OBSERVATION OF SOME ANOMALIES IN THE ICRP METABOLIC MODEL OF URANIUM

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A simple and sensitive analytical method was developed for determining the concentrations of uranium (^{238}U) in biological and environmental samples. The minimum detection limit (M.D.L) of 0.08ng uranium (0.1 Bq of ^{238}U) in a sample was obtained with this method. Data on daily intake (through diet, drinking water and air), the excretion (through urine) and organ burdens of uranium were obtained for human subjects living in Bombay (India), and were employed to obtain important human metabolic parameters of U such as, gastrointestinal absorption factor(f_1), excretion ratio and biological half-lives of uranium retained in various human organs. The study showed that the f_1 factor for uranium available to humans at dietary level (0.5-5ug.d^{-1}) is 0.016, which is much lower than 0.2 suggested in ICRP 30. The biological half-lives, of uranium stored in kidney and muscles were estimated to be much longer than those proposed by ICRP.

INTRODUCTION

The large variations reported in the uranium concentration of similar human tissues and body fluids for subjects with comparable intakes of uranium, underline the need for the developement of sufficiently sensitive and accurate analytical method for uranium determination (Wrenn et al.1985, Dang 1993). The consistently lower concentrations of uranium in human tissues and body fluids reported by different workers in recent years (Fisenne et al., 1987, Igarashi et al., 1987, Wrenn etal., 1985, Dang et al., 1992, 1993, 1995) in comparison to the values reported in ICRP 30 (1979) also indicated that the ICRP metabolic model for uranium, may need revision.A program was initiated at Bhabha Atomic Research Centre (India) with the main objectives: 1) to develop a sensitive and reliable analytical method (with extremely low reagent blank) for the determination of uranium in biological and environmental samples, 2) employ the method to obtain the daily intake, excretion and organ burdens data on human subjects, 3) to use these data for evolving various important human metabolic parameters of uranium, 4) to compare the metabolic parameters thus obtained with those proposed by ICRP..

MATERIALS AND METHOD

The human tissue (obtained at autopsy), diet and body fluids (urine and blood serum) samples were obtained from population living in natural background area of urban India (Bombay). The samples of air and drinking water were also collected, for uranium analysis. While collecting samples, it was ensured that various samples were obtained from subjects with similar social background, food habits and that all the subjects were living in Bombay City for long duration of time.

The tissue, diet and blood serum samples were homogenised, freeze-dried and powdered before analysis. Urine and drinking water samples were first digested in nitric acid and, then the U present in the samples was co-precipitated with calcium phosphate. The procedure is described in detail by Dang et al (1992). The air samples obtained from various parts of the city were directly analysed along with the filter paper used for air sample collection.

The freeze dried samples and calcium phosphate precipitate were sealed and then irradiated for 24-36h in a nuclear reactor in neutron flux of = 10^{13} n cm^{-2} sec^{-1}. The neutron irradiation converted the uranium present in the samples to neptunium (^{238}U (n,)^{239}U-----^{239}Np). The uranium standard (200 ng of uranium nitrate solution dried on ashless filter paper) was also sealed and taken up for irradiation along with the samples. The irradiated samples containing ^{239}Np were digested in acid and then the ^{239}Np was chemically separated by two step chemical separation, using first DOWEX-1 (an anion exchange resin)and then its co-precipitation with barium sulphate precipitate. ^{237}Np was also used as tracer for the chemical yield determination. The gamma activity (227.5 keV and 278 keV gamma rays) of ^{239}Np was counted using 54 CC Hyperpure Germanium detector coupled to 4096 channel analyser. The uranium standard was also processed in the same way and counted, to compare the activities of sample and standard for the quantification of U. The details of analytical procedure are given elsewhere (Dang et al.,1992 Dang et al., 1993). The reliability of the analytical method was tested using the Standard Reference Materials.

RESULTS AND DISCUSSION

The total daily intake of uranium by adult Indian subjects are shown in Table 1. The daily urinary excretion levels of uranium are also included in the same Table.

Gastro-intestinal absorption factor(f_1)

The uranium entering the blood stream (transfer compartment) after absorption through the gastrointestinal tract, gets deposited in various systemic body organs such as, kidney, skeleton, muscles. Because of the relatively short biological half-lives of U deposited in various organs, as compared to the average age of the subjects (45Y), an equilibrium between the uranium entering and leaving the organs could be assumed. The U leaving the organs is excreted through the kidney into urine. Contribution of air was <2% of total intake. The f_1 factor was estimated to be 0.016 from the daily urinary excretion and intake of uranium.

Excretion Ratio

The excretion ratio is the fraction of uranium excreted daily in urine from the uranium present in the transfer compartment which is assumed to be the blood serum. Table 2 gives the uranium burdens for some of the body organs and total uranium in blood serum. The burdens were calculated from the concentrations of uranium determined in tissues obtained from these organs and average weight of the organs of an adult Indian. Employing the daily urinary excretion of 12.2 nanogram and the 30 ng of U in total serum pool, the excretion ratio was worked out to be 41%. Th excretion ratio is quite close to the clearance fraction of 54% proposed by ICRP. The excretion ratio studied in the present work could provide a guideline for an upper limit which may be used for screening the subjects for any impending kidney malfunction.

Biological half-lives of uranium retained in some organs

The organ burdens calculated on the basis of daily intake of uranium, the gut absorption factor obtained in the present study and the ICRP distribution factors (f_2) for these organs are also shown in Table 2 along with the measured values.

Table 1. The daily intake and urinary excretion (ug d^{-1}) of uranium for an adult Indian population group.

Source of intake	No. of samples analysed	Daily intake Range	Mean ± SD
Duplicate Diet	18	0.24 - 1.3	0.64 ± 0.4
Drinking Water	10	0.05 - 0.39	0.14 ± 0.1
Air	9	0.002 - 0.03	0.01 ± 0.1
Total Intake			0.79 ± 0.2
Daily Urinary Excretion ($ng.d^{-1}$)	28	4.0 - 52.0	12.3 x 2.2

Daily Intake of water 2.6L, and Air $20M^3$ Daily Urine vol. 1.3L
Daily Urinary excretion is based on Geometric mean (GM) conc. of U.

The calculated and measured values for skeleton are comparable, but for kidney and muscle, they are 4 and 70 times lower, indicating that the true biological half-lives for uranium retained in kidney and muscle may be much longer. As the concentration of U in muscle is extremely low,the longer biological half-life may not have significant effect on human health the longer biological half-life of uranium deposited in kidney (upto four times longer than that proposed in ICRP metabolic model) however, may warrant the reduction in the annual limit if soluble class (D&W) of uranium..

Table 2. Organ burdens of kidney, skeleton and muscles and blood serum of Indian subjects

Organ	Organ Weight (g)	Measured Uranium Burden (ug) Range	Geometric Mean (GM)	calculated
Kidney	230	0.02 - 0.58	0.13	0.03
Skeleton	7000	0.70 - 11.4	2.18	2.16
Muscle	21000	0.06 - 1.54	1.20	0.03
Blood Serum	2300	0.02 - 0.06	0.03	-

CONCLUSIONS

The f_1 factor 0.016 for uranium incoporated in diet, obtained in present sudy, is much lower than 0.2, as reported in ICRP 30. This study also showed that the biological half-life of uranium for kidney may be upto four times longer than that is proposed in ICRP model. The excretion ratio obtained in the present study may provide a useful guideline for screening the occupational workers handling uranium for any impending kidney damage.

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