THORON EXPOSURE FOR WORKERS WITH NATURALLY OCCURRING RADIOACTIVE MATERIALS

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1 ABSTRACT

Thoron levels and consequently radiation dose by its inhalation were determined for workers that handled naturally occurring radioactive materials in two facilities at the Instituto de Pesquisas Energéticas e Nucleares (Ipen), São Paulo, Brazil.

2 INTRODUCTION

Naturally occurring radioactive materials (NORM) as uranium and thorium, are hazardous to health and it has long been recognized that work with NORM can rise significantly the exposure in the workforce (1). NORM can affect workers, basically by inhalation or ingestion of airborne radioactive dust and by inhalation of radon and thoron isotopes and their daughters. By far, inhalation of ^{222}Rn (radon), a radon isotope with a half-life of 3.8 days from the ^{238}U series, represents the main contribution to the effective dose received by the workers. Since ever, thoron (^{220}Rn, a radon isotope with a half-life of 55 s, from the ^{232}Th series) contribution, had been neglected, mainly due to its relatively short half-life and lower concentrations of ^{232}Th, when compared to ^{238}U ones, but, lately, this approach has been reconsidered.

It is well known (2) that residents of high thorium background areas, like Brazil, China and India, receive elevated doses. In addition, recent studies shown that, besides the residents of those high background areas, workers in the mineral sand processing industries receive elevated doses up to 180 mSv.y^{-1}. Also, it was observed a higher probability of lung cancer for uranium miners than in control groups and great discrepancies in the risk estimates in those investigations that cannot be explained only by the levels of radon and its daughters (3). Some of the discrepancies may be due to differences in thoron concentrations, as in places with high thorium concentration, the thoron concentration exceeds the radon concentration. So, the thoron contribution can leave to a rise of the dose in the lungs and must be considered.

Beyond the environmental aspects and the mineral sand processing industries, there are other relevant uses for thorium to be considered, like fuel for nuclear reactors or conventional uses (4) such gas lamp mantles, pigments or certain optical glasses.

At the Instituto de Pesquisas Energéticas e Nucleares (IPEN), São Paulo, Brazil, there are two facilities dealing with great thorium concentrations, the nuclear materials storage site and the thorium purification plant.

Uranium and thorium safeguarded materials are stored at the nuclear materials storage site, a 150 m length per 20 m width place with an unique door and no windows or artificial ventilation. This safeguarded site remains closed all the time and only authorized workers open it.
At thorium purification plant, the thorium sulfate is converted in concentrated thorium nitrate with high purity degree. The thorium purification is processed in four stages, as follows: stage 1, the thorium sulfate is weighted, the process takes 10 minutes and 150 kg are handled; stage 2, sodium carbonate is added to the thorium sulfate during 10 minutes, a total of 50 kg; stage 3, water is added to the compound of stage 2 during 60 minutes and 80 kg are handled; and stage 4, with addition of HNO_3 to the thorium oxi-carbonate, a total of 10 kg; this process also takes 10 minutes and is made 3 times a day. The operation stages 1, 2 and 3 are realized into the same large room and by the same workers and stage 4 is realized elsewhere at the plant, by another group.
All workers at those places are continuously handling thorium and certainly their health risks by thoron inhalation cannot be neglected. In order to estimate the lung cancer risk for the workers, it was necessary to determine the levels of thoron concentrations in the air.
The thoron direct determination is difficulted by its short half-life (55.3 s). So, to obtain the thoron concentration in the air, we use the concept of equilibrium equivalent concentration (EEC), defined as the thoron concentration, in equilibrium with its short-lived daughters, that has the same potential alpha energy concentration per unit volume as exists in a sample mixture (5).
The contribution from ^{220}Rn and its progeny was determined by sampling the air within the work areas and measuring the filters by high resolution gamma-ray spectrometry, alpha spectrometry and total alpha counting.
The committed effective dose from exposure to thoron progeny was obtained from the thoron equilibrium equivalent concentration and dosimetric models for inhalation and distribution of airbone radioactivity by human body. These models incorporate the data on respiratory tract deposition and clearance of particles, as well as the behaviour of inhaled radionuclide into the body.
The primary pathway for health detriment associated with ^{220}Rn arises from inhalation of the short-lived decay products (^{212}Pb e ^{212}Bi) and the resultant alpha particle exposure to the respiratory tract, but, due to the relatively long half-life of ^{212}Pb (10.6 h), a considerable fraction of the energy deposited in the lungs is transferred to other organs. For this reason, the committed effective dose from thoron daughters inhalation was calculated considering the activity incorporated as being only the activity of ^{212}Pb.

3 SAMPLING AND MEASUREMENT TECHNIQUES

The ^{220}Rn equilibrium equivalent concentration was obtained by the active detection method, when the air pumped by an air sampler is collected on glass fiber filters with diameter of 47 mm. The methodology of sampling and the measurement techniques were previously described (6).
At the nuclear materials storage site, 54 air filters were collected, at different dates, one hour of sampling, with door open. The samplings were done when the authorized personal was working as well as with no workers. At the thorium

purification plant, we sampled 28 air filters, at different days, following the purification process stage and the working time.
We must point, however, that the purification process does not necessarily follows in chronological order the four stages. Great quantities can be weighted at stage 1 and kept stored, which explains why several samplings for stages 2, 3 or 4 are older that the stage 1 ones.

4 THORON EQUILIBRIUM EQUIVALENT CONCENTRATION RESULTS

The equilibrium equivalent concentration (EEC) of thoron was obtained as described elsewhere (6). The results are presented in figures 1 and 2.
Figure 1 shows the thoron equilibrium equivalent concentrations (EEC) at the nuclear materials storage site between August 1996 and June 1997. Figure 2 shows the results of thoron equilibrium equivalent concentrations (EEC) at stages 1, 2, 3 and 4 of the thorium purification process, between June 1997 and December 1998.

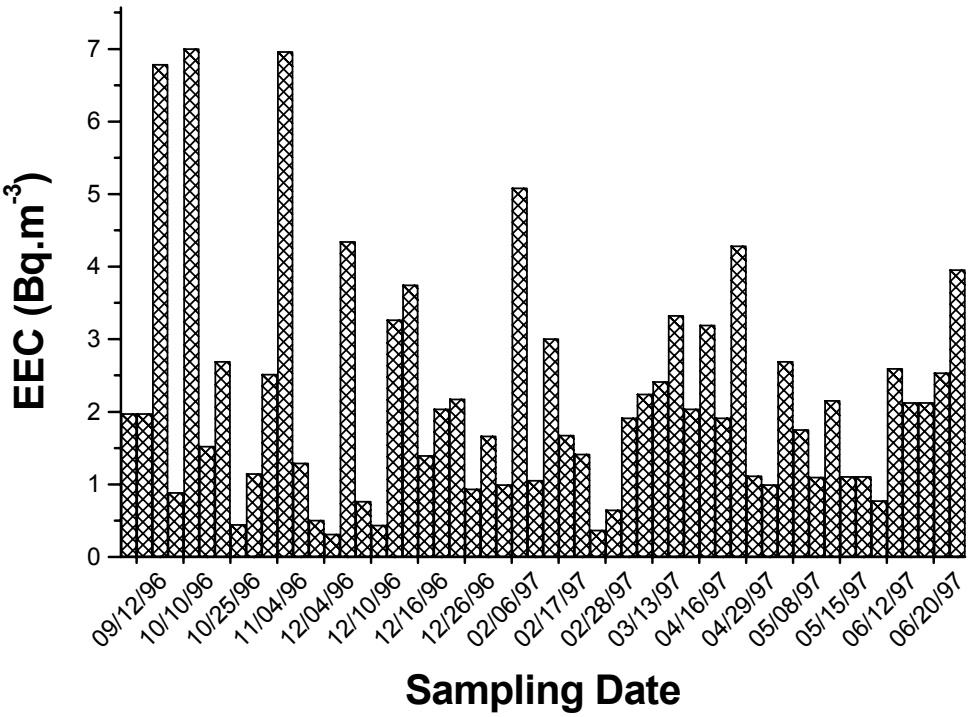

Figure 1: Thoron equilibrium equivalent concentrations (EEC) at the nuclear materials storage site between August 1996 and June 1997

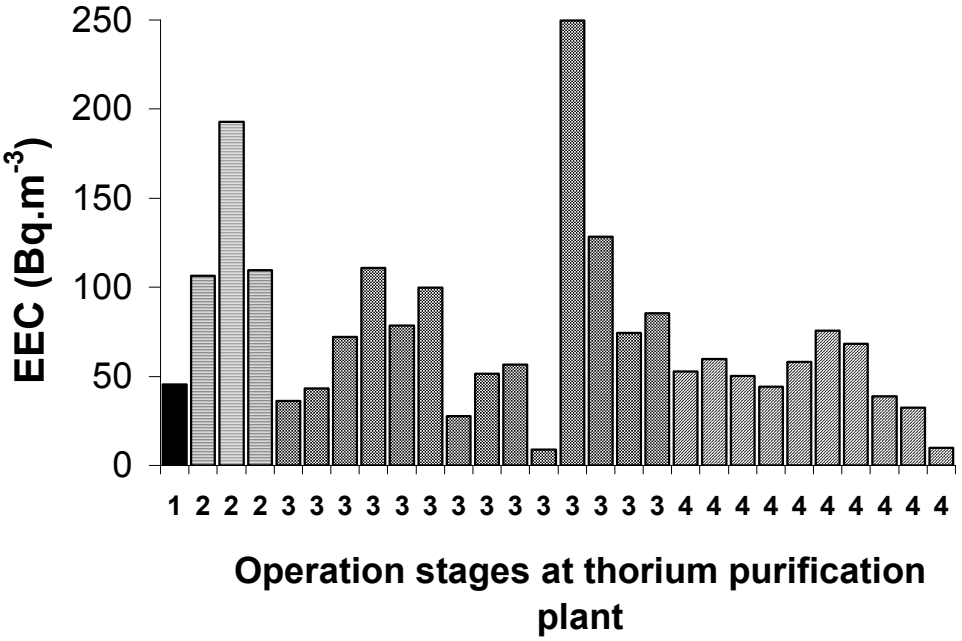

Figure 2: Thoron equilibrium equivalent concentrations (EEC) at stages of the thorium purification process between June/1997 and December/1998

5 COMMITTED EFFECTIVE DOSE RESULTS

The inhalation is the main pathway of thoron daughters intake and the ^{212}Pb is the main contributor for radiation dose, so we use the ICRP 66 lung compartment model (7) and the ICRP 67 lead metabolic model (8) to describe the radionuclide distribution into the body and to calculate the committed effective dose.

For purposes of modeling, four regions represent the respiratory tract. The extrathoracic region comprises the anterior and posterior nasal passages and the oral passages, including the mouth, pharynx and larynx with associated lymphatic tissues. The thoracic regions are bronchial consisting of the trachea and bronchi, bronchiolar consisting of the bronchioles and terminal bronchioles and alveolar interstitial consisting of the respiratory bronchioles, the alveolar ducts and sacs with their alveoli, also with associated lymphatic tissues.

The model evaluates fractional deposition of an aerosol in each dosimetric region considering the aerosol size, inhalability and nose and mouth breathing and gives reference values of regional deposition for reference workers.

Materials deposited in the respiratory tract are cleared by three main routes: into blood by absorption, to the gastrointestinal tract via the pharynx and to regional lymph nodes via lymphatic channels.

The functions describing uptake and retention in a body tissue following particle intake are quite complex and therefore, it is convenient to describe the transfer of radionuclide from air to body by simple models which facilitate calculation and yet still allow sufficiently accurate dose estimates.

According to the metabolic model described in the ICRP 67, lead leaving the transfer compartment is distributed to blood, liver, kidneys and skeleton. More than 99% of the activity in blood is associated with red blood cells. Skeleton

behaviour of lead appears to be similar to that of the heavier alkaline earth elements, if account is taken of the slower deposition of lead in the skeleton due to competition with other tissues and fluids, particularly red blood cells.
Considering the ICRP 66 lung compartment model, the ICRP 67 lead metabolic model and the lead annual intake (obtained through the thoron equilibrium equivalent concentration, breathing rate and the exposure period), we calculated the committed effective dose due to thoron daughters inhalation using the AnaComp code (9). We assumed that the workers were exposed during 2000 hours per year at the nuclear materials storage site and 500 hours per year at the thorium purification plant and we considered a breathing rate of 1.2 m^3/h (10).
The AnaComp code uses the MIRD (Medical Internal Radiation Dose) formalism to calculate the radiation doses through the compartment model.
Figure 3 shows the values of the committed effective dose at the nuclear materials storage site between August 1996 and June 1997.

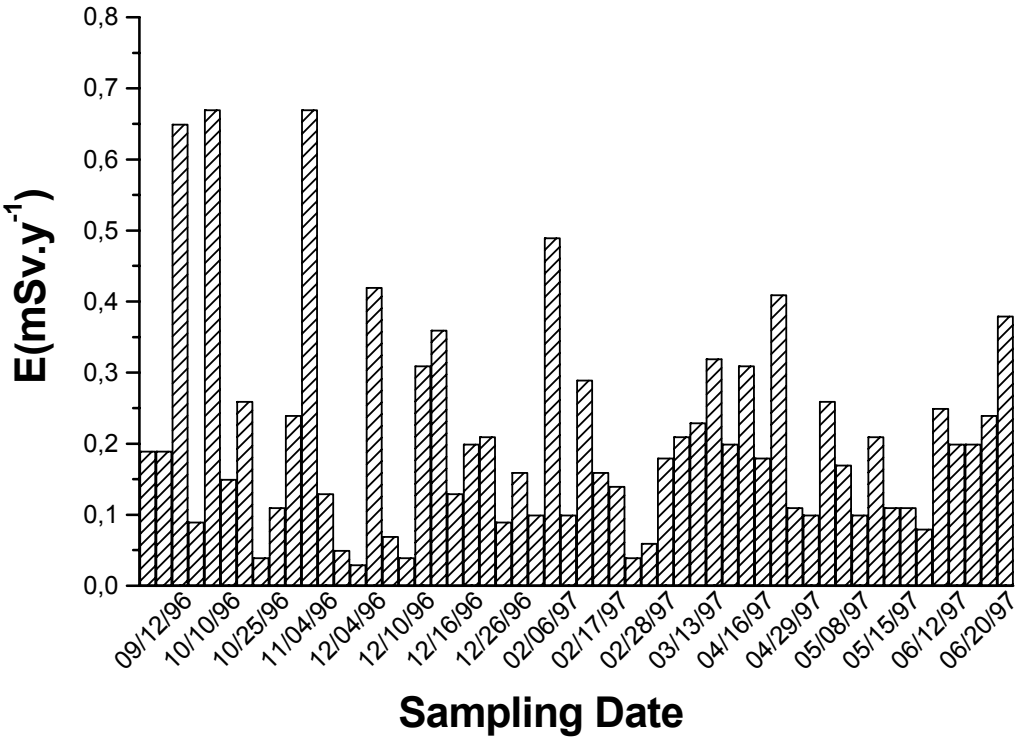

Figure 3: Committed effective dose at nuclear materials storage site between August 1996 and June 1997.

Figures 4 shows the values of committed effective dose at stages 1, 2, 3 and 4 of the thorium purification process between June 1996 and December 1998.

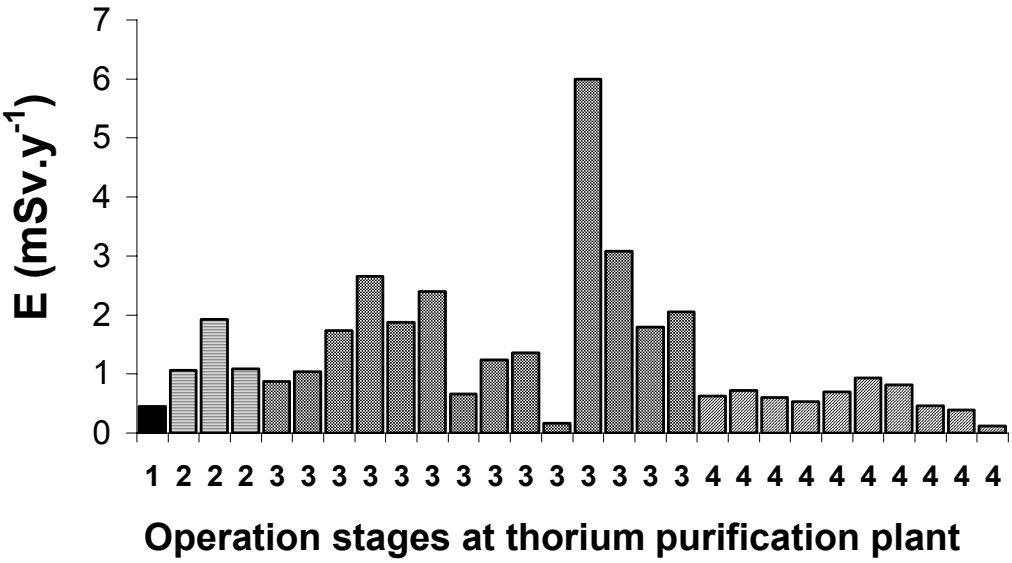

Figure 9: Committed effective dose (E) at stages of the thorium purification process (between June/1997 and December/1998)

6 CONCLUSIONS

As it can be seen from figures 1 and 2, at the thorium purification plant, the thoron equilibrium equivalent concentrations are greater than at the nuclear materials storage site. This is to be expected, as at the nuclear materials storage site, the thorium compounds are stocked in sealed barrels and at the thorium purification plant every time are processed great quantities of thorium compounds are processed in order to extract the thorium.

The variation from 0.32 to 6.97 $Bq \cdot m^{-3}$ in the equilibrium equivalent concentration at the nuclear materials storage site is easily explained by the different sampling conditions, as, when sampling was done with no workers, the door remained open only one hour, and, when sampling was done while workers were handling nuclear materials, the door remained continuously opened, resulting in a considerable exchange air rate.

At the thorium purification plant, the large dispersion, even at the same stage, in the thoron equilibrium equivalent concentration is explained by the different thorium activities and quantities of the manufactured materials handled by the workers every time we sampled the air.

The effective committed dose received by the workers varied from 0.03 to 0.67 ($mSv.y^{-1}$) at nuclear materials storage site and from 0.16 to 6.00 ($mSv.y^{-1}$) at thorium purification plant. At both places the results are below 20 $mSv.y^{-1}$ (11), suggested as an annual effective dose limit for occupational exposure by ICRP 60, considering a working time of 2000 hours per year. Nevertheless, it

must be pointed that, at the thorium purification plant, the Radiological Health Service established a working time of only 500 hours per year. So, the local work routine is correct, as if, for a working time of 2000 hours per year, the annual dose limit will be exceeded.
As result of this study, the thorium purification is now processed in 3 stages, the stage 1 (weighing of the thorium sulfate) was removed in order to optimize the process and to reduce the radiation dose received by workers. The stage 2 is always initiated with the same known thorium sulfate quantity.

Acknowledgements: Campos M. is indebted to CNPq for the financial support as fellowship student during the work.

7 REFERENCES

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