Occupational Dose Assessment in the Cyclotron Installations of IEN During Ultra Pure ^{123}I Production from ^{124}Xe

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ABSTRACT

Since 1998 the Nuclear Engineering Institute (Instituto de Engenharia Nuclear, IEN), Brazil, has produced ultra pure ^{123}I using its KIPROS system and CV-28 Cyclotron. During this production, neutrons and gamma radiation are generated and scattered at different places in the IEN areas, inside and outside the Cyclotron Installations building, increasing the level of radiation exposure at these places. The radiation exposures to the workers of the Institute have been assessed by its occupational monitoring program. The area monitoring program results indicate that the increase of neutron and gamma dose rates, inside and outside the Cyclotron building, is more important close to the load gate of its experimental area. Dose rate values found outside the experimental area, near the gate, are higher than the dose limit established by the Brazilian national radiation protection standards for non restrict areas. IEN individual monitoring program results indicate no measurable individual whole body doses due to this increase of radiation exposure level detected by the Institute area monitoring program.

1. INTRODUCTION

The Nuclear Engineering Institute (Instituto de Engenharia Nuclear, IEN), Brazil, was created in 1962 by an agreement firmed between the Brazilian Nuclear Energy Commission (Comissão Nacional de Energia Nuclear, CNEN) and the University of Brazil, presently the Federal University of Rio de Janeiro. Since then the Institute has been acting in different areas, nuclear and non-nuclear ones, of research and scientific and technological development. Its actuation areas are [(1)]:

- Reactor Physics and Engineering;
- Nuclear Physics, mainly, Radioisotope Production;
- Control and Nuclear Instrumentation;
- Chemistry and Metallurgy;
- Radiation Protection and Radioactive Waste Management.

IEN is constituted by a nuclear installation, the Argonauta research reactor, and by several non-nuclear installations.

The production of radiopharmaceuticals, mainly ^{123}I, is considered as one of the main activities of IEN. Since 1980 IEN has been developing studies aiming the radioisotopes production for nuclear medicine using the CV-28 Cyclotron. Methods were established for the production of ^{77}Br, ^{201}Tl, ^{111}In, ^{67}Ga, ^{131}I and ^{123}I. In the near future the Institute aims to produce ^{18}F as well. Until about 1997, IEN was producing ^{123}I from ^{124}Te, according to the reaction [(1, 2)]:

$$^{124}Te\ (p,\ 2n)\ ^{123}I$$

These radioisotope was produced once a week, at low production levels. Since 1998 IEN has been producing and commercialising ultra pure ^{123}I (^{123}I UP) using the KIPROS system (Karlsruhe Iodine Production System) and CV-28 Cyclotron. In this new production method, ^{123}I is obtained from the following nuclear reactions [(2)]:

$$^{124}Xe\ (p,\ 2p)\ ^{123}I \text{ and}$$
$$^{124}Xe + p \rightarrow {}^{123}Cs + 2n \rightarrow {}^{123}Xe + \beta^+ \rightarrow {}^{123}I + \beta^+.$$

The ^{123}I UP production, using the new method, is carried out twice a week, at levels too much higher than the previous one.

IEN acquired its CV-28 Cyclotron from The Cyclotron Corporation (TCC), USA, in 1973. This machine is a multiparticle compact accelerator, with variable energy, capable to accelerate protons, deuterons, alpha particles and ^{3}He. It was installed in IEN in 1974. It deserves to point out that, although the Cyclotron of IEN is able to accelerate several particles, as it is presented in table 1, since 1998 the machine was dedicated only for the production of ^{123}I UP, using 24 MeV protons, currents of 10, 15 and 20μA and irradiation periods from approximately 3 to 5 hours, normally.

Table 1: Particles and maximum energy ranges delivered by IEN CV-28 Cyclotron (1)

Particle	***Energy (MeV)***	***Current (μA)***
p	2 - 24	40
d	3 - 14	50
$_{2}He^{3++}$	4 - 36	20
α	7 - 28	20

The KIPROS unity is composed by a target modulus, the control modulus and the technical processing modulus. The production of ultra pure ^{123}I is carried out through the irradiation of ^{124}Xe with 24 MeV protons accelerated by the CV-28 Cyclotron. During this production, neutrons and gamma radiation are generated and scattered at different places in the IEN areas, inside and outside the Cyclotron installations building, increasing the level of radiation exposure at these places (2).

The ^{123}I production is carried out in the Cyclotron installations. Basically, these installations are the CV-28 Cyclotron cave, the irradiation lines and the maintenance workshop of the accelerator, the cave of the KIPROS target modulus, the processing and quality control laboratories and the expedition area, where the processed radiopharmaceutical is packed and sent to the hospital (1).

The radiation exposures to the workers of IEN during ^{123}I UP production have been assessed by the occupational radiation protection program (2), that was implanted specifically for this activity, from March 1998 up to September 1999. This program was elaborated according to the requirements established in the Brazilian radiation protection standards (3), that are based on the recommendations contained in the publication 26 of the International Commission on Radiation Protection (ICRP). The objective of this work is to present and to discuss the results of this program.

2. CV-28 CYCLOTRON INSTALLATIONS

The IEN CV-28 Cyclotron and its associated laboratories are installed in a building with an area of 1800m². All restricted areas of the Cyclotron installations are provided with control, safety and detection systems, such as: access control; workplace air exhaust system provided with specific filters; safety interlocks; area monitoring system with portable and fixed gamma radiation monitors; use of special working clothes and of other equipment of individual protection; ordinary concrete shield; radioactive sewerage system with contention and decay tanks; area and individual monitoring program (1).

The Cyclotron building presents two great contiguous construction groups with different foundations and structures, namely:

- Group 1: place where the experimental area and Cyclotron and external irradiation caves are installed. It was especially projected and built to support the high weight load, mainly, due to the concrete shield of the Cyclotron cave;
- Group 2: area composed of three floors, namely:
  - 1st floor (ground floor) it includes the following restricted areas: Nuclear Measurements, Radioisotope Processing and Quality Control laboratories; Expedition area; Cyclotron maintenance workshop; Cyclotron operation room; access control area (access to controlled areas); and some non restrict areas, as the laboratory of cold chemistry;
  - 2nd floor, where the principal auxiliary systems and utilities are installed;
  - 3rd floor, where the exhausters, filters and stacks of the air exhaust systems and refrigeration towers are installed.

Besides these facilities, there are other constructions such as: personnel's rooms, electric substation and the contention and decay tanks for radioactive liquid effluents.

Figure 1 presents the layout of the main existent facilities in the 1st floor of the Cyclotron installations building.

The activities of radioisotope processing are carried out in hot cells situated in the Processing laboratory, located beside the Cyclotron cave. This laboratory is separated from the Cyclotron cave by a concrete wall of 1.8 m. The only connection between the laboratory and the Cyclotron is a tunnel in form of Z, through where the irradiated target arrives to the accelerator for processing (1).

The Quality Control laboratory is situated besides the Processing laboratory. In this laboratory is carried out the radiochemical and microbiological control of the final product. The ambient pressure of both laboratories is smallest than that one of the non active areas, in order to prevent dispersion of the air contamination to the non-active areas, in case of the accidents with radioactive materials leakage (1).

In the areas of the Cyclotron installations, the following radiation sources can be found (2):

1) Cyclotron in operation:

During the operation of the cyclotron, neutrons can be generated, from the nuclear reactions that take place in the target and in the accelerator structural materials, that may interact with the own materials constituting the cyclotron and the target components, as well as of the walls and air materials of the cave ambient, forming several activation products. Additionally, there may be the scattering of the γ radiation and neutrons produced in the mentioned nuclear reactions for other areas of the building.

2) Turned off Cyclotron:

Before the turn off of the Cyclotron, the radiation levels in the accelerator cave ambient are due to the induced activity in the accelerator, target and beam lines components, as well as in Cyclotron cave materials.

Just as it happens to the Cyclotron cave, after the turn off of the accelerator, the radiation levels in the ambient of cave 5, where the system KIPROS is installed, are due to the induced activity in the walls and air of that place, as well as in the target and beam line components. It is also possible to find in that area, γ radiation, coming from the decays of the ^{123}Xe and ^{123}I.

3) Processing and expedition of ^{123}I

After a period of 6 hours, the ^{123}I produced in the Kipros system is dragged by a water stream into the hot cell of the Processing laboratory, where it is concentrated and purified in a ion exchange column. Later, it is eluted from this column, mainly, in the iodide chemical form. The resulting solution is, then, homogenised through a nitrogen gas flow and, finally, fractionated in aliquots that are sealed in glass flasks, still inside the processing cell. The sealed flasks are sent to another intermediate cell, where they are placed in a lead shield. The product, already in the shield, is, then, sent to the Quality Control laboratory where it is put into the canister. The radiopharmaceutical, already canned, is led until the Expedition area, where, finally, is conditioned in its final packing. After monitoring, it is, then sent to the hospitals.

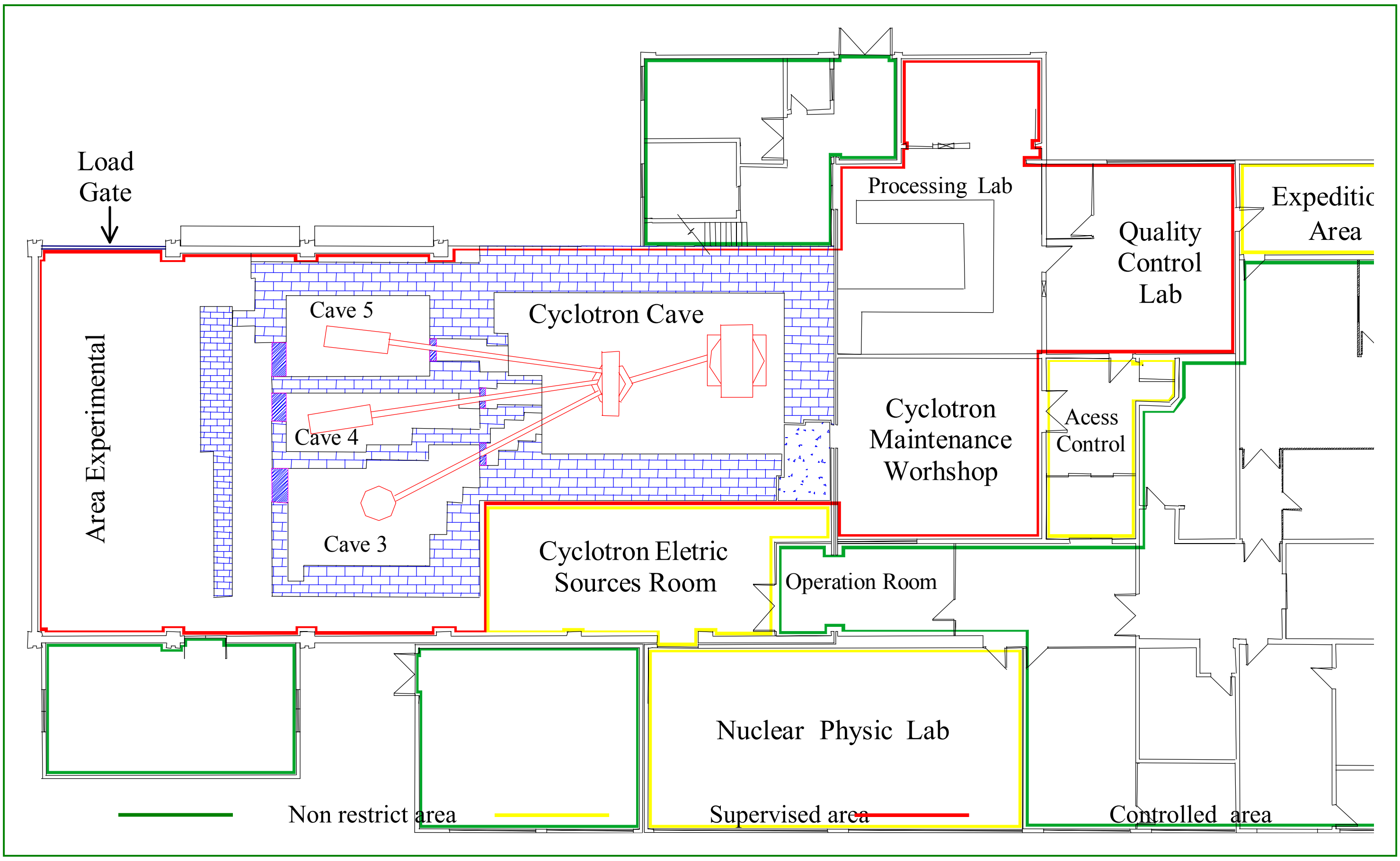


Figure 1: Layout of the main existent facilities in the 1st floor of the IEN Cyclotron installations building

3. IEN OCCUPATIONAL RADIOLOGICAL CONTROL (2, 4, 5)

IEN occupational radiological control, for the production of ^{123}I, is carried through the individual and area monitoring programs. The results of these programs are filed by a period of 30 years, when applicable.

a) Individual Monitoring Program

Everyone working in the IEN radiation areas is required to wear personal dosimeters, especially those who work at the Cyclotron installations, appropriate to their professional activities. Workers, that execute activities in areas with potential radioactive contamination, are also submitted to periodic exams, or whenever necessary, to evaluate the possibility of internal exposure. The control procedures used in personal monitoring are described below.

- External monitoring:
  - Photographic dosimeter (Film Badge);
  - Direct reading dosimeter (dosimeter pen);
  - TL dosimeter (ring);
  - Neutron dosimeter (albedo).

- Internal monitoring
  - Whole body counter;
  - Bioanalysis.

b) Area Monitoring Program

A IEN health physics staff provide all time control and surveillance for all activities in IEN radiation areas that involve radioactive materials or ionising radiation. They make frequent radiation surveys of occupied areas using radiation monitoring equipments. The area monitoring program aims to evaluate and control the radiological conditions of the IEN workplaces. This program involves the following procedures:

- Routine conditions:
  - Restricted areas monitoring through the radiation external fields measurement;
  - Restrict areas monitoring with potential of radioactive contamination being used:
    - surfaces contamination evaluation using directly and indirect methods;
    - air contamination evaluation (aerosol and iodine).

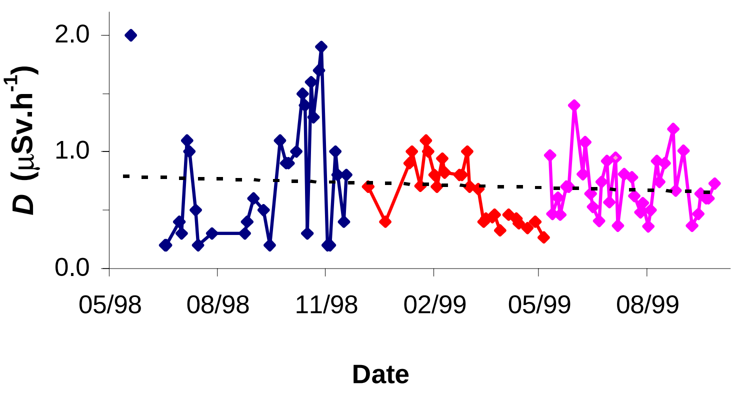
- Operational and maintenance conditions:
  - Cyclotron in Operation (I^{123} production):
    - external fields measurement of γ radiation and of neutrons.

  - Cyclotron in maintenance:
    - external fields measurement of γ radiation;
    - contamination surfaces evaluation;
    - air contamination (aerosol).

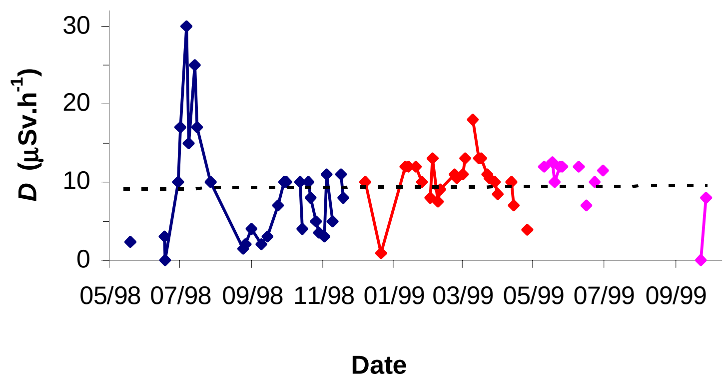
- ^{123}I Processing:
  - external fields measurement of γ radiation;
  - contamination surfaces evaluation;
  - air contamination (iodine).

4. RESULTS AND COMMENTS (2, 3)

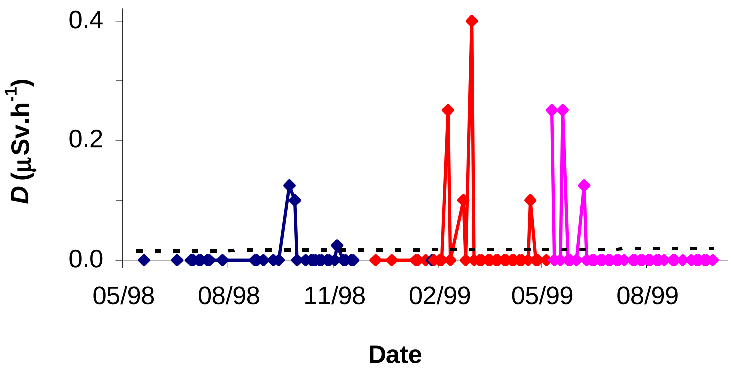
Figures 2 and 3 show dose rate values found in the main internal and external areas of the Cyclotron installations during the ^{123}I production in cyclotron operation to 10, 15 and 20 μA.



(A)



(B)



(C)

Figure 2: Neutrons dose rates *(D)* measured in the Cyclotron operator position (A); external area close to the load gate of the Cyclotron experimental area (B); and in the internal non restrict areas of the 1st floor of Cyclotron installations (C). Operation to 10 µA (blue line), 15 µA (red line) and 20 µA (purple line). Black stippled line: tendency line

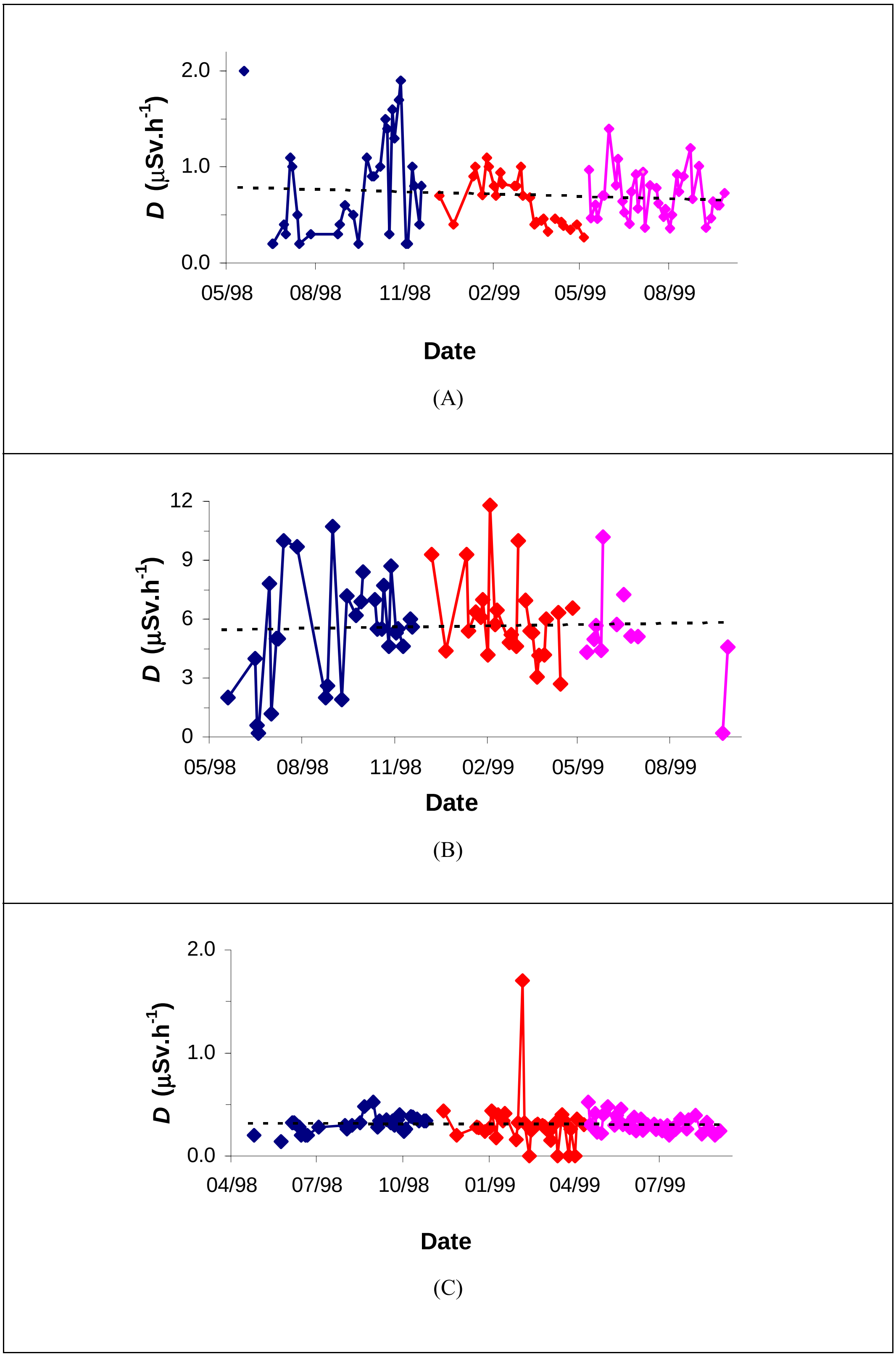


Figure 3: Gamma dose rates *(D)* measured in the Cyclotron and Kipros system operator position (A); external area close to the load gate of the Cyclotron experimental area (B); and in the internal non restrict areas of the 1st floor of Cyclotron installations (C). Operation to 10 µA (blue line), 15 µA (red line) and 20 µA (purple line). Black stippled line: tendency line

The results of the area monitoring program presented in figures 2 and 3 indicate that there was no important variation in the average values of dose rate as a consequence of the increase of the Cyclotron operation current, from 10 to 20 μA. A small tendency of increase of the neutron dose rates at the workplace of Cyclotron and KIPROS system operators (operation room) and in the external non restrict area, close to the load gate of the Cyclotron experimental area, during the operation of the accelerator, as a consequence of this current increment, can, also, be observed. On the other hand, in the same operation conditions, in relation to γ radiation, it is observed a small decrease of dose rate values assessed in the Cyclotron and KIPROS system operation room. Additionally, it is observed very pronounced sporadic variations in the values of dose rate found, for some situations, which can be explained as a result of the oscillation of the Cyclotron operation current, during the ^{123}I production.

However, the most important aspect to stand out is the dose level measured at the load gate of the experimental area. These dose values are higher than the dose limit established by CNEN for non restrict areas, namely 1 mSv.yr^{-1}, independent of the current oscillation. It was verified that these doses are a consequence of the inefficiency of the cave 5 shield, where the irradiation of the ^{124}Xe target takes place, and consequently, the nuclear reaction for the formation of ^{123}I UP too. In order to improve this situation, some improvements in the cave 5 shield has been carried out along the time.

However, it deserves to be mentioned that:

- The iodine production has been carried out, usually, under those conditions, between 00:00 and 07:00 h, out of the normal office hours in IEN, that is, 07:30 to 16:30 h. During the ^{123}I production only the workers involved in this activity stay at IEN. It is important to mention that IEN security guard is present in the Institute during the ^{123}I production, however their components aren't occupational workers at all and their workplaces are non restrict areas, where dose rates are comparable with local background.
- The external area close to the load gate is a unoccupied place (no personnel's permanency), being used, some times, at special occasion, for the discharge of materials, with turned off accelerator;
- The gamma dose values at the internal non restrict areas of Cyclotron installations are small and of same order of the local background. In those situations where neutron dose rate were observed in same places, the values present a high unexplained variation;
- Dose rate measurements vary, considerably, from one production to another. These variations are, possibly, related to the variations of the Cyclotron operation current observed during ^{123}I production;
- Even with the increase of the cyclotron current, the measured total dose in the accelerator operation room, tends to decrease. Although the operation room is classified as a non restrict area and the increment of the radiation levels verified during the irradiations in this place, the estimate of the total annual dose, that could be received by a person that there stayed during the whole time of irradiation (considered 5 hours) it would be about 0,5 mSv. It should be observed that, in the operation conditions of IEN Cyclotron, reported in this work, only IEN workers involved in the ^{123}I production stay in the operation room. These workers during their professional activities are monitored properly with personal dosimeters, such as: thermoluminescent dosimeters (rings), film badge, albedo dosimeters and dosimeter pen.

Occasionally, it was found ^{123}I in air at the Processing laboratory, at low levels, maximum of 6 Bq.m^{-3}. In some circumstances, radioactive contaminations were also found in surfaces of the Cyclotron installations controlled areas, specifically, in the Processing laboratory (^{123}I) and in the Cyclotron maintenance workshop (^{65}Zn), however also at small levels.

Considering the results of the program of individual monitoring,, it was verified that the workers involved in the ^{123}I production of IEN:

- Didn't receive important whole body external doses. Basically, these doses values were below the detection limit of the film Badge, that is 20 mSv. Exceptionally, in 18 cases of a total of 714 observations, external whole body doses not higher than 30 mSv were found. However, the total external body whole dose resultant of these exposures didn't exceed 100 mSv.yr^{-1} per worker;
- Some extremity exposures were received, about 54 cases of a total of 640 observations. These doses were lower than the dose limit for extremities established by CNEN (500 mSv.yr^{-1}) and didn't exceed 230 mSv.yr^{-1} per worker;
- External whole body doses registered for some workers can be explained as resultants of maintenance activities in the processing cell or accelerator, as well as other activity carried out by radiation protection workers not related to ^{123}I production;
- Doses due to neutrons were below the detection limit of the neutron dosimeter, that is, 20 mSv;
- Important incorporations with radioactive materials present, occasionally, in the air or in the work surfaces of the restricted areas of the Cyclotron installations (products of activation of the Cyclotron components or ^{123}I) were not observed. The resulting doses of this possible exposition pathway were not important either.

5. CONCLUSIONS (3, 4, 5)

A radiation protection system should be based on three fundamental principles: justification of practice, optimization of protection and limitation of individual exposures. It should be noted that the first two are the most essential. The third principle, limitation of exposures, supplements the use of the justification and optimisation procedures, being in that way, an individual guarantee because the distribution of the benefits and costs of a certain practice, throughout the population, is not uniform.

According to the concepts exposed above, as well as the results obtained in this work, it is recommended that an optimisation study is accomplished, especially concerning an effective project to the cave 5 shield, in order to verify the application of the ALARA principle to the existing work conditions. In this sense two questions should be answered: - Although the measured occupational doses are low, are they justified? - Is it possible to reduce them, decreasing the risks of the stochastic effects that can be induced by the ionising radiation? In other words, the radiation protection project was optimised?

It is also suggested, that the areas classification of the Cyclotron installations is revised, especially, the external area close to the load gate of the experimental area. This area during the ^{123}I production should be delimited (restricted access).

6. REFERENCES

1. **Peres, S. S**., “Estimativa das Implicações Radiológicas Ambientais Resultantes da Operação do Instituto de Engenharia Nuclear na Ilha do Fundão no Rio de Janeiro”, tese de mestrado, IBCCF/UFRJ, RJ, julho, 1999 (In Portuguese).
2. **Peres, S. S. , Da Rosa, L. A. R and et al**, "Sistema de Radioproteção e de Gerência de Rejeitos Radioativos do IEN", internal document, maio, 1999 (In Portuguese).
3. **Comissão Nacional de Energia Nuclear**, “Diretrizes Básicas de Radioproteção", CNEN-NE-3.01, RJ, 1988 (In Portuguese).
4. **International Atomic Energy Agency**, " Radiological Safety Aspects of the Operation of Proton Accelerators ", Technical Reports Series no. 283, Vienna, 1988.
5. **International Atomic Energy Agency**, " Radiation Protection in Occupational Health ", Safety Series no. 83, Vienna, 1987.