SURVEYING AND ASSESSING THE HAZARDS ASSOCIATED WITH THE PROCESSING OF URANIUM

Jan Kruger

Isotopes and Radiation Division, Atomic Energy Board, Private Bag X256, Pretoria 0001 South Africa

INTRODUCTION

The mining and milling of uranium covers the physical mining process, followed by the extraction of the uranium to a final concentrated uranium compound, usually ADU or oxide. Further processing and conversion of the uranium to other chemical forms or to the metal is normally associated with the nuclear industry and such plants are also located separately from the mining industry. It is generally accepted that exposure to radon constitutes the main hazard in mining operations whereas exposure to dust constitutes the main hazard in milling operations (1). The monitoring of radon and exposure during mining operations have received, and is still receiving, extensive attention. Uranium processing plants associated with the nuclear industry are controlled according to the norms applicable at nuclear installations, including full radiological surveying and monitoring. At uranium plants associated with the mining industry it is found that radiological surveying is infrequent if done at all. Assessment of the working environment is based on a limited air sampling program. Biological monitoring by means of urinassay, sometimes performed on selective sampling of the work force is used to assess the exposure of the work force (2, 3). Reliance is placed on good housekeeping and visual observation is used to judge the effectiveness of measures taken to prevent the spread of material.

The toxicology of uranium and the hazards associated with uranium processing have been extensively documented. A comprehensive résumé on occupational health experience with uranium was presented at a conference in Arlington, Virginia, USA in 1975 (4). Exposure due to the uptake of natural uranium depends on the solubility of the compounds in the body fluids. For soluble compounds the chemical toxicity to the kidney limits the uptake of material to 2,5 mg per day by inhalation, or 150 mg in two days by ingestion (5). For insoluble compounds the radiation exposure to the lung becomes the limiting factor. The permissible lung burden for long-term exposure can be calculated to be 26 mg (6). Since the original method of monitoring urinary excretion for assessing exposure due to the uptake of uranium (7), bioassay methods have been extended (8). There is, however, a lack of information on epidemiological data on human exposures.

This paper reviews the involvement in terms of radiation protection of several natural uranium processing facilities over a number of years in South Africa. The extent of the involvement differed appreciably for different facilities, and also changed in time. An effort will be made to use the large amount of data available from surveys to relate acceptable environmental conditions to exposure standards and to indicate how contamination control results in a de=

crease of internal exposures.

THE SURVEYING OF PROCESSING FACILITIES

The survey information used in this analysis consists of routine static air sampling data and routine surface contamination smear data. Special survey data obtained during abnormal operating conditions or accidental releases to evaluate such situations, have not been included. The longer term consequences of such abnormal conditions are however reflected in the routine data. The frequency and extent of the routine surveys varied from time to time and were determined by prevailing circumstances. It is assumed that the routine data are a true reflection of the environmental levels over a period of time. It must be appreciated that due to such uncertainties as the representativeness of the static air sampling data, the selected frequency of surveying, and accidental releases, this assumption may not be true at any particular time.

The only method available for assessing personnel exposure over the period under review was bioassay by urinalysis. The routine urine sampling results are used as an indicator of personnel exposures. No effort is made to relate this to actual dose commitment, chemical or radiological. Where personnel exposure could be attributed to accidental releases which were not reflected in the routine surveys, the data were excluded from the analysis. As in the case of the survey routines, the frequency of urine sampling was determined by prevailing circumstances.

The collection of survey data refers to three different sets of circumstances:

Study A (9): In the early sixties a facility was operated for pilot plant work on the extraction of uranium from ore, and conversion to oxides, fluorides and metal. In 1964 health physics coverage was provided on a part-time basis, and this evolved to full-time coverage from 1967 onwards. Whereas the previous period included infrequent surveys, the latter period included regular routine surveys. A routine urine sampling program was undertaken from the beginning of operations, with a sampling frequency of once a month. Urinalysis results are available for the whole period under review.

Study B: In the middle seventies several pilot plants for experimental work on the conversion of uranium to oxides, fluorides and metal started operating. Health physics assistance was provided from the design stage of the plants and full coverage and surveys were provided from the onset of operations. Frequent and regular routine surveys were initiated. A monthly urine sampling program was initiated. As circumstances dictated and the work load increased, the frequency of urine sampling was increased to once a week at some of these plants. The feed materials to these plants are normally ADU.

Study C: Since the late sixties a number of surveys have been made on request at several uranium plants associated with the mining industry. These surveys do not constitute anything like routine coverage and represent infrequent samples taken at different plants. During any one survey an effort was made to obtain representative samples of the working environment of that particular plant. The surveys cannot be considered comprehensive as only a few of the existing plants were covered. Although routine urine sampling programs are

in existance, the results were not available to the author. The operations at these plants include extraction of uranium from the ore and concentration to ADU or calcining to the oxide.

ASSESSING THE HAZARDS ARISING FROM URANIUM

Considering the uranium series it is seen that although the parent emits only weak gamma-rays, and in itself presents a small external exposure risk, the daughter products do present an external exposure risk. During extraction of uranium the daughter products (with radium-226 the main hazard) are separated and these can be present in the waste stream, collected somewhere in the process, and may even to a smaller extent be carried through the product stream. After separation the first two daughters of uranium build up again to reach 50 % of equilibrium in about 24 days. The alpha contamination from uranium as well as the beta contamination from these daughters should therefore be considered in any uranium processing plant. The exposure due to external radiation will not be considered in this paper.

The working environment is judged against Derived Working Levels (DWL). For airborne material the (MPC) for uranium for a 40 hour week is used (10), namely 7×10^{-11} Ci/m^3 or 200 µg/m^3 (11). For surface contamination the following DWL's were used (12):

Uncontrolled areas : 10^{-5} µCi/cm^2 for alpha and 10^{-4} µCi/cm^2 for beta.

Controlled areas : 10^{-4} µCi/cm^2 for alpha and 10^{-3} µCi/cm^2 for beta.

The relaxation by a factor of 10 for controlled areas was not used.

The exposure of personnel was judged against two urinary levels of uranium. The first level is considered as the minimum significant level of 10 µg/ℓ and serves to indicate a positive exposure. The second is the maximum permissible level of 100 µg/ℓ for uranium excretion.

RESULTS

Study A: For the purpose of comparison two periods are considered and totalled, i.e. before 1967 and after 1968. The surface contamination levels as they were found to exist in 1967, are used to describe conditions prior to routine health physics coverage, and levels at the beginning of 1969 to describe conditions after contamination control had been implemented. A survey consisted of 20 smear points and 8 air sampling points. A total of about 1 800 urine results over the whole period was used to describe personnel exposures. The results are tabulated in Table 1. The percentage of smears (either alpha or beta activity) that exceeds the DWL for controlled areas is shown together with the percentage of urine samples exceeding 10 µg/ℓ, those exceeding 100 µg/ℓ and the maximum average alpha air activity. (Subsequent to the publication of reference 7 a collection efficiency of smear sampling of 10 % has been defined as realistic. The smear results in reference 7 has been corrected accordingly).

TABLE 1

Period	% smears > DWL	% urine > 10 µg/ℓ	% urine > 100 µg/ℓ	Max. av. air activity in DWL
Before 1967	10	45,6	0,4	0,43
After 1968	0	9,3	0,1	0,1

Study B: The available data was considered for periods of time according to the sampling periods for urinalysis, i.e. for monthly urine sampling a period of one month was defined, and for weekly urine sampling a period of one week was defined. The data was furthermore divided into two groups as determined by the smear results over the period considered:

Group 1: All smear results during the period defined show levels less than DWL for controlled areas (alpha or beta contamination).

Group 2: During the period defined smear results showed one or more values in excess of the DWL for controlled areas (alpha or beta contamination).

Data from a total of 3 400 urine samples, 40 000 smear samples and 1 600 air samples are used in the analysis.

The results are given in Table 2. In each group the percentage of smear samples in excess of DWL (uncontrolled areas for group 1 and controlled areas for group 2), the percentage of urine samples in excess of 10 µg/ℓ, the percentage of urine samples in excess of 100 µg/ℓ, the percentage of air samples in excess of the appropriate DWL, and the maximum average air activity is given.

TABLE 2

Group	% smears > DWL	% urine > 10 µg/ℓ	% urine > 100 µg/ℓ	% air > DWL	Max. av. air activity in DWL
1	9,9	9,3	0,1	1,8	1,8
2	3,3	19,9	0,7	4,2	15,3

Study C: The results from the individual surveys were totalled in two groups. It was found that in these plants access to stages subsequent to the precipitation of uranium (be it mechanical or chemical) should be and, in some cases, was controlled. All stages preceeding the precipitation of uranium (i.e. leaching, gravitational settling, extracting and ion exchanged) can be considered uncontrolled areas. In practice, analytical laboratories are also in uncontrolled areas. The results are therefore divided into:

Group 1: Uncontrolled areas

Group 2: Controlled areas

A typical survey consisted of 40 to 60 smear samples and 10 to 16 air samples.

Results are given in Table 3. For each group the percentage of smears showing levels in excess of the appropriate DWL, the per=

centage of air samples in excess of the DWL and the maximum air contamination level is given.

TABLE 3

Group	% smears > DWL	% air > DWL	Max. air activity in DWL
1	14	0	0,1
2	22	30	3 000

The highest air activities were measured at specific points where, during normal operations, release of uranium was taking place. These points were defined as : ADU filtration at installations where these were not enclosed, oil-fired calciners and drum loading areas. Excluding these particular samples, the highest air activity was found to be 0,3 DWL.

CONCLUSIONS

It must be emphasized that the conclusions are of a general nature. A number of complicating factors are not represented. At different plants the uranium compounds differed largely and the uptake and excretion patterns differed widely. Effects of abnormal releases and of undesirable operating practises were not defined, although they may have been included in the routine survey results.

The survey data totalled over periods of bioassay show the relationship of higher exposures during larger contamination of the working environment. Air sample data, especially grab sampling, do not show a clear relationship.

In study A the improvement in exposure after reduction of surface contamination levels correlates well with the surface smear results. The air sampling, performed on a grab basis, does not correlate to the same extent. In study B higher exposures correlate clearly with higher contamination levels of the working environment. The air sampling was more continuous and therefore more descriptive of the air contamination than in study A. In total they also show the correlation of higher average air samples to higher exposures. However, over shorter periods they do not necessarily correlate with higher exposures - see for example the maximum averages recorded in the two groups. It is believed that this does not indicate exposure by ingestion in stead of by inhalation, but rather points to the unrepresentativeness of static air sampling.

The Derived Working Levels used for surface contamination are illustrated to closely resemble levels that give rise to significant personnel uptakes, as judged from urinalysis. The use of an in-vivo method of dosimetry is under way to further investigate and assess exposure of personnel. It is, however, concluded that in the light of uncertainties in personnel dosimetry, the lack of epidemiological data and to keep exposure to as low a level as is reasonably achievable, the DWL's above are realistic for control on personnel exposures. The relaxation by a factor of 10 is not advisable.

Although a DWL for uncontrolled areas (which is equivalent to 3 mg/100 cm^2 for uranium) is considered to give a visible indication of uranium contamination, study C shows that visual control is not truly representative of the control of material, and accordingly also not of the risk of exposure involved. Infrequent grab air sampling only is not adequate for assessing the hazard. Surface contamination surveys, together with frequent air sampling, are required for uranium processing plants. Proper area control and personnel access is indi= cated.

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