Implication of the default absorption factor in the determination of the internal dose from the dietary intake of uranium in Nigerian foodstuffs

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Abstract

Daily urinary excretion of uranium for 12 adults occupationally exposed to uranium and 7 adults living in normal areas without radionuclides exposure besides what is assumed as background values in Nigeria has been determined using high resolution sector field inductively coupled plasma mass spectroscopic (HR-SF-ICP-MS) analytical method. The mean daily urinary uranium excretion values for the unexposed and exposed population for infant, child and adult subjects are 3.0 µg d^{-1}, 5.0 µg d^{-1} and 8.0 µg d^{-1}, and 5.3 µg d^{-1}, 11.3 µg d^{-1} and 16.7 µg d^{-1} respectively. The predicted excretion rates using biokinetic model of uranium given by the International Commission for Radiation Protection (ICRP) were also presented and compared with the measured data. Large discrepancy was observed between the measured data and the model predictions using the default ICRP f_1 values for uranium, which suggested the need for the use of an appropriated f_1 values to fit the measured data.

Keywords: urine, excretion, human subjects, uranium, model, absorption factor

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Introduction

Biotechnology development and the threat posed by the global climate change are twin issues that will either impact positively or negatively on the global food security. The quality and quantity of food products will either be enhanced or diminished. In the discussion of the quality of food products, food safety is an important parameter. In view of the various methods involve in biotechnology development, especially the use of ionizing radiations in gene mutation and increasing the chef life of foodstuffs. Radiological impact assessment of foodstuffs will be an important safety criterion. Food gets to the systemic circulation through the small intestine and the fraction of radionuclide like uranium that gets to the blood is the absorption factor for the radionuclide. Uranium is a primeval radionuclide and it is ubiquitously occurring element with several radioactive isotopes. It is the heaviest naturally occurring element and its radiotoxicity and chemical toxicity have been described extensively in the literature (UNSCEAR, 2000), with average activity concentration in the range of 25 – 50 Bq kg^{-1}. However, the activity concentration may vary considerably from one location to the other depending on the local underlying geology.

During decay process, highly penetrating gamma rays are emitted, thereby causing intensive damage to the tissues where they are localized. Improper handling of materials containing uranium can lead to occupational exposure and hence, radiation hazard. Apart from the occupational exposure, population in the vicinity of the numerous applications of uranium element and its compound can also be exposed. Environmental exposure to a population stems from constant daily inhalation and ingestion of the radionuclide from natural, air borne particulates and dietary sources.

Body burden of uranium in any population can be enhanced by human activities resulting in the alteration of the natural constituents of radionuclides in the soil. Mining and milling of ores all over the world have contributed immensely to this observed disequilibrium and the enhanced naturally occurring radioactive materials (Dowdall et al, 2004) and their subsequent processing can lead to the exposure of personnel from external radiation and from intake (UNSCEAR, 2000). This affects the terrestrial ecosystem due to the excavation of large amount of sand and the eventual accumulation of large volume of tailings. Tin mining in Nigeria resulted in the removal of considerable amounts of generated waste as tailings from the mining sites to neighbouring vicinities.

In tin mining industries, the main technologically enhanced naturally occurring radioactive materials (TENORM) are the Uranium and Thorium series and their progenies, together with ^{40}K (Dowdall et al, 2004).

In Nigeria, tin mining and processing lead to the accumulation of tailings with significant amounts of natural radionuclides like ^{238}U. Tin mining activities started in the Jos Plateau some decades ago around 1904 shortly after airborne radiometric mapping revealed high deposit of cassiterite and columbite (niobium) ores. Monazite sands and zircons were later discovered as accessory minerals, which have been known to contain very high amount of thorium (UNSCEAR, 2000). It follows from this that foodstuffs like cereals, tubers, vegetables will contain traces of ^{238}U owing to the radionuclides transport processes in the soil and underground water. Elevated activity of radionuclides in foodstuffs and soil have been reported in the tin mining sites of Jos Plateau (Jibiri et al, 2007) and no efforts have been made to determining the extent of internal health hazards to the inhabitants of the area and even those that are occupationally exposed.

In order monitor and ensure the radiation protection of occupationally exposed person, it is important to have reliable information on the biokinetic behaviour of radionuclide in humans by considering the natural intake scenarios and the body content. Monitoring of occupational incorporation of radionuclide 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. However, for a reliable estimation of the occupational uptake of workers, baseline data of daily urinary excretion in subjects non-exposed occupationally is needed. Furthermore, the knowledge of the ingestion rate of radionuclide with the application of an appropriate absorption (f_1) factor will enhance realistic assessment of the internal dose. In Nigeria, the activity of radionuclides 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 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 radionuclides 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 uranium 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 uranium 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 uranium 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). 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. In the case of uranium, its natural standard solution was used to prepare the calibrating solution and the samples were measured directly by using 100 µg l^{-1} of internal standard solution of ^{193}Ir to correct for matrix interference.

Biokinetic modelling of Uranium

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 ^{238}U intakes of 1.9 mg y^{-1} (23.2 Bq y^{-1} fresh weight) 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 (8.0 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 3.0 µg d^{-1}, 5.0 µg d^{-1} and 8.0 µg d^{-1}, respectively. The age adjusted intake values for the exposed population are 5.3 µg d^{-1}, 11.3 µg d^{-1} and 16.7 µg d^{-1}, respectively. In modelling the lifetime excretion rates, the biokinetic transfer coefficients governing the distribution and retention of uranium 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

Urinary uranium excretion

Baseline data of the daily uranium excretion of human 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 in the population, the results of the daily urinary uranium excretions measured in 52 samples obtained at different times from 19 healthy adult males subjects in Jos, Akure and Bisichi including their ages and weights are presented in Table 1. The range of excretion values along with the mean (±SD), median (95% confidence interval), and geometric mean (±GSD) for the subjects were also shown in Table 1. Large intra-individual variations can be seen in the data provided in the table. These are in agreements with similar studies conducted elsewhere (Paul Roth et al, 2005). It is clear from the frequency distribution of the daily urinary excretion for the radionuclide, the arithmetic mean values of the excretion, and the associated standard deviation that the excretion does not seem to follow a Gaussian distribution pattern. In view of the above, the data could best be represented by the median value provided along with the 95% confidence interval. Furthermore, Figure 1 suggested that the data is log-normally distributed and could best be represented by the geometric mean provided along with its geometric standard deviation calculated using a log-normal distribution function.

Comparison of urinary thorium and uranium excretion with literature values

The present urinary uranium excretion data fall within the normal range obtained in literatures. Figure 2 shows the urinary uranium excretion in an unexposed population from different studies. The figure suggested that the daily urinary uranium excretion range between 3.4 ng and 34 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 uranium excretion with its model data

The expected daily urinary excretion of uranium was determined for adult male using the two intake scenarios and the ICRP biokinetic model discussed earlier. The urinary excretion rates for uranium during lifetime were simulated for the unexposed and exposed groups using the age dependence intake given earlier for the Nigerian population and the result is as shown in Figures 3. The default ICRP f_1 value of 2.0×10^{-2} for uranium was initially used for all the calculations as presented in the Figure. The Figures 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 model prediction using the f_1 value of 0.02 % and the intake scenario for the unexposed population, overestimation of the excretion using the default f_1 value can be seen and this may lead to gross underestimation of intake when using the model for monitoring purposes. 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 by the ICRP (ICRP, 1995). The need to specified f_1 value for the dietary incorporated radionuclides in the population can be clearly seen in view of the present discrepancies in the application of the default ICRP value. The f_1 value should also take care of situation with high intake scenarios, which is currently lacking as observed in the present study especially in the case of the dietary incorporated uranium. The form in which the foodstuffs are consumed must be taken into account in the assessment of an appropriate f_1 value, the diet constituents of the population are mostly solid unprocessed root tubers, which contribute about 70% of the local foodstuffs. In this form, the radionuclides may be bio-available in the gastrointestinal environment (GIE) but not bio-accessible for absorption from the GIE apparently due to some rebounding processes. Moreover, bio-accessibility may be hampered due to some additives, fat from palm oil, which is majorly used in the cooking process of most foodstuffs can be a contributing factor. Furthermore, the intake values used in this study may not represent the actual intake since the value was obtained from raw foodstuffs (Arogunjo et al, 2009). In Nigeria, foodstuffs usually go through the process of cooking before they are consumed. Uranium is soluble in water by forming uranyl complexes; this can result in the radionuclide going into solution and its eventual removal from foodstuffs during the cooking process.

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 uranium excretion data at the mean ages for the two groups of subjects considered in this work, new f_1 of 0.07 % was proposed for the unexposed (dash-dot-dot line) and the exposed groups (dash-dot line) as shown in Figure 3.

Conclusion

The urinary uranium 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 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 median for the measured urinary uranium excretion values are 22.2 (0.7) ng d^{-1} and 7.2 (0.9) ng d^{-1} for the exposed and un-exposed, respectively. The urinary excretion rates simulation during lifetime by assuming the default ICRP f_1 values and the age dependence intake for infant, child and adult of 3.0 µg d^{-1}, 5.0 µg d^{-1} and 8.0 µg d^{-1}, respectively predicted an excretion of 142 ng d^{-1} at the mean ages for the unexposed groups. The result shows wide difference between the measured data and the model prediction. These discrepancies suggested the need for the reconsideration of the use of the default ICRP f_1 values for the dietary incorporation of the radionuclide. The model prediction at the mean ages of 31y and 38 y using a new absorption factor for the daily uranium excretion are 22.16 ng and 4.98 ng for the exposed and the unexposed population, respectively.

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Table 1: Daily urinary excretion of ^{238}U in unexposed and exposed adult subjects in Nigeria

Subject	Age	Weight (kg)	U excretion values (ng d^{-1})
Unexposed Group			
PAC1	39	91	2.96; 3.94; 3.86
PAC2	40	71	8.08; 7.28; 7.19
PAC3	37	67	10.58; 15.29; 7.22
PJS1	34	70	6.06; 7.28; 7.19
PJS2	37	91	0.99; 4.95; 4.90
PJS3	38	84	19.37; 10.44; 7.28
PJS4	44	70	1.35; 6.74; 6.68
Number of samples			21
Range			0.99 – 19.37
Mean (SD)			7.13 (4.23)
Median (95 % confidence interval)			7.19 (0.86)
Geometric mean (GSD)			5.90 (2.00)
Exposed Group			
TPJ1	27	70	25.35; 32.57; 12.38
TPJ2	26	70	17.63; 30.97; 15.21
TPJ3	36	64	21.71; 31.10; 22.19
TPJ4	26	88	22.23; 28.39; 20.90
TPJ5	24	72	34.91
TMB1	30	58	62.70; 89.25; 59.63
TMB2	34	66	10.01; 35.70; 12.88
TMB3	35	58	3.89; 9.17; 7.64
TMB4	26	64	10.23; 13.17; 10.07
TMB5	28	60	30.75; 32.00; 18.25
TMB6	32	70	26.83; 33.06; 12.98
PAC4	52	84	7.13; 10.98; 10.91
Number of samples			34
Range			3.89 – 89.25
Mean (SD)			24.20 (17.78)
Median (95 % confidence interval)			22.19 (0.67)
Geometric mean (GSD)			19.38 (1.97)

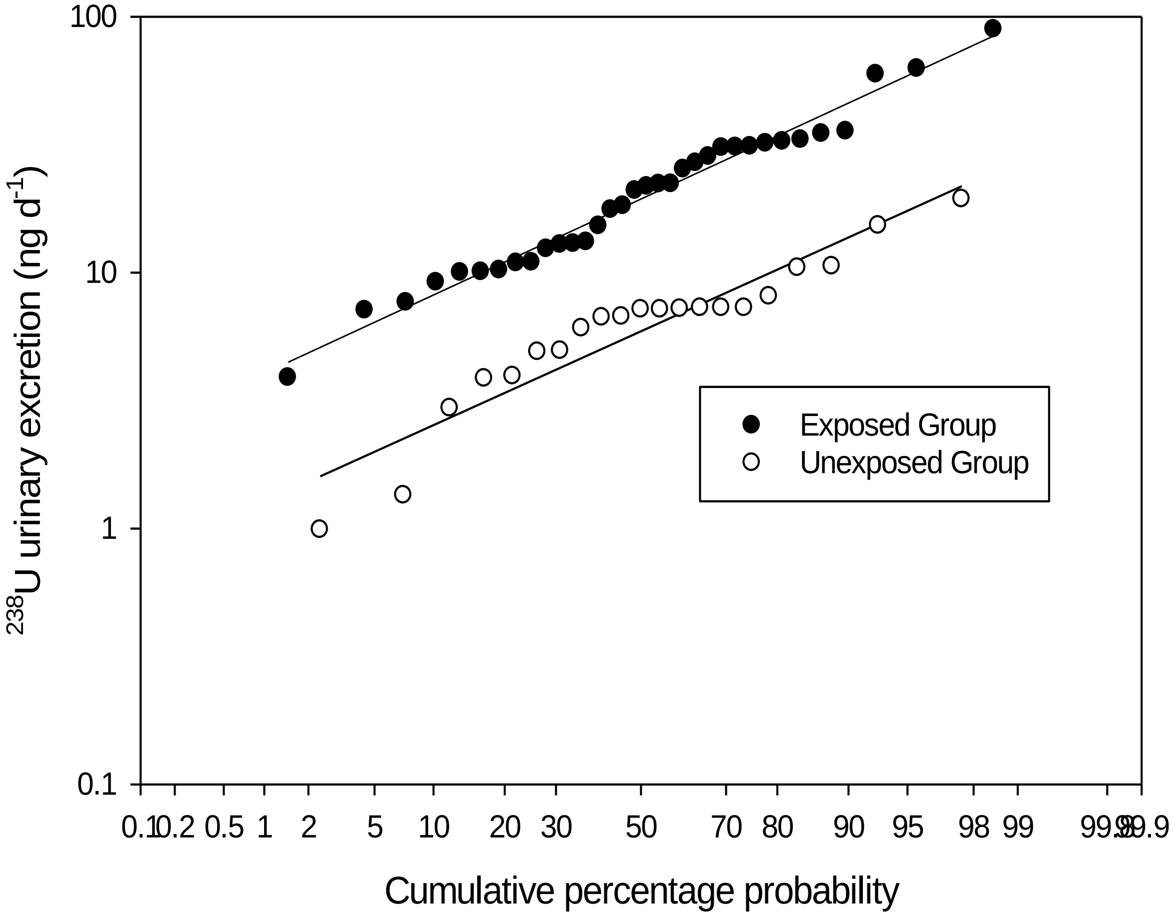

Fig 1: The log-probability plot of urinary ^{238}U excretion rate and the fitted curve.

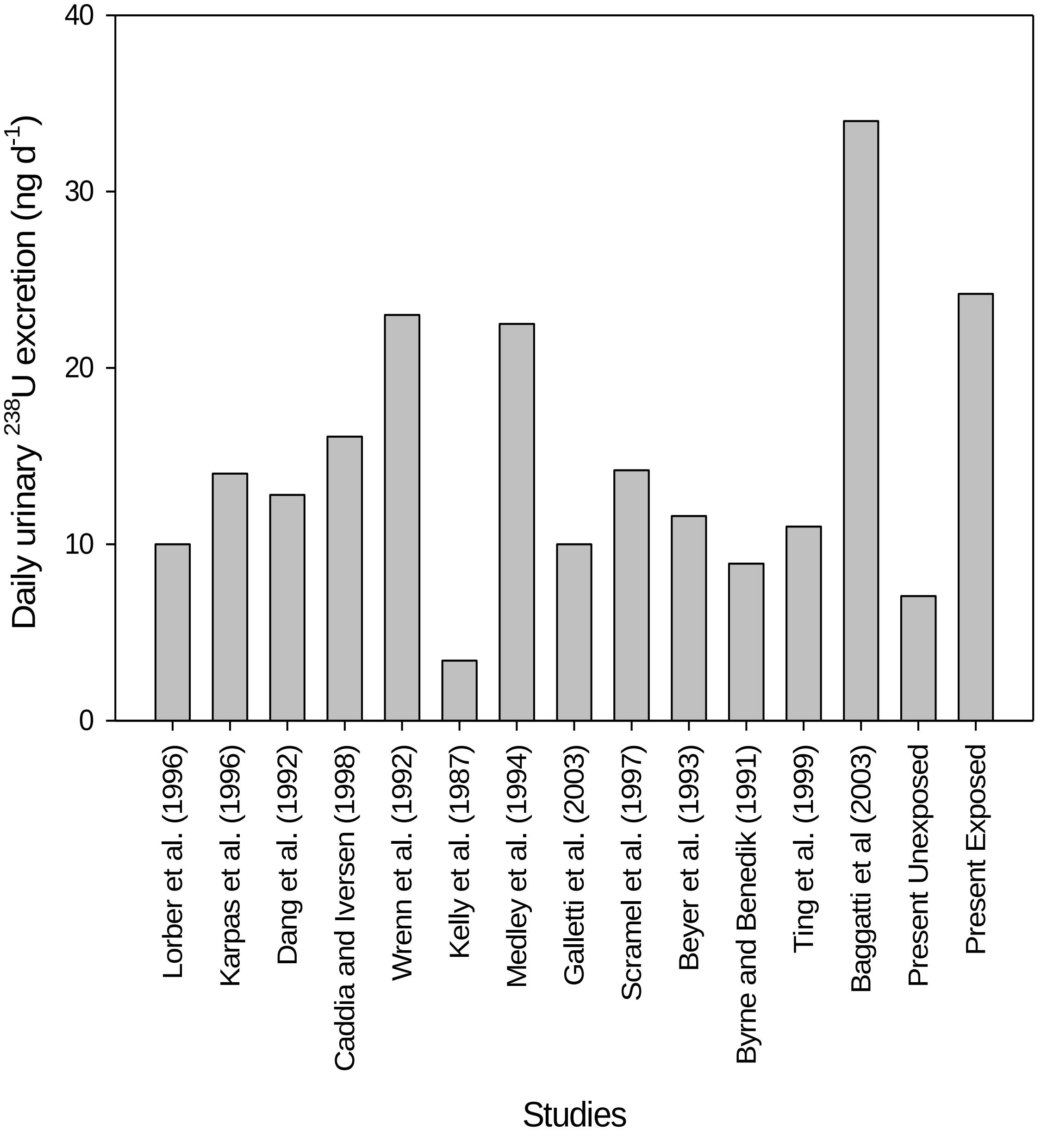

Fig 2: Comparison between the present and other studies on the daily urinary ^{238}U excretion

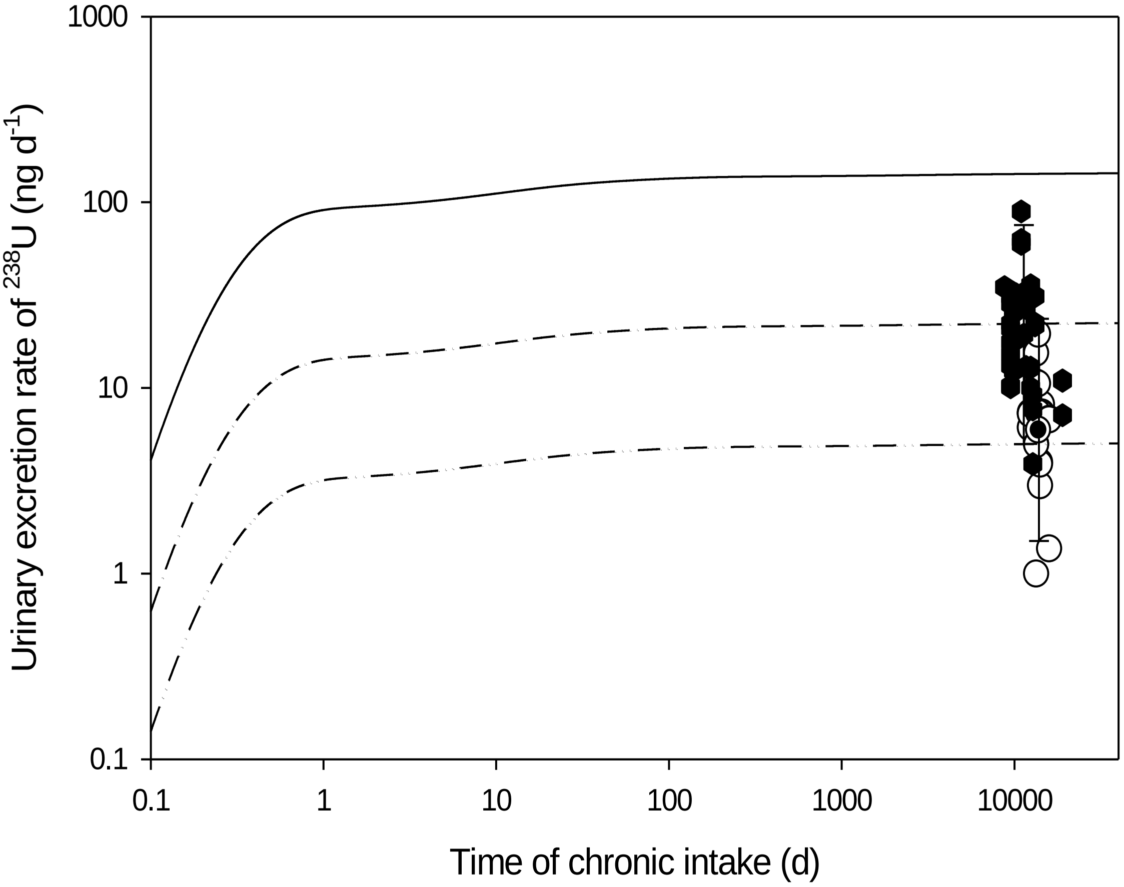

Fig. 3: Predicted urinary uranium excretion during lifetime and the measured excretion values. Solid line: ICRP biokinetic model (ICRP 1995) using the default f_1 value and uranium intake for the exposed; Dash dot line: Model prediction using the modified f_1 value and daily intake for the exposed; Dash 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.