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Overview of the technological enhancement of natural radiation in the Brazilian non-uranium mining industry

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Abstract. The mining and milling of ores with significant amounts of uranium and thorium associated to the main ore has the potential to pose undue health risks to members of the general public and workers. In order to assess the status of this problem in the Brazilian non-uranium mining industries a comprehensive investigation project has been undertaken. The adopted methodology was based on the detailed analysis of each investigated industry operational flowplan, mass balance calculations, risk assessment (operational and post-operational scenarios taken into account) and environmental management principles. This papers addresses the main issues arising from the investigation effort, reports the most relevant conclusions and states the future studies to be implemented. It could be observed that these industries have the potential to cause relevant radiological impacts and must be regulated/controlled as to avoid these problems.

1. INTRODUCTION

The mining and milling of ores with significant amounts of uranium and thorium minerals may lead to the radiological environmental impact of nearby water streams, soils and aquifers. These impacts may be observed during the operational phase of the facilities as well as after the facility close out, if no proper reme diation is carried out.

During the life of the mining operation, and after the plant close out, the tailings and waste dumps that have been formed are constantly subjected to weathering. Upon close out, weathering process, although unchanged externally, may be considerably different within these areas. The rate of leaching of pollutants from the solid material will change with time and be dependent on several factors that will regulate the rate of pollutant release from these sources.

In the assessment of the potential impacts caused by these installations, monitoring programs, envisaged to detect increases in radioisotopes concentrations in environmental samples like superficial water, sediments, soil, vegetables, fish, etc, used to be carried out. This strategy lacks of consistence once it can not cope with impacts in future scenarios. In addition to this, an overall understanding about the amount of radionuclides disposed off with the wastes, radioisotope concentrations in by products that could be used in a wide variety of situations remain unknown.

Another important issue to be emphasised is the fact that these industries, despite the potential radiological impacts that they may cause are not regulated, i.e., they are not licensed as practices, nor controlled by the Regulatory Authority. However, as it will be showed in other paper in this book [1] undesirable exposures of members of the public and workers may result from their operation.

Several are the facilities in Brazil which present potential radiological problems caused by the presence of uranium and thorium associated to the main ore like coal, phosphate, niobium, gold, tin, copper and lead mining and milling facilities. Exposures of members of the general public will also depend, on a very large extent, on the regional characteristics associated to a particular facility location. This is of special concern in a country like Brazil where the development stages of the different regions are not the same, leading to social and economical patterns completely different from region to region.

Presently, a broad research program concerning the assessment of the extent of the technological enhancement of natural radiation caused by non-nuclear mining industries is being developed by the Institute of Radiation Protection and Dosimetry (IRD). The questions to be answered by the investigation program are:

1. Do the non-uranium mining industries have the potential to cause environmental (occupational) radiological impacts?
2. If yes, what are the extent of these impacts?
3. How to deal with the assessment of the problem?

In order to deal with these questions a working strategy was designed. The program relies on a pragmatic approach to identify, characterise and implement remedial actions, whenever necessary, to the sites affected by the operation of these installations.

2. WORKING METHODOLOGY

Once mining operations begin a mineral deposit is disturbed in such ways that many accessory elements and compounds are redistributed in markedly different patterns and concentrations from those that would occur by natural geologic processes. Redistribution of elements can occur during any of the steps involved in the material cycle.
During mining, various elements and minerals are made more accessible to environmental exposure. Waste rock or overburden is generally removed and piled away from the working area; some may be used in road and site construction but most goes into spoil piles.
With most raw materials, the mined ore must be beneficiated to separate the desired minerals from the waste material. This is carried out first by comminution to a size at which the minerals are liberated from each other, followed by concentrating mechanism for the desired ore mineral. Beneficiation technologies utilise physical or surface chemical means for separating minerals, and consequently the minerals are not changed chemically at this processing stage. Generally, chemical processing is required to convert the desired ore minerals into the chemical form suitable for use. Elements in the metallic state, for instance, may accompany the metal whereas elements forming oxides may be separated and discarded as slag. In addition, volatile constituents can separate in gaseous phase and be vented to the atmosphere. In the processing of certain ores or concentrates, leaching with aqueous solutions may be a major separation step. Here, element redistribution follows from the discard leach liquid and any entrained leach residue. During the chemical processing, there may be an opportunity to recover by-products because of the chemical changes induced in the material at this stage. Recovery can often be achieved by modification of specific process in the flowplan,
From the previous discussion it is clear that a precise understanding of the potential impacts caused by a mining industry shall begin by the detailed analysis of the operational flowplan. Concentration of radiosiotopes in the different generated wastes and effluents must be determined, and multiplied by the flux that each waste is produced in the operational process. This will allow (by means of mass balance calculations) the exact knowledge of the amounts of radioactivity involved on each step of the operational process. It will also allow for a better waste management system if it appears that the adopted strategy is likely to cause undue environmental impacts. Moreover, the likelihood for radioisotopes (and any other potential pollutant) mobilisation from the wastes must be addressed. This may be of key relevance in terms of future contamination of aquifers and superficial waters. External exposures as well as radon exhalation shall also be determined in case of unrestricted use of depositional areas are intended in the future. The impacts associated to the use of by-products (e.g., coal ashes and phosphogypsum as building materials and phosphogypsum as fertiliser in the agriculture) shall also be assessed. Finally, the adoption of remedial actions may be suggested. This may include modifications of the waste disposal strategy, alteration of the processing route, economical recovery of by-products and so on.
The adopted methodology consisted of:

(1) Analysis of the operational process of the industry;
(2) Sample collection (and radionuclide determination) from specific points of the operational process;
(3) Mass balance calculations and determination of the total activity accumulated in each step of the operational process and those deposited as wastes. The amount of radioactivity in the final product is also considered;
(4) Determination and quantification of the potential mechanisms involved on the mobilisation of the pollutants from the waste deposit areas (tailings dam/pile and waste dumps);
(5) Estimation of the environmental compartment concentrations associated to effluent releases;
(6) Risk assessment concerning the operational impacts of the facility and those associated to the facility close-out;

(7) Examination of the need for remediation concerning the contaminated area (waste deposit areas) for irrestricted use.

3. RESULTS

Table 1 shows the activity concentrations determined in samples from the run of mine collected in some industries.
It can be seen that, with the exception of gold ore sample, the radionuclides are not in equilibrium. The highest concentration of uranium was found in the Niobium II industry. In the other Nb-industry Th showed a higher concentration than uranium. The phosphate II industry showed higher concentrations of the natural radionuclides in comparison to the phosphate I industry.
The total inventory of radionuclide fluxes among the different industries is shown in table 2. These fluxes are obtained by multiplying the different radionuclide concentrations in the run of mine by the total mass of the run of mine entering the process. The highest values are observed for the phosphate industries and one of the investigated niobium industries.

Table 1. Activity Concentrations in some Ores of the Investigated Industries

Mining Industry	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Niobium I	930 +/- 62	805 +/- 97	1,330 +/- 231	6,390 +/- 655	5,176 +/- 716
Niobium II	4,550 +/- 300	3,390 +/- 319	7,890 +/- 1,121	904 +/- 94	2,040 +/- 312
Coal	359 +/- 24	457 +/- 57	754 +/- 167	33+/- 3	68 +/- 49
Gold	114 +/- 8	136 +/- 17	169 +/- 104	49 +/- 5	< 42
Phosphate I	114 +/- 8	330 +/- 41	800 +/- 172	204 _+/- 21	350 +/- 123
Phosphate II	880 +/- 58	700 +/- 85	990 +/- 192	753 +/- 77	1,550 +/- 255

Obs: Results refer to one composite sample. The analysed sample was composed by five sub-samples taken during five consecutive days by the mining operators (according to their quality control protocol).

Table 2. Fluxes of Radioactivity Associated to the Investigated Mining Industries

Industry	Total Radioactivity Flux (Bq/year)
Niobium I	1.8×10^{13}
Niobium II	6.4×10^{12}
Coal	1.5×10^{10}
Gold	3.3×10^{11}
Phosphate I	2.0×10^{13}
Phosphate II	2.7×10^{13}

The redistribution of the radionuclides along the operational process will cause that some of the radionuclides will be leaving the process in higher amounts associated to a specific waste. The redistribution will depend on the type of the operational process and on the mineralogical composition of the ore. Table 3 shows the redistribution of the radionuclides in the operational process of two of the investigated niobium industries.
By far, the highest concentrations are observed in the slags of both Nb industries and also in the barium sulphate residue of the Niobium I industry. However, the mass flux involved in these steps of the operational process (0.85 to 3.0 t/h) are very small if compared to the mass flux of the other steps. It means that these wastes may be disposed of in separated areas with proper preparation of the disposal area. Shielding against gamma radiation exposure shall be provided. Mobilisation of radionuclides from these wastes (with the exception of barium sulphate residue) will not be a major issue, once this material is very refractory. On the other hand, ground water bellow the barium sulphate disposal area shows (in the case of the Niobium I industry) Ra isotopes (226 and 228) concentrations as high as 5.0 Bq/L. These waters need to be pumped and treated with $CaSO_4$ to precipitate the dissolved Ra isotopes before being released to the open environment.

Table 3. Radionuclide Redistribution in the Different Steps of the Operational Processes of two of the investigated Niobium Industries

Operational Process Step	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Niobium I					
Magnetic Separation (22 t/h)	(1,091+/-74) 2.0×10^{11}	(340+/-43) 6.3×10^{10}	(720+/- 63) 1.3×10^{11}	(863 +/- 90) 1.4×10^{11}	(5,176+/-716) 6.9×10^{11}
Disliming (17 t/h)	(1,265+/-83) 1.8×10^{11}	(1,381+/-158) 2.0×10^{11}	(2,029+/-311) 2.9×10^{11}	(8,995+/-924) 1.3×10^{12}	(5,278+/-720) 7.7×10^{11}
Floatation (90 t/h)	(770+/-50) 5.8×10^{11}	(990 +\- 120) 7.5×10^{11}	(2,440 +\- 360) 1.8×10^{12}	(3,100 +/- 320) 2.3×10^{12}	(1,440 +\- 243) 1.1×10^{12}
Slag (3 t/h)	(23,138 +/1520) 5.8×10^{11}	(3,290 +\- 313) 8.3×10^{10}	(2,400 +\- 355) 6.1×10^{10}	(117,600 +\- 12,000) 3.0×10^{12}	(20,200+\- 3,606) 5.1×10^{11}
Barium Sulphate (0.6 t/h)	(42 +/- 2) 2.1×10^{08}	(26,250+/- 3,100) 1.3×10^{11}	(7,400+/- 1,000) 3.7×10^{10}	(43 +/- 1) 2.2×10^{08}	(19,7300+/- 27,600) 9.9×10^{11}
Niobium II					
Magnetic Separation (13 t/h)	(843+/-56) 9.2×10^{10}	(941+/-112) 1.0×10^{11}	(1,730+/-276) 1.9×10^{11}	(293+/-30) 3.2×10^{10}	(315+/-120) 3.4×10^{10}
Disliming (8 t/h)	(6,700+/- 145) 4.5×10^{11}	(6,900+/-404) 4.6×10^{11}	(6,180+/-860) 4.2×10^{11}	(1,754+/-180) 1.2×10^{11}	(3,080+/-439) 2.1×10^{11}
Floatation (43 t/h)	(4,985+/- 327) 1.8×10^{12}	(3,400+/-320) 1.2×10^{12}	(7,530+/- 1,065) 2.7×10^{12}	(1,040+/-107) 3.8×10^{11}	(1840+/-289) 6.6×10^{11}
Slag (0.83 t/h)	(34,819+/- 2,294) 2.4×10^{11}	(5,160+/-393) 3.6×10^{10}	(435+/-132) 3.0×10^{09}	(167,700+/- 1,713) 1.2×10^{11}	(6,450+/-900) 4.5×10^{10}

Obs: Concentration results in parenthesis

In both Nb industries the highest radioactivity mass fluxes are associated to the floatation step. However, the radionuclide concentrations in these wastes are different between both industries. ^{238}U concentration in the floatation wastes of the Niobium II industry is about 5,000 Bq/kg. The total flux of ^{238}U in these wastes is 1.8×10^{12} Bq/year which corresponds to 145 $t_{U\text{-}238}$/year. It means that 145 tons of ^{238}U are being lost as waste in the operational process. The obvious conclusion is that an economical feasibility study concerning the recovery of uranium from these wastes shall be commenced. This idea is even more plausible if one takes into account that the average production of the only uranium mining and milling facility to operate in Brazil until now was about 100 t_{U3O8} per year. The estimated production of a future Uranium mining and milling facility (located at Bahia state – Lagoa Real Project) is 300 t_{U3O8} per year.

Table 4 summarises the potential uranium recovery in some of the investigated mining industries. The data are very preliminary ones, however they have the potential to reveal that the recovery of uranium from the wastes of non-uranium mining industries may constitute an important source of uranium in the future.

One of the mining industries that is generally associated to major radiological problems is the phosphate industry. In Brazil, the total installed capacity for phosphate rock concentrate production is about 4.8×10^6 ton/year, being the mean national production about 3.4×10^6 ton/year. Differently from what is observed in the rest of the world, phosphates in Brazil are mainly associated to igneous rock. It is reported that 4 to 5 tons of phosphogypsum are generated in the production of 1 ton of phosphoric acid. It is estimated that by the year 2,000 about 80 million tons of phosphogypsum will be stocked in the country.
Table 5 shows radionuclide concentrations found in some phosphogypsum samples.

Table 4. Potential ^{238}U recovery in Different Industries Non-Uranium Industries

Mining Industry	Potential for Uranium Recovery (tons of ^{238}U per year)
Niobium I – industry	88
Niobium II – industry	200
Phosphate II – industry	400

Table 5. Radionuclide Concentrations is Some Phosphogypsum Samples

Radionuclide	n	Minimum Value	Maximum Value
^{238}U	62	5.52	116
^{226}Ra	62	28	319
^{210}Pb	31	20	513
^{232}Th	61	12	268
^{228}Ra	62	20	513

Source: Costa e Silva (1998) [3]

The wide range of concentrations for the individual radionuclides is caused by the different ore composition in respect to those radionuclides (see table 1). It may be said that the phosphogypsum is the most relevant aspect in terms of environmental radiological impacts associated to the phosphate cycle. It is reported that radon exhalation from the phosphogypsum piles may be represented by a generic value of 3,600 $Bq/m^2/h$, corresponding to a total input of 25 TBq/year. In addition to this, one has to consider the potential problems of groundwater contamination (especially by Ra isotopes) and the use of phosphogypsum for different purposes. Two works connected to this project have assessed the potential impacts of the use of phosphogypsum as building material and as a fertiliser in the agriculture. The first one [2] demonstrated that the use of phosphogypsum as building material would not cause undue exposures to members of the public in the considered scenario and that ^{220}Rn inhalation would account for 80% of the total dose. The second one [3] assessed the individual effective dose related to the consumption of different agricultural products cultivated with phosphogypsum being used as a fertiliser. The estimated dose values, for the examined situations, were lower than 2.3 μSv/year. A week point of this work was the fact that the collective dose associated to the use of phosphogypsum as fertiliser was not assessed. An alternative to the sulphuric route, to avoid the generation of phosphogypsum, is the nitric or chloridric route, as represented by equation 1.

$$\mathbf{Ca_{10}(PO_4)6F_2 + 14\ HCl + 3\ H_2O <=> 3\ Ca(H_2PO_4).2H_2O + CaCl_2 + 2\ HF}\ \text{(equation 1)}$$

In this case, U, Th and Ra isotope dissolve and enter the waste stream. ^{226}Ra formed together with $CaCl_2$ (12 Ci_{Ra}/year per 1,000 ton_{ore}/day) may be precipitated with $BaSO_4$. Uranium can be recovered with OPPA or DEHPA-TOPO and the rare earths may also be recovered. In addition to these advantages, fertilisers relatively free of U, Ra and Th will be produced.
A last point to be observed is the exposure of workers to radon in the working places (especially in underground mines). As it will be showed in other contribution in this book [3] ^{222}Rn concentrations in the range of 10^4 to 10^5 Bq/m^3 were observed in some of the investigated mining sites.

Dose limits to be applied to these workers shall not be the same as those applied to workers in an uranium mining facility. As a consequence these workers must be faced as members of the general public and whenever the effective dose have the potential to exceed the value of 1 mSv/year, remedial actions shall be applied.

4. CONCLUSIONS

The investigation of several non-uranium mining and milling facilities revealed that radionuclides are remobilised and redistributed in the generated wastes reaching activity concentrations orders of magnitude higher than those observed in the run-of-mine material. The precise knowledge of the wastes composition and fluxes is of extreme relevance in aiding the adoption of adequate waste management strategies. As a general trend it must be proposed that the potential radiological problems are mostly associated to the solid wastes. The habitation of these areas and the use of these materials for house construction may lead to the undue exposure of members of the general public to radiation (external gamma exposure and radon inhalation). Operational impacts (by means of liquid effluent releases into the environment) were not found to be relevant. This was due to the fact that the chemical treatment applied to these effluents, to reduce the emissions of the non-radioactive pollutants, was effective to reduce the effluent concentrations of the radioactive ones. In as much, in some of the investigated industries process waters are recycled minimising the liquid effluent release into the environment. Natural processes like acid drainage generation may also mobilise significant amounts of radionuclides (mainly uranium) and must be properly managed. Changes in the operational process may reduce environmental problems and favour the economical recovery of products of interest, e.g., uranium and rare earth elements.

5. FUTURE STUDIES

Future studies to be developed comprise:

(1) Assessment of the present extension and situation of dump sites in all the mining industries where uranium and thorium occur associated to the main ore in significant amounts;
(2) Experimental and modelling studies on radionuclide migration from deposited wastes and assessment of the contamination of groundwater in the influence area;
(3) Development of methods to immobilise radionuclides in the wastes;
(4) Assessment of radon exhalation from dumpsites and experimental tests to reduce the gas exhalation (with emphasis on phosphogypsum piles);
(5) Priorization of studies involving radon exposure in working places (especially in underground mines).

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