$^3$ reported detection levels for $^{238}$U ranging from 14–42 ng using α-spectrometry procedures. WELFORD and
BAIRD\(^4\) used a fluorimetric method to determine the concentration of \(^{238}\text{U}\) in urine and other biological materials and reported a detection level of 5 ng. They found the reagent blank for the whole analytical procedure to be 7 ng. The detection limit for \(^{232}\text{Th}\), using fluorimetry, was reported by SILL and WILLS\(^5\) to be 10 ng. PETROW and STREHLOW\(^6\) studied the concentration of \(^{232}\text{Th}\) in bone using spectrophotometry and reported a detection limit of 100 ng. PLESKACH\(^7\) used the technique of neutron activation for the determination of these two radionuclides in urine, but used low neutron flux and the short-lived irradiation products, \(^{233}\text{Th}\) and \(^{239}\text{U}\) for their analysis, and reported a detection level of 100 ng and 600 ng, respectively, for Th and U nuclides. PICER and STROHAL\(^8\) also used a neutron activation method for the analysis of human tissue and body fluid samples, but the results obtained by them were higher than those reported in recent literature.\(^9,10\)

An IAEA interlaboratory comparison for the analysis of Th and U in bioassay samples showed that there were very large variations in results reported on the same sample by different laboratories,\(^11\) clearly demonstrating the inadequacy of the available methods for reliable analysis and underlining the urgent need for the development of sufficiently sensitive and reliable methods for the determination of these nuclides.

This paper reports on the development of a simple and sensitive method for Th and U determination, which employed neutron activation, post-irradiation chemical separation and counting of the activated products. This method was applied to determine the concentrations of the two radionuclides in food samples from high background areas to assess the daily dietary intake by the subjects living there.

### Materials and method

#### Sample preparation

The duplicate diet samples were collected from volunteers, living in high background area of India (Chavra in Kerala Sate) in precleaned polythene bags. The diet samples consisted of all the cooked and uncooked components of food, which an individual ate during 24 hours. These samples were homogenized in a mixer, freeze-dried and then powdered. About 200–250 mg of the powdered food sample was taken up for analysis.

The food sample along with the mixed standard of U and Th, prepared from standard uranium and thorium nitrate solutions, were sealed separately in polythene bags. These polythene bags were further sealed in aluminium foil before irradiation. Three standard reference materials: Orchard Leaves,\(^12\) Bovine Liver,\(^13\) and Animal Muscle (IAEA–H–4)\(^14\) were also analyzed to study the reliability of the method.