COMPARISON OF DAILY URINARY EXCRETION OF THORIUM IN EXPOSED AND UNEXPOSED SUBJECTS WITH THEIR BIOKINETIC MODEL PREDICTION

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Abstract

In view of the increasing population pressure and malnutrition in spite of the enormous wealth from long year dependence on oil export in Nigeria, effort should be geared toward harnessing sustainable agricultural products. However, the safety of such products is a major issue of great environmental concern. Ionizing radiation from radionuclides in such products especially foodstuffs results in deleterious health effect in internal organs of the body. The estimation of daily intake of radionuclides and the amount getting to the systemic circulation are very crucial to the determination of internal dose to the various organs/tissues of the body. The extent to which human subject is exposed to thorium and the related risk can be deduced from its daily excretion. In this work, daily urinary thorium excretion of 19 adult subjects living and working in different locations have been measured in Nigeria using high resolution sector field inductively coupled plasma mass spectroscopic (HR-SF-ICP-MS) analytical method. The subjects included 12 adults occupationally exposed to thorium and its compounds and 7 adults living in normal areas without radionuclides exposure besides what is assumed as background values. The mean daily thorium excretion values are 15 ng and 4 ng for the exposed and un-exposed, respectively. These values were within the normal range of 3.4 ng and 34 ng obtained in literatures in spite of the high intake scenario considered. The predicted excretion rates using bioassay model of thorium given by the ICRP was also presented and compared with the measured data.

Keywords: urine, excretion, human subjects, thorium, model,

[1]Research activities conducted under the frame of the Alexander von Humboldt foundation (AVH) fellowship

Introduction

Thorium is a primeval radionuclide and it is ubiquitously occurring element with several radioactive isotopes. The average activity concentration of ^{232}Th is in the range 25 – 50 Bq kg^{-1} and can concentrate in certain rocks like granites and alkaline igneous rocks (UNSCEAR, 2000). Thorium, along with it progenies, emits six alpha particles and is therefore considered as one of the highly radiotoxic elements. The health hazards associated with these radionuclides stem from their ability to accumulate in human tissues, especially bone surface (ICRP, 1993). During the decay process, highly penetrating gamma rays are emitted, thereby causing intensive damage to the tissues where they are localized. The element (^{232}Th) has many industrial applications like the production of gas mantle, welding rods, thermisters, crucibles, alloying, etc. Improper handling of thorium materials can lead to occupational exposure and hence, radiation hazard. Apart from the occupational exposure, population in the vicinity of the numerous applications of thorium element and its compound can also be exposed. Environmental exposure to a population stems from constant daily inhalation and ingestion of thorium from natural, air borne particulates and dietary sources.

In order monitor and ensure the radiation protection of occupationally exposed person, it is important to have reliable information on the biokinetic behaviour of thorium in humans by considering the natural intake scenarios and the body content. Monitoring of occupational incorporation of thorium should preferably be carried out by analysis of its urinary excretion since the quantity lost per day via urine is related to the systemic body content (Höllriegl et al, 2002). However, for a reliable estimation of the occupational uptake of workers, baseline data of daily urinary excretion in subjects non-exposed occupationally is needed. In Nigeria, the activity of thorium from natural environmental sources in human bodies and its excretion of both exposed and unexposed subjects are yet to be studied. In fact, studies on urinary excretion of these radionuclides from Africa are not available.

The International Commission on Radiological Protection (ICRP) provides guidelines to assess the exposure to thorium using the daily urinary excretion data (ICRP, 1997). In response to this, daily urinary thorium excretion data are now available from studies conducted in many countries (Höllriegl et al, 2005a&b; Roth et al, 2005). However, data on daily urinary excretion of thorium are not available in Nigeria, and to the best of are knowledge, little or no data are available from African countries for comparison. This work is a pioneering effort in Nigeria, especially through the ingested pathway in the determination of internal contribution to the overall radiation body burden. In this study, the concentration of thorium in urine samples of unexposed and exposed subjects was quantified and its daily urinary excretion determined. The daily urinary excretion values were compared with the biokinetic model prediction of thorium using the dietary intake values for the same population recently reported (Arogunjo et al, 2009). The excretion values were also compared with values from literature.

MATERIALS AND METHODS

Sample collection

Twenty-four hour urine samples were collected from four different groups of subjects, which include six mine workers in Bisichi mining site, five processing workers from tin processing company in Jos, four members of the public living in the city of Jos and four members of the public in Akure about 900 km away from Jos Plateau as control. The occupational (mining and tin processing workers) groups have age range of 24 – 52y including a subject in Akure who by virtue of his job might have been occupationally exposed to radionuclides. The public group around the mining and processing sites but are not exposed occupationally has age range of 34 – 44y. The public group used as control has age range of 37 – 40y that are not exposed to any artificially higher levels of thorium and its compounds. The 24-h urine samples were collected from both the occupational group and the public group around the mining and processing sites between 14th and 15th September 2006. The public control group 24-h urine samples were collected between 22nd and 23rd September 2006. The 24-h urine was collected starting early in the morning. After wake-up, the subjects emptied their bladder in the toilet noting the time, and all urine thereafter was collected in a graduated 3000 ml pre-cleaned polyethylene container until the following morning, and for the last time at the exact time the bladder was emptied the previous morning. The first void collected at the start of the sampling was acidified with 0.5 ml HCL to prevent decomposition. Thirty (30) ml aliquots of the total urine collected from each subject was put into a plastic vial, which was placed inside a plastic cylinder and stored at 4 ^{0}C until analysis.

Sample Preparation and measurement

All the samples were measured at the Central Analytical Service of GSF, using high resolution sector field ICP-MS Model ELEMENT 1 (Finnigan MAT, Germany). The instrument parameters and the method applied have been described elsewhere (). Prior to measurement, all the urine samples were removed from the storage site and allowed to defrost at room temperature. The acidified samples were diluted into ratio 1:2 by the addition of 0.25 ml of concentrated HNO_3, 0.5 ml of concentrated HCL and 4.5 ml of H_2O to 5 ml of the sample. Thorium standards were used to calibrate the instrument for its direct measurement and reagent blanks using deionised water were also measured at intervals during the entire measurement process.

Biokinetic modelling of the radionuclide

Radionuclides transport in the human body can be investigated using deterministic model. This process involves model simulation of the linear transfer processes represented by sets of linear differential equations governed by first order kinetics. In order to be able to compare the measured urinary excretion rates with that predicted by the ICRP biokinetic models for thorium and uranium, expected excretion rates through lifetime were simulated using the age dependent biokinetic transfer coefficients for the six age groups given by the ICRP Publication 69 (ICRP 1995). For the purpose of simulating the behaviour of the radionuclides between compartments after ingestion, the systemic model was coupled to the gastrointestinal (GI) tract model. The ICRP age-dependent transfer rates in the GI tract and the transfer rate from the small intestine to blood was calculated according to ICRP (1995). According to the dietary intake values for the same adult population recently reported, the annual ^{232}Th intakes of 6.1 mg y^{-1} (24.7 Bq y^{-1}) was obtained for the unexposed population (Arogunjo et al, 2009) in Nigeria. This value was added to the intake value for milk and meat given by UNSCEAR (2000), which were not included in the study to represent the adult population. The resultant value (45.0 µg d^{-1}) was age-adjusted according to the respective food consumption rates ratio for the different age groups namely: infant, child and adult given by UNSCEAR (2000) to 16.0 µg d^{-1}, 29.0 µg d^{-1} and 45.0 µg d^{-1}, respectively. The age adjusted intake values for the exposed population are 24.5 µg d^{-1}, 29.0 µg d^{-1} and 71.9 µg d^{-1}, respectively. In modelling the lifetime excretion rates, the biokinetic transfer coefficients governing the distribution and retention of thorium in the various compartments of the systemic and the GI tract models during the integral time course were performed using age-dependent linear interpolation. The distribution and retention of the radionuclide in the various compartments is governed by linear transfer processes represented by sets of linear differential equations. The transfer between the various compartments therefore, follows a system of first-order kinetics. To solve these sets of linear differential equations, different software packages are available for solving multi-compartmental systems and one of such packages is the SAAM II computer program. The SAAM II software package version 1.2.1 was used to perform the biokinetic modelling.

RESULT AND DISCUSSION

Baseline data of the daily thorium excretion of un-exposed subjects is very crucial to the overall emergency response in case of gross contamination and the assessment of subjects exposed occupationally. In order to determining the extent of radiation health hazards to the exposed and non-exposed population, the results of the daily urinary thorium excretions measured in 37 samples including their ages and weights are presented in Table 1. The Table included two groups of subjects namely the exposed and the un-exposed groups. The range of excretion values along with the mean (±SD), median (95% confidence interval), geometric mean (±GSD) for both the exposed and the un-exposed groups were also shown in Table 1. Thorium daily urinary excretion varies from 1.2 ng to 41.8 ng in the exposed group and from 0.78 ng to 8.9 ng in the un-exposed group.

In the test for significance between the urinary excretion of thorium for the exposed and the unexposed group, statistical test of the Mann-Whitney U-test was employed. The results show that the exposed group excrete more thorium ($p < 0.01$) than the unexposed group (Z: 2.897, $p = 0.00377$). This result is expected in view of the risk of exposure associated with the daily routine work in an elevated activity area. The mine workers are not only exposed via ingestion through food and water, but also via inhalation and dermal contact at workplace.

In spite of the notable differences in the excretion of thorium between the exposed and the unexposed groups considered in this work, the data still fall within the normal range obtained in literatures. Figures 1 show the mean urinary thorium excretions in unexposed populations from different studies. The figure suggested that the daily urinary thorium range between 1.5 ng and 15.5 ng although the authors used different statistical parameters and units. The values were converted to the same units by assuming daily urinary volume of 1.4 l d^{-1} proposed by the International Commission on Radiological Protection (ICRP) for adult male and female (ICRP, 2003).

Comparison of urinary thorium excretion with its model data

The expected daily urinary excretion of thorium was determined for adult male using two different intake scenarios for the unexposed and the exposed groups in Nigeria by applying the current ICRP biokinetic model discussed earlier. The urinary excretion rate for thorium during lifetime was simulated using the age dependence intake given earlier for the unexposed population as shown in Figure 2. The excretion rates for the exposed group were also calculated using the intake values given earlier for this group. The default ICRP f_1 values of 5 x 10^{-4} for thorium, was initially used for the calculations as presented in the Figure. The figure also included all the individual excretion values plotted to show the large variability in the measured data and the discrepancy with the model prediction. In the figure, the model prediction was obtained using the default f_1 value for thorium and by applying the unexposed intake (long dash line) and the exposed intake (solid line). The model predictions fall in the range of the measured data, however, it overestimated the excretion rate value at the mean age (38 y) for the unexposed data set by more than 100 % and that of the exposed data set at the mean age (31 y) by about 24 % when compared with the geometric mean values plotted at these ages. This result clearly disagrees with the report of similar study conducted with German subjects, which indicated an underestimation of urinary thorium excretion using the default f_1 value. The default value multiplied by a factor of 10 was then proposed to fit their data (Roth et al, 2005). The general observation from the results of the present work and that of the study reported for German subjects, although the subjects are from different geological, ethnic and environmental backgrounds, is that the disagreement between the measured data and the model predictions could be traced to the assumption of a default f_1 values for all unspecified compounds by the ICRP (ICRP, 1997). The need to specified f_1 value for all dietary incorporated radionuclides can be clearly seen in view of the present discrepancies in the application of the bioassay models. The f_1 value should also take care of situation with high intake scenarios, which is currently lacking as observed in the present study.

In view of the above, it then suggest a question as to whether the f_1 value proposed by the ICRP should be applied uniquely in all situation especially when using the bioassay model as a monitoring tool in an emergency response programme. In order to fit the model to the median values of the measured urinary thorium excretion data at the mean ages for the two groups of subjects considered in this work, new f_1 of 4.0×10^{-4} was proposed for both the unexposed (dash-dot-dot line) and the exposed groups (dash-dot line) as shown in the figure.

Conclusion

The urinary thorium excretion rates have been calculated for the exposed and the unexposed populations in Nigeria using ICP-MS analytical method. The predicted excretion rates using bioassay model of thorium and uranium given by the ICRP (1997) were also presented and compared with the measured data. The results show that the median values for the exposed and un-exposed groups are 10.8 (1.0) ng d^{-1} and 4.0 (1.3) ng d^{-1} for thorium daily urinary excretion, respectively. The predicted urinary excretion rates simulated using the default ICRP f_1 value at the mean ages of 31y and 38 y for the daily thorium excretion are 11.48 ng d^{-1} and 6.10 ng d^{-1} for the exposed and the unexposed population, respectively. The difference between the predicted excretion values and the measured values suggested the need for the reconsideration of the use of the default ICRP f_1 values for the dietary incorporation of the radionuclide for different intake scenarios. The application of a new f_1 value proposed in this work for thorium however predicted the daily excretion values of 9.18 ng and 4.88 ng for the exposed and unexposed group, respectively.

Acknowledgements

The author recognised the contributions of Dr A. Giussani, Dr U. Oeh, Dr V. Höllriegl, and Dr W.B. Li of Helmholtz-Zentrum Munchen, German Research Center for Environmental Health, Institute of Radiation Protection, Ingolstadter Landstr. 1, 85764 Neuherberg, Germany.

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Table 1: Daily urinary excretion of ^{232}Th in unexposed and exposed adult subjects in Nigeria

Subject	Age	Weight (kg)	U excretion values (ng d^{-1})
Unexposed Group			
PAC1	39	91	0.78; 4.28
PAC2	40	71	1.45; 7.61
PAC3	37	67	0.84; 4.37
PJS1	34	70	3.77; 8.19
PJS2	37	91	0.99; 5.29
PJS3	38	84	2.64; 8.94
PJS4	44	70	1.35; 7.28
Number of samples			14
Range			0.78 – 8.94
Mean (SD)			4.13 (2.94)
Median (95 % confidence interval)			4.03 (1.26)
Geometric mean (GSD)			3.01 (2.44)
Exposed Group			
TPJ1	27	70	1.95; 27.10
TPJ2	26	70	21.45; 33.93
TPJ3	36	64	1.62; 15.18
TPJ4	26	88	8.58; 11.08
TPJ5	24	72	33.54
TMB1	30	58	41.78; 41.70
TMB2	34	66	4.32; 4.83
TMB3	35	58	1.20; 6.65
TMB4	26	64	2.03; 15.37
TMB5	28	60	10.76; 7.00
TMB6	32	70	1.45; 8.28
PAC4	52	84	23.98; 32.12
Number of samples			23
Range			1.20 – 41.78
Mean (SD)			15.47 (13.57)
Median (95 % confidence interval)			10.76 (1.00)
Geometric mean (GSD)			9.30 (3.00)

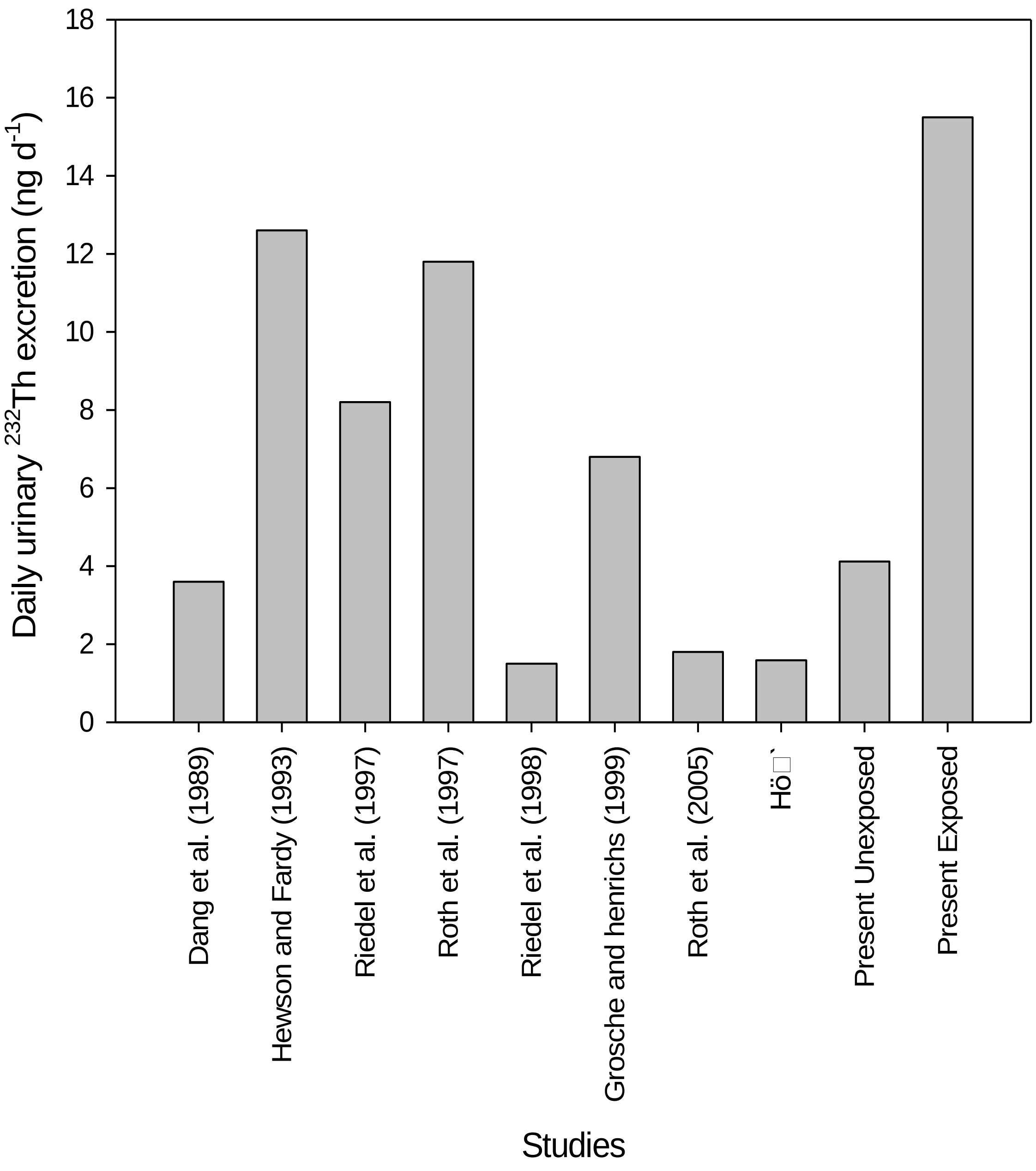

Fig 1: Comparison between the present and other studies on the daily urinary ^{232}Th excretion

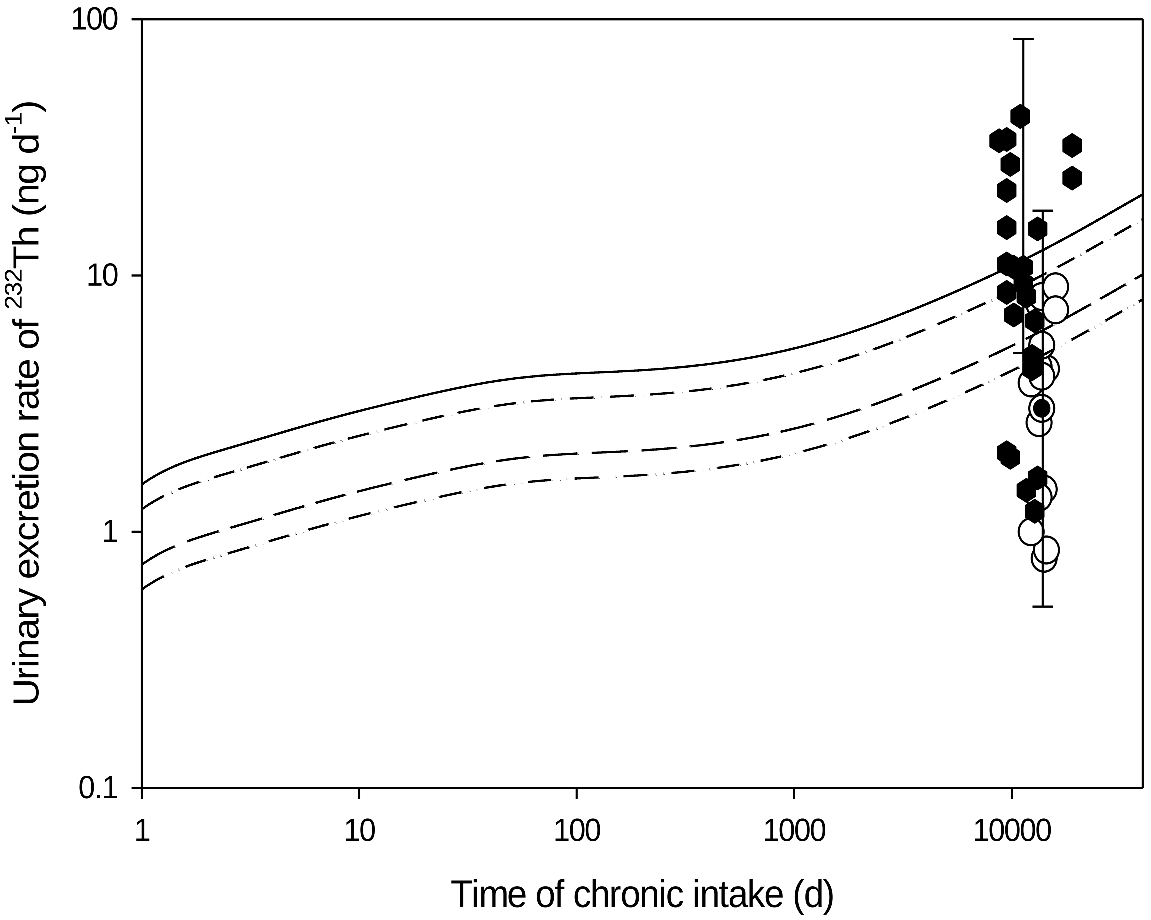

Fig. 2: Predicted urinary thorium excretion during lifetime and the measured excretion values. Solid line: ICRP biokinetic model (ICRP 1995) using the default f_1 value and thorium intake for the exposed; Long dash line: Model prediction using the default f_1 value and daily intake for the unexposed; Dash dot line: Model prediction using the modified f_1 value and daily intake for the exposed; Dash dot-dot line: Model prediction using the modified f_1 value and daily intake for the unexposed. Open circle symbol represent individual excretion values for the unexposed group; Closed Hex symbol represent individual excretion values for the exposed group. Error bars are the upper and lower bound at 95 % confidence interval of the GSD for the two groups.