OCCUPATIONAL EXPOSURE TO NATURAL RADIATION IN BRAZIL

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Occupational Radiation Exposures
UNSCEAR 2000

AVERAGE DOSES TO WORKERS

Radiation source	*Number of monitored workers*	*Effective dose (mSv per year)*
Man-made sources		
• Nuclear fuel cycle	900,000	2.9
• Other industry	600,000	0.9
• Defence activities	400,000	0.7
• Medicine	2,200,000	0.5
Total	**4,100,000**	**1.1**
Enhanced natural sources		
• Mining (excluding coal)	700,000	6
• Coal mining	3,900,000	0.9
• Aircrew	250,000	3
• Other	~300,000	<1
Total	**5,200,000**	**1.7**

Occupational Exposure to Natural Radiation

- **Extracting and Processing Industries:**

  - Uranium mining and milling
  - Coal mines
  - Metal milling and smelting
  - Phosphate industry
  - Fertiliser industry
  - Rare earth and ceramic industries
  - Pigment industries
  - Oil and gas exploration and production operating companies

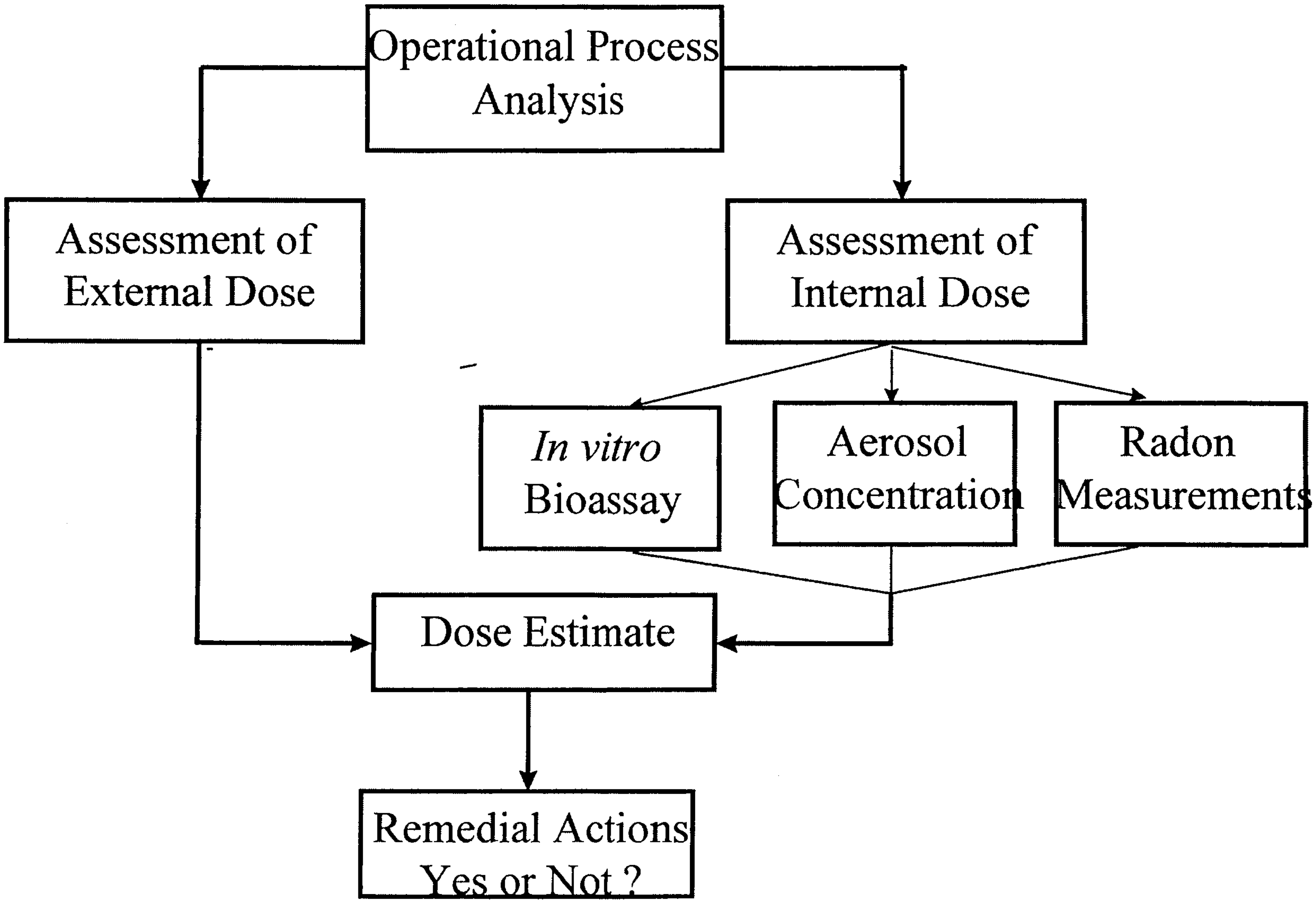
Occupational Exposure to Natural Radiation

- **Power production using coal**
- **Drinking water supply facilities**
- **Aircraft operation**

SURVEY PILOT PROGRAM IN CONVENTIONAL MINING INDUSTRIES IN BRAZIL

- Monazite sand extraction
- Coal mines
- Gold mine
- Nickel mine
- Niobium mine
- Phosphate mine

APPLIED METHODOLOGY FOR OCCUPATIONAL DOSE ASSESSMENT



INTERNAL DOSE ASSESSMENT

Mine	Average committed effective dose (mSv)
Coal	> 1
Phosphate	< 1
Nickel	< 1
Gold	< 1
Niobium	> 1
Monazite sand	> 1

PRELIMINARY CONCLUSIONS

- Coal Mines – Rn control program
- Niobium Mines – Internal monitoring program for Th, U and their decays products
- Monazite Sands - Internal monitoring program for Th, U and their decays products
- Nickel and Gold Mines - No enhanced internal contamination
- Phosphate Mines – No need of internal monitoring program.

SURVEY PROGRAMMES BEING CARRIED OUT IN BRAZIL

- **Oil and gas exploration and production operating companies**
- **Drinking water supply facilities**
- **Aircraft operation**

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Exposure of workers in mineral processing industries in Brazil

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Abstract

The mining, milling and processing of uranium and thorium bearing minerals may result in radiation doses to workers. A preliminary survey pilot program, that included six mines in Brazil (two coal mines, one niobium mine, one nickel mine, one gold mine and one phosphate mine), was launched in order to determine the need to control the radioactive exposure of the mine-workers. Our survey consisted of the collection and analysis of urine samples, complemented by feces and air samples. The concentrations of uranium, thorium and polonium were measured in these samples and compared to background data from family members of the workers living in the same dwelling and from residents from the general population of Rio de Janeiro. The results from the coal mines indicated that the inhalation of radon progeny may be a source of occupational exposure. The workers from the nickel, gold and phosphate mines that were visited do not require a program to control internal radiological doses. The niobium mine results showed that in some areas of the industry exposure to thorium and uranium might occur. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Enhanced natural radiation exposure; Mining workers internal exposure; Uranium; Thorium; Polonium

1. Introduction

In the mining industry, workers may be exposed to enhanced natural radiation. In some mines, thorium and uranium are the principal nuclides of concern in

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occupational radioprotection programs. In others the main problem is radon exposure. A preliminary survey pilot program, that included six mines in Brazil, was established in order to determine the need to control the radioactive exposure of the mine-workers. The mines were chosen based on the association of thorium and uranium with the main mineral. They were part of a list selected by the department of prime materials, responsible for control of minerals and prime material. The industries that were chosen comprised a niobium mine, two coal mines, one gold mine, a nickel mine and one phosphate mine. Brazil is responsible for 80% of the world production of niobium and the mine that was visited is one of the three main installations in Brazil for niobium production. The coal mines are known for their radiological impact and one of the mines that was chosen is located near a discontinued uranium mine. Gold production is substantial in Brazil and has been increasing in the last decade. The levels of uranium and thorium in the gold mine that was chosen are 9 and 12 ppm, respectively. The nickel mines, located in a town economically dependent on the mine and named Niquelândia, were selected due to the toxicity of the element. The phosphate mineral from the mine that was chosen occurs associated with titanium, niobium and rare earths and the thorium and uranium levels are 55 and 27 ppm, respectively.

2. Materials and methods

The internal contamination of the professionals working in the mines was evaluated through indirect monitoring methods, consisting of the analysis of the concentrations of uranium, thorium and polonium in excreta samples and of thorium and uranium in air samples.

For the analysis of the concentration of uranium and thorium in excreta samples, different analytical and detection methods were used, depending on the mine. When the survey program started only alpha-spectrometry methods and fluorimetry were available at our laboratory. The methods used for analysis of samples are described in the literature (Azeredo, Melo, Dantas & Oliveira, 1991). The nickel mine was visited after the acquisition of inductively coupled plasma-mass spectrometry (ICP-MS) equipment and thus, samples from this mine as well as samples from one of the coal mines and from the last visit to the niobium mine were analyzed by this technique. The lower limits of detection (Altshuler & Pasternack, 1963) for thorium and uranium were around 1 mBq per liter of urine and 1 mBq per gram of fecal ash, using our analytical separation technique followed by alpha-spectrometry, 5 μg per liter of urine for uranium fluorimetry and 7.4×10^{-5} ppm for thorium and 1.95×10^{-5} ppm for uranium using the ICP-MS technique.

The ICP-MS technique for the analysis of urine samples only requires dilution. An aliquot of 1 ml of the 24 h urine sample is diluted 1 : 20 by the addition of 19 ml of 1 N HNO_3, prepared using hyper-pure HNO_3 and deionized, doubly distilled water. The feces samples required 24 h calcinations, with temperatures that were slowly raised to 400°C and further calcinations for 12 h at 600°C, before the dissolution of 0.01 g with 20 ml of HNO_3 on a hot plate.

Polonium was measured in urine samples only for screening, as an indication of a possible radon progeny exposure. The measuring method is described elsewhere (Azeredo & Lipsztein, 1991).

Some workers used personal air samplers with cyclones, with cut-off diameter of 2.5 μm, during an 8 h working period. In some installations, a stacked filter unit, with a flow rate of 17 ml/min and a six-stage cascade impactor with an operational flow rate of 12 l/min were placed near the workers at the point of highest total dust concentration and at 1.5 m from the ground and were used to identify the main elements present in the dust, their concentrations and particle size distributions mass medium aerodynamic diameter (MMAD). Nuclepore filters, 47 mm diameter, and 0.4 and 8.0 μm porosity were used in the stacked filter. The impacting surfaces of the cascade impactor consisted of Mylar films. The particles collected through the personal air samplers and through the stacked filter unit were analyzed for uranium and thorium using alpha-spectrometry techniques. The elemental composition of the mass impacted at each stage of the cascade impactor was measured by particle induced X-ray emission (PIXE), using a 4.0 MV van de Graff accelerator. The concentrations of thorium and uranium measured through the cascade impactor were derived assuming that the thorium and uranium masses were due to ^{232}Th and ^{238}U. The lower limits of detection (Altshuler & Pasternack, 1963) in activity concentrations for ^{232}Th, ^{228}Th, ^{238}U and ^{234}U in samples collected using personal air samplers were 0.9, 1.1, 0.8 and 0.9 mBq/m^3, respectively; and for samples collected by the stack filter unit were 0.7, 0.8, 0.6 and 0.7 mBq/m^3, respectively. The minimum mass concentrations for thorium and uranium in samples collected using the cascade impactor were 3.1 and 3.9 ng/m^3, respectively. The methodologies used in the analysis of all samples are described elsewhere (Dias da Cunha, Lipsztein & Barros Leite, 1994; Dias da Cunha, Lipsztein, Fang & Barros Leite, 1998).

Each mine was treated differently, with respect to the experimental design for sample collection and comparison with background. As these mines are exempted from radiological protection control, samples were collected on a voluntary basis, following agreements with the managers of the mines. All collaboration was welcome but the degree of collaboration varied from mine to mine. A sample comprising 68 people from Rio de Janeiro was used as a control for the uranium and thorium excretion data. The average excretions of uranium and thorium in urine were 0.068 ± 0.028 and 0.159 ± 0.051 μg /l, respectively. The average excretion rates of ^{232}Th and ^{228}Th in feces were 2.3 ± 1.5 and 14.3 ± 6.8 mBq/g ash. The average excretion rate of uranium was 0.25 ± 0.2 μg/g ash. For the polonium excretion the control group from Rio de Janeiro comprised 28 male individuals of the same age as the mine-workers, 15 of them non-smokers. Average ^{210}Po excretions of 9.9 ± 4.1 and 5.2 ± 2.2 mBq/d were found for smokers and non-smokers, respectively. In the statistical treatment of data, analysis of variance and the Student's–*t* tests were used for comparing the nickel and niobium mine data with the control, using $\alpha = 0.05$ (Rosner, 1990). The non-parametric Kruskal-Wallis and Dunns' multiple comparison statistical tests were applied (Hollander & Wolfe, 1973) to the coal mine data in comparison with the control ($\alpha = 0.05$).

The evaluation of the risks from the contamination is accomplished through the interpretation of the measurements in terms of intake and the assessment of dose from the intake. The biokinetic models for thorium and uranium recommended in publication 69 of the ICRP (1995) were used to calculate the daily urinary or fecal excretion as a fraction of the intake and at specific times after intake. When calculating the intakes incurred at work, a chronic 5 days-a-week inhalation exposure, was assumed. When calculating the public exposure, a chronic constant exposure through inhalation or ingestion was assumed. The air measurements were used either to calculate standard man intakes or the MMAD, which were assumed to be equal to the AMAD. The assessments of the effective doses from the intakes were accomplished using the dose coefficients from the ICRP database of dose coefficients for workers and members of the public (ICRP, 1999).

The polonium bioassay data were used with the sole purpose of indicating possible radon or progeny exposure and the need for further investigations.

3. Results and discussion

3.1. Coal mines

Two coal mines were visited. Coal mine I is situated in the north of the state of Paraná, located in the proximity of an old discontinued uranium mine. There are three excavation sites. The first one, with an average depth of 138 m, has a vertical access by elevator. The second one is reached by foot, through a slanted ramp, and has an average depth of 41 m and will be referred in the text as site II. The last one has a vertical access, 128 m average depth, and at the time of the visit was being prepared to begin operation. Another vertical well is used to ventilate the other mining areas. Fifteen people were working in the first excavation site, 80 people in the second one and 250 preparing the third cave, when the visitation took place. The workers take approximately 1 h to reach the place of coal excavation, where they work for 6 h. The temperature inside the mine is about 23°C. During the working period there is a 15-min snack time, at the work-place. The workers were observed smoking while working. They do not use respiratory protective masks.

The mineral ores are loaded into cable cars. At the vertical well they are manually emptied into boxes, which are then taken to the surface by the elevator and mechanically thrown into the silos. At the inclined ramp the cars are taken to the surface and mechanically brought to the silos. Coal is transported to the processing area in open trucks. At the processing area (open area) coal is crushed, washed and classified by granulometry for sale.

Urine samples from 14 workers in the excavation and coal-processing jobs and two from administrative personnel were collected. These samples were analyzed for uranium and polonium concentrations. Feces samples from six workers in the excavating job were analyzed for thorium and uranium contents. Thorium and polonium were analyzed by alpha-spectrometry and uranium by fluorimetry. All employees had measurable polonium concentrations in urine but only one worker, from site II, had uranium concentrations, in urine, above the detection limit. From the feces results, only two results were significant, both for uranium and from two workers from site II. The worker that presented uranium in urine did not show concentrations above detection limits in the feces, which is an unexpected result. The committed effective doses associated with the hypothesis of chronic 5 days-a-week inhalation exposure for the positive uranium bioassay results are of the order of 10^{-3} Sv.

The amounts of polonium excreted by personnel working at site II were significantly higher than the quantity excreted by other employees, indicating a possible radon exposure problem at this location, although it may most probably be indicative of inhalable ^{210}Pb rather than of radon itself or the shorter-lived progeny. In Fig. 1 the concentrations of polonium in urine from workers of this coal mine are compared to the background concentrations from smokers and non-smokers from Rio de Janeiro. The amounts of polonium excreted at site II were much higher than the amounts excreted by all other groups, dispensing all considerations for statistically confounding factors.

Four workers and four of our staff used personal air samplers. All thorium and uranium results were below detection limits.

Coal Mine II is located in the state of Santa Catarina, also in the south of Brazil. The mine is 150 m deep. Only the main excavation site was visited. The ores are

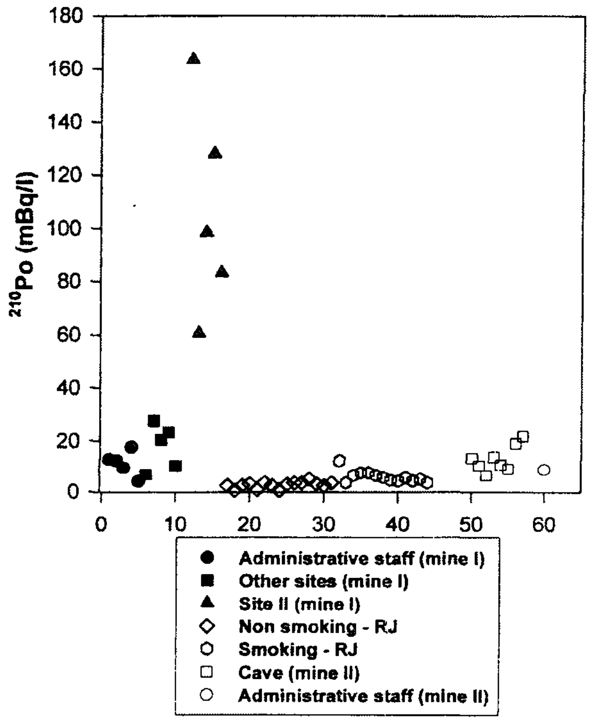

Fig. 1. Excretion of polonium in urine samples of coal miners.

collected and transported in bobcats with pneumatic propulsion. Ten excreta samples, urine and feces, from employees were collected. Seven were from workers doing tasks inside the main cave of the mine (two in the ore excavation, four in the maintenance and preparation of the exploration sites and one in the transportation), one from a worker at the crushing site and two from administrative personnel. Samples from the wives of three workers were also collected. These samples were analyzed for thorium and uranium contents, using the ICP-MS technique. For thorium, there were no significant differences amongst the different jobs or between workers, their wives and the background samples taken in Rio de Janeiro. For uranium, the concentrations excreted in the feces of the workers were similar to those excreted by their wives and by the people living in Rio de Janeiro. The concentrations of uranium in urine samples obtained in Rio de Janeiro were lower than those in the workers, although there were no significant differences amongst the different jobs or between workers and their wives, as presented in Fig. 2.

A cascade impactor was used to characterize the MMAD from the thorium and uranium aerosols. Samples were analyzed using PIXE. Inside the mine and at the crushing site the MMADs varied between 1.0 and 1.7 μm and the thorium and uranium concentrations were about 0.05 and 0.22 $\mu g/m^3$, respectively. Eight of the workers from whom excreta samples were collected used personal air samplers, as also the two other workers from the crushing jobs. Samples were analyzed for thorium and uranium contents. Results were below detection limits for thorium. Positive results for uranium were found, in ranges of 2.2–5.9 mBq/m^3 for ^{238}U and 2.5–5.9 mBq/m^3 for ^{234}U.

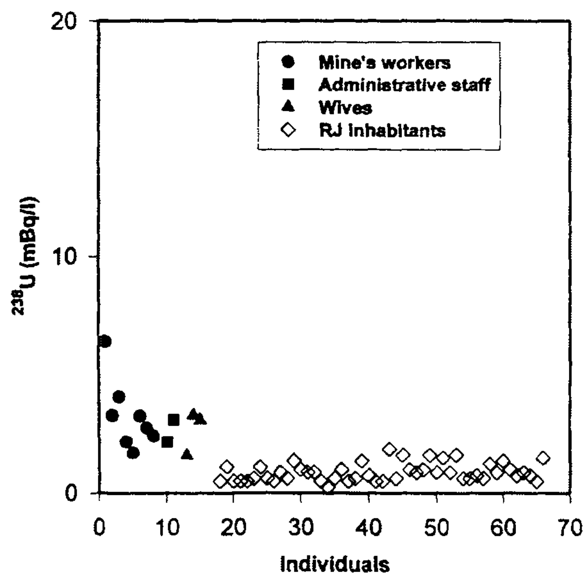

Fig. 2. Concentrations of uranium in urine samples from coal miners and their wives.

The hypothesis of chronic continuous inhalation exposure was used in the interpretation of the excreta results. A daily intake of uranium that varied from 0.20 to 0.58 Bq was calculated, for the workers and their wives. The committed effective doses and the annual effective doses for uranium for coal mine workers and their wives are of the order of μSv.

Urine samples from the workers were also analyzed for polonium concentrations. These results are shown in Fig. 1, in comparison with coal mine I workers and Rio de Janeiro background. The amounts excreted by the workers were in the same range as those excreted by mine I workers, with the exception of site II.

3.2. Phosphate mine

The phosphate mine is located in the state of Goiás, in the central part of Brazil, in an area known for its elevated natural uranium and thorium occurence in the soil. The ore is extracted in an open-pit mine and transported by trucks to the crushing area. The ore is homogenized, crushed again and sent for separation, conditioning, flotation, filtration and final drying processes. Waste is produced in the conditioning and filtration steps. Four workers performing tasks in the extraction of the ore, crushing, flotation and loading of the transportation train were asked to wear personal air samplers. The same four workers, plus two administrative staff, were asked to collect 24 h feces and urine samples. The uranium concentrations were measured using fluorimetry and results were below detection limits for all samples. All workers, including the administrative staff, presented positive results for ^{228}Th, in the range of 33–175 mBq/g of ash, probably originating from food consumption. Three workers from the crushing, flotation and final dispatch sections presented measurable quantities of ^{232}Th in their feces, in the range of 15–23 mBq/g of ash. The associated effective doses are of the order of μSv. The personal air sampler used by one worker from the final dispatch process (loading of the transportation train) was the only one to present measurable activities of thorium, but the committed effective dose associated with this result is again of the order of μSv. In the thorium dose calculations a chronic intake by inhalation, five days per week was assumed.

3.3. Nickel mine

The nickel mine is located in the north of the state of Goiás, central-west region of Brazil. A cascade impactor was used to characterize the air particles in the mine. Aerosols containing thorium and uranium were not detected. The aerosol samples were measured by PIXE, and uranium and thorium masses were below detection limits (about 12 and 15 ng for thorium and uranium, respectively). Bioassay samples from workers, as expected, did not show significant concentrations of the nuclides. Urine and feces samples were analyzed by ICP-MS. The average uranium excretion for 24 mining workers was 0.10 ± 0.02 μg/l, and this result was shown to be not significantly different from the background excretion rate for the nuclide in Rio de Janeiro ($p > 0.05$). The average thorium excretions for the same workers, 0.18 ± 0.04 μg/l, were also not significantly different from the background in Rio de Janeiro ($p > 0.05$). The average concentrations of thorium and uranium in the feces of the workers were respectively 0.36 ± 0.23 and $0.33 \pm 0.19\,\mu g/g$ of ash, not significantly different from the control group and from the values excreted by the 26 workers' wives ($p > 0.05$). Nickel was detected in significant amounts in air samples, urine, feces and hair from the workers and their wives. The potential risks for the workers were evaluated by Azeredo (1998). There are no additional risks from radioactive exposure to uranium and thorium in this mine.

3.4. Gold mine

The gold mine that was visited by our staff is located in the northeastern region of Brazil, in the state of Bahia. There are two extraction sites. The deepest part of the first is situated at 655 m from the surface and is accessed by ramp. The deepest part of the second one is located at 360 m from the surface. Air monitoring was conducted using a cascade impactor and static air samplers, at 18 different places, including the extraction sites and the crushing and milling locations. Workers from these sites and from the waste barrage wore personal air samplers. All air monitoring results for thorium and uranium were below detection limits. Seventeen urine samples were collected over from workers spread in all sites of the mine, including two samples from administrative personnel. The concentrations of thorium, uranium and ^{210}Po were all below detection limits.

3.5. Niobium mine

The niobium mine, located in the south of the state of Goiás, is a major producer of ferro-niobium alloy. The installation comprises an open-pit mine and a metallurgical plant. The production process consists of the mineral extraction, crushing, leaching and concentration of niobium, followed by the aluminothermic production of the Fe–Nb alloy, its further crushing, screening, packing and dispatch. Our staff visited this mine several times (Dias da Cunha, Lipsztein & Barros Leite, 1998). The results reported in this work refer to the last visit, which took place in 1998.

A six-stage cascade impactor, placed near the workers at the point of highest total dust concentration and at 1.5 m from the ground, was used to identify the main elements present in the dust, their concentrations and particle size distributions (MMAD). The main elements identified were Nb, Th, U, Pb and Zr. During the aluminothermic process and Fe–Nb crushing, the Nb concentrations were the highest and the MMAD was about 1.0 µm in both locations. Detectable amounts of uranium were generated during the crushing of the mineral and in the aluminothermic process. The MMAD varied from 1.1 to 2.2 µm. Thorium was present at all stages of the process, with average concentrations less than $0.5\,\mu g/m^3$ and MMAD from 0.8 to 1.4 µm.

A group of nine workers used personal air samplers with a cyclone, and collected feces and urine samples. The wives of the workers were asked to collect feces and urine samples. The ^{232}Th and ^{228}Th activities in these samples were determined, as well as the niobium and uranium concentrations.

Three workers had measurable thorium activities in the air filters. They were involved in different mining activities, open-pit mine, mineral crushing and Fe–Nb crushing. Their peers, engaged in the same jobs, did not show activities higher than the detection limits. All workers presented measurable thorium concentrations in feces. The results did not correlate with the personal air sample data. The ^{228}Th activities in feces were much higher than the ^{232}Th activities, in contrast with the air samples, in which the two nuclides are in equilibrium. The ^{232}Th concentrations in feces varied from 2.4 to 60 mBq/g ash and the ^{228}Th varied from 10.3 to 131 mBq/g ash. The amounts of thorium present in the feces of the wives were lower than their husbands, but again there was a lack of equilibrium between ^{232}Th and ^{228}Th. The ^{232}Th and ^{228}Th concentrations in feces of these workers were measured in samples collected before and after 30 days vacation, prior to their return to work. The thorium content of feces samples after a prolonged absence period from work reflects the occupational exposure from inhalation, since all ingested thorium at work would have been excreted after 30 days. The samples taken at all other periods of the working year characterize both the inhalation and the ingestion pathways of intake during work (Azeredo, Juliáo, Santos, Melo, & Lipsztein, 1994; Julião, Azeredo, Santos, Melo, Dantas, & Lipsztein, 1994; Dias da Cunha et al., 1998). After vacation, the ^{232}Th concentrations in the feces of the workers varied from the detection limit up to 11.7 mBq d^{-1} and the ^{228}Th concentrations were in the range of 1.8–63.4 mBq d^{-1}. The ^{232}Th concentrations in feces samples from wives are in the same range as the ^{232}Th concentrations in feces from the workers in samples collected after their vacations. These results indicate that the thorium excreted in feces is not a consequence of inhalation exposures during work. The amount of thorium present in 21 urine samples from workers performing different jobs did not differ from the amount present in the urine samples of a control group in Rio de Janeiro. These results indicate that ingestion is the most important pathway of thorium intake. For the workers, contaminated hands and mouth are important vehicles for ingestion as well as inhalation of coarse particles. Since the lack of radiological equilibrium was also observed in food consumed in the region, locally produced foodstuffs are a substantial source of thorium intake. The workers' committed effective doses for thorium, considering the ingestion pathway of intake, are of the order of µSv.

The uranium concentrations in personal air samplers were only significant in the aluminothermic process and in the crushing of the Fe–Nb alloy. The uranium concentrations in urine samples from 19 workers engaged in different jobs were not different from the control group in Rio de Janeiro. The amounts of uranium excreted in the feces of six workers were significantly higher than the average concentrations excreted by the control group and by the group of wives, as visualized in Fig. 3. Using the hypothesis of chronic continuous exposure through inhalation, the workers committed effective doses for uranium are of the order of mSv. If the exposures to uranium were from ingestion the committed effective doses would have been much lower.

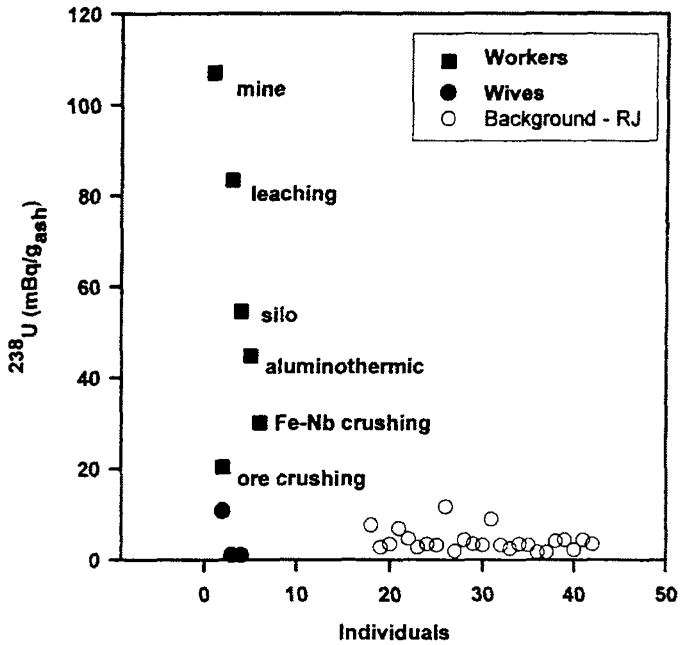

Fig. 3. Concentrations of uranium in feces samples from niobium miners and their wives.

The average niobium concentrations in feces and urine samples from workers were 42.4 ± 39.6 and $1.34 \pm 0.45\,\mu g\,d^{-1}$, respectively. The average niobium concentrations in feces and urine samples from the control group were 3.4 ± 3.2 and $0.60 \pm 0.45\,\mu g\,d^{-1}$, respectively. The amounts of Nb excreted by the workers are significantly different from the control group ($p > 0.05$). The niobium concentration in feces samples from workers suggests that the workers inhale coarse particles and ingest mineral dust particles probably as they touch their mouths with dirty hands. The niobium concentrations in urine samples indicate that there are systemic incorporations of the element.

Although the committed effective doses for both uranium and thorium are low, an annual monitoring program for evaluating workers' exposures is recommended.

4. Conclusions

The results of this preliminary survey program indicate that there might be a need for a program to control radon (short- and long-lived progeny) exposure in the coal mines and for a minimum internal monitoring program for uranium and thorium in the niobium mine. In the nickel facility and in the gold mine that were visited, there are no enhanced internal contaminations due to radioactive elements. In the phosphate facility, the level of internal contamination does not justify the implementation of radioprotection programs. Future studies should be carried out to further evaluate the need to control in these industries.

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