Study of thorium uptake by inhabitants of a high background radiation area

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Abstract. Buena, located in the North of Rio de Janeiro, is characterized by its high natural radiation background, due to large deposits of monazite sand. The foodstuffs consumed by the population are basically composed of local products, which contain significant amounts of thorium. The analysis of complete cooked meals have shown an average daily intake of 18 $mBq.d^{-1}$ of ^{232}Th and 189 $mBq.d^{-1}$ of ^{228}Th. The average urine to feces ratio of ^{232}Th from samples of volunteers was found equal to 7.5×10^{-2}. The comparison of the experimental data with the predicted urine to feces ratios derived using the biokinetic model for thorium described by the ICRP publication 69 and simulating inhalation and ingestion separately, lead to the conclusion that the thorium intake is a combination of inhalation and ingestion. The clearance rate of thorium of monazite in lungs has apparently behaved as Type M compound. Inhalation is the biggest contributor for the committed effective dose due to thorium internal exposure.

INTRODUCTION

In Brazil, there are some areas where the concentrations of uranium and thorium in the soil are elevated. The inhabitants of theses areas are chronically exposed to radionuclides from the series of thorium and uranium. Most of the data available in the literature are based on the internal dose estimates through daily diet intake instead of bioassay data. In this study, Buena, a town located in North of Rio de Janeiro State in Brazil, where there is a large deposit of monazite sand, was selected for bioassay data collection and estimation of effective doses due to thorium natural exposure. An important characteristic of the families living in this area is the low mobility. Most of them have been in town for more than 10 years. The food consumed by the population is basically composed of local products. The quantification of the activities present in the diet from the village of Buena and from the city of Rio de Janeiro has been reported by Lauria et al. [1, 2]. The authors have found a significant concentration of thorium, uranium and nuclides from their radioactive series in the cooked meals. The average values of the daily diet intake of some radionuclides found in the cooked meals from Buena and from Rio de Janeiro are shown in Table I. These data characterize a chronic ingestion of foodstuffs with high concentrations of natural radionuclides. As this study is still being conducted, the data related to the control area are not available yet and Rio de Janeiro has been used as a reference of low background radiation area. Radium is the larger contributor to the intake, but this investigation will be focused on thorium.

METHODOLOGY

A total of 17 healthy adult volunteers inhabitants from Buena were selected and monitored by bioassay techniques. The individuals were asked to collect 24 hours fecal and urinary excretion. As thorium in monazite is an insoluble compound, only traces of thorium were expected in urine. The urine samples were analyzed for thorium content by inductively coupled plasma atomic mass spectrometry (ICP-MS), which is suitable for measurement of small amounts of thorium. The thorium in feces samples were high enough to be detected by alpha spectrometry method, which provided the isotopic ratio of these two radionuclides and allowed the comparison of ^{232}Th and ^{228}Th excreted in feces with the daily intake in diet.

The interpretation of the excreta results was done using a computer code, developed in our laboratory, to derive the expected fractions of thorium excreted in urine and in feces due to chronic intakes of the nuclide, through ingestion and inhalation. The thorium biokinetic model described in ICRP publication 69 [3] was used, but the value of the gastrointestinal uptake factor f_1 was changed several times, to test different proposals existing in the literature [4, 5]. The computer code reproduces the thorium predicted values of body contents and daily urinary and faecal excretion as a function of time, published in publication 78 of the ICRP [6], following the intake patterns and the parameters described in the publication. For the dose calculation, the dose coefficients from the ICRP database of dose coefficients for workers and members of the public [7] was utilized.

Table I. Average daily intake of radionuclides in the diet (cooked meals) from Buena and Rio de Janeiro ($mBq.d^{-1}$)

	^{226}Ra	^{228}Ra	^{210}Pb	^{238}U	^{234}U	^{232}Th	^{230}Th	^{228}Th
				Buena				
Average	246	722	216	108	111	18	32	189
				Rio de Janeiro				
Average	179	456	89	36	35	8	15	146

RESULTS AND DISCUSSION

The ^{232}Th daily urinary and fecal excretion of a group of a group of 20 adults are shown in Table II. The ^{232}Th fecal excretion varied from below to 4 to 82 $mBq.d^{-1}$, with an average value of 23 $mBq.d^{-1}$. The average value for ^{228}Th in feces was 160 $mBq.d^{-1}$. The comparison of the average value of ^{232}Th excreted in feces with the amount of thorium present in the diet shown in Table I, suggests that the fecal excretion reflects the daily intake through diet. The thorium urinary excretion varied from 1.2 to 2.9 $mBq.d^{-1}$, with an average value of 1.9 $mBq.d^{-1}$. The variation of ^{232}Th in feces was not correlated to ^{232}Th in daily urinary excretion. Consequently, the urine to feces ratio varied widely from 0.03 to 0.23 and it was influenced by the amount of ^{232}Th in feces, the average ratio was 0.12. While the average values of ^{232}Th found in the daily urinary and fecal excretion from inhabitants of Rio de Janeiro were 0.7 and 9 $mBq.d^{-1}$, respectively, the urine to feces ratio was 0.072 [8]. The average amount of ^{232}Th in urine samples from inhabitants of Buena was almost three times higher than in urine samples from inhabitants of Rio de Janeiro, characterizing a chronic intake.

In order to investigate the main pathways of thorium intake and interpret the excreta data it was necessary to test some hypotheses and compare with the experimental data. As the amount of ^{232}Th and ^{228}Th in feces samples were similar to quantities present in the daily diet consumed by the population, the first hypotheses assumed that ingestion was the only pathway of thorium intake. The predicted values of daily urinary and fecal excretion were derived using the thorium biokinetic model described in ICRP publication 69 [3], simulating a chronic intake of thorium by ingestion, considering a gastrointestinal uptake f_1 equal to 5×10^{-4}, as suggested by ICRP publication 69 [3] for members of the public. The predicted urine to feces ratio was 1.2×10^{-4}, which is smaller than the experimental one, 1.2×10^{-1}. As a large variety of values of gastrointestinal uptake (f_1) of thorium has been reported in the literature, for insoluble thorium compounds, like thorium compound found in monazite, other values for f_1 were tested. The f_1 value equal to 1×10^{-3} for thorium in food recommended by NEA/OECD [4] by analogy with plutonium ingested in the tetravalent state as recommended in Publication 48 [9] was used. The predicted urine to feces ratio was still low, 2.5×10^{-4}, compared to the experimental one. As Johnson and Lamonte [5] have reported f_1 values in a

range of 1×10^{-3} to 1×10^{-2} as suitable values for dietary intake of thorium, the highest f_1 value, 1×10^{-2}, was tested, but the predicted ratio 2.5×10^{-3} was still too small in comparison to the experimental values.

Table II. Thorium in 24 hours excretion of the adult people from Buena

#	Age (y)	^{232}Th urine ($mBqd^{-1}$)	^{232}Th feces ($mBqd^{-1}$)	^{228}Th feces ($mBqd^{-1}$)	^{232}Th U:F ratio
TMOP	19	1.56	7.94	86.75	0.20
ACV	26	1.26	-	-	-
ROGS	32	1.18	37.08	257.50	0.03
FMOP	32	2.99	-	-	-
SVR	33	1.34	6.35	63.48	0.21
ARC	35	2.44	11.72	72.48	0.21
SR	37	2.44	-	-	-
GMS	37	2.73	14.07	338.23	0.19
JLV	40	1.19	47.06	139.44	0.03
GCV	43	1.16	<LMD[a]	-	-
MBS	44	1.73	34.39	209.92	0.05
MMSG	46	1.76	-	-	-
SFR	47	1.57	-	-	-
ADG	47	1.48	<LMD[a]	85.05	-
MD	48	2.69	81.99	214.92	0.03
AR.S	48	2.04	<LMD[a]	44.57	-
ABV	56	2.03	-	-	-
ICS	57	2.78	45.65	157.85	0.06
W AV	59	1.37	17.32	200.47	0.08
ACS	67	2.49	10.75	210.27	0.23

a = 4 $mBq.d^{-1}$

The comparison above has suggested that the contribution of thorium inhalation may be significant. The expected values of daily urinary and fecal excretion were tested using the thorium biokinetic model described by ICRP publication 69 [3], simulating a chronic intake of ^{232}Th by inhalation. As the thorium in monazite is classified as type S, AMAD = 1 μm, the predicted urine to feces ratio would be 1.4×10^{-3}, which was still low compared to the experimental one. If the compound was classified as Type M, AMAD = 1 μm, the predicted urine to feces ratio was 8.5×10^{-2}, which is close to the average of the experimental one, 1.2×10^{-1}.

In Figure 1 it is shown that there was an inverse correlation between the urine to feces ratio with the amount of thorium in feces. The analysis of this figure has reinforced the suggestion of a combination of intake through inhalation and ingestion. The low urine to feces ratio values were influenced by the high amounts of thorium in feces, indicating a significant contribution of the thorium of diet in feces. The urine to feces ratios higher than 0.08 has indicated that the predominant route of intake was through inhalation. The thorium content in urine may be influenced by the inhalation. These results have indicated that the f_1 value for thorium is really low as suggested by ICRP 69 [3], about 5×10^{-4} or even 1×10^{-3} as suggested by ICRP 48 [9] for plutonium in diet, and the method for thorium determination in urine was not sensitive enough to detect the increment from the contribution of thorium from diet.

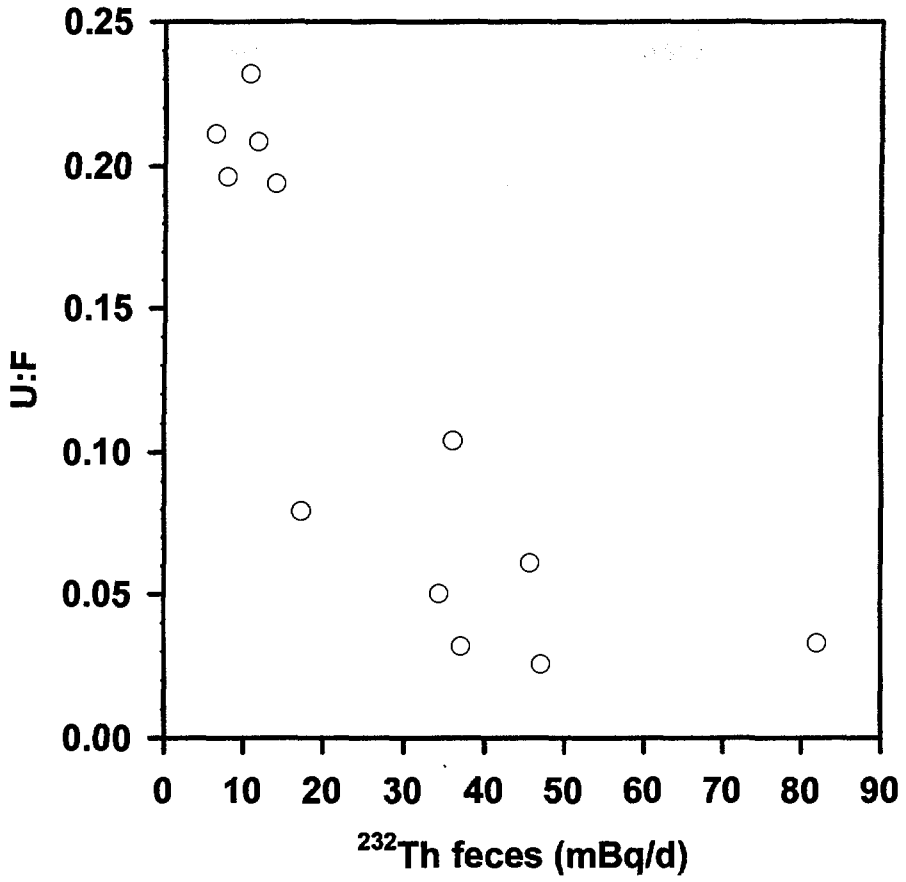

Figure 1. ^{232}Th urine to feces ratio as a function of the ^{232}Th fecal excretion.

Dose Calculation

For the adults, it was assumed that the thorium chronic intake began after 20y old. The biggest contributor for the dose was the intake through inhalation, since the dose coefficient for inhalation of a ^{232}Th compound Type M (4.5×10^{-5} Sv.Bq^{-1}) is almost two orders of magnitude higher than the dose coefficient for ingestion (2.3×10^{-7} Sv.Bq^{-1}) [7]. The average value for committed effective dose per year of exposure, by inhalation of ^{232}Th, for the chronically exposed adult living in Buena is 1.3 mSv. The committed effective dose from inhalation of ^{228Th} is about 70% of the one due to ^{232}Th. Although the ^{228}Th intake through ingestion was the highest, as seen in Tables I and II, the average value for committed effective dose per year of exposure, by ingestion was of order of 4 μSv.

CONCLUSIONS

- The thorium excretion in urine is related to the amount solubilized from the lungs, the clearance of the thorium from the skeleton and clearance from other organs and soft tissue, with very small contributions from the fraction absorbed from the daily intake through diet. The amount excreted in feces reflects the intake through diet, including the lack of equilibrium between ^{232}Th and ^{228}Th in fecal excretion.
- Although thorium in monazite has been classified as Type S in the literature, the use of Type M compounds was compatible with the experimental data.
- Inhalation was the main contributor for the internal dose due to chronical exposure to ^{232}Th.

- The excretion rate of thorium in urine do not increase as the excretion rate of thorium in feces increases for samples from inhabitants of Buena, reflecting the poor absorption of thorium through the gastrointestinal tract.

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