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AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT

LUCAS HEIGHTS RESEARCH LABORATORIES

PROGRESS REPORT

FOR

ENVIRONMENTAL SCIENCE DIVISION

JULY 1981 - JUNE 1982

Chief: D.R. Davy

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FOREWORD

The dislocation caused by both the transfer of staff from AAEC to CSIRO and the co-location of the AAEC and some CSIRO research activities at the Lucas Heights Research Laboratories was significant during the year. Three sections of the Division were relocated, and in some cases this meant premature termination or delayed commissioning of some long-term experiments.

The reallocation of staff and functions between CSIRO and the AAEC affected the Environmental Science Division in the following ways:

- The Chemical Engineering Section of the AAEC Chemical Technology Division was not, as was most of the rest of that Division, transferred to CSIRO; it is now part of the Environmental Science Division.

- The responsibility for low-level radiochemical analyses was acquired, but only one of the staff members involved in the work was transferred to the Division.

- Chemical quality assurance for the reactor HIFAR and the Effluent Control Laboratory became a divisional responsibility through transfer of the professional supervisor, but the routine analyses are still done by the technical staff who are now CSIRO officers.

- The responsibility and staff for laboratory animal production, together with the project on total body irradiation of mice (Sections 5.4 and 5.5 of AAEC/ES-PR 1981), were transferred to the AAEC Isotope Division.

- Two technical staff were transferred to CSIRO.

- Six officers - three professional, three technical - were transferred to the Environmental Science Division from the AAEC Nuclear Fuel Cycle and Power and Energy Assessment Units.

Under the reporting system now adopted by the AAEC, the division with prime responsibility for an inter-Divisional project reports on its progress. Thus the work done by Environmental Science Division on the leaching characteristics of SYNROC appears in the progress report from Materials Division.

CONTENTS

1. CHEMICAL ENGINEERING

1.1 Uranium Mining and Milling

1.1.1 Leaching of uranium ores

Leaching of Mary Kathleen ore

(R.J. Ring)

Mary Kathleen Uranium Ltd (MKU) sponsored research to determine the reasons for excessive acid consumption in the mill leaching circuit. Earlier work involved laboratory tests on a single composite ore (No.1) supplied by MKU. This year, the research program was extended to examine the leaching characteristics of a second MKU composite (No.2). The mill was also visited to assess the operation of the leaching circuit. Following these visits, a plant trial was carried out to examine changes in leaching conditions suggested from AAEC test work.

The leaching characteristics of composites Nos.1 and 2 were considerably different. For the same leaching conditions, residue grades for composite No.2 were higher, even though acid requirements were increased by 10-15 kg t^{-1} ore. The higher acid consumption for composite No.2 was mainly attributable to its oxidant demand, which was about three times greater than for composite No.1. Despite these differences, acceptable uranium extractions were obtained for both ores at the target acid consumption of 65 kg t^{-1} ore.

The experimental program showed that, to achieve maximum efficiency, the leaching circuit must be operated within a fairly narrow pH range at a relatively low redox potential. Subsequent plant visits enabled a number of areas to be identified as being in need of close attention during the operation of the mill leaching circuit to achieve the required conditions. A short plant trial achieved very promising results and confirmed the conclusions of AAEC laboratory tests. Since the trial, the leach circuit operating procedures have been modified to alleviate these problems and leaching efficiency has improved.

Caro's acid as an oxidant in acid leaching

(R.J. Ring)

Interox Chemicals has continued its sponsorship of research on Caro's acid (permonosulphuric acid) as an alternative oxidant to pyrolusite in the acid leaching of uranium ores.

Batch leaching tests were carried out on samples of primary and blended Koongarra ore containing 0.85 and 0.7% U_3O_8 respectively. The ore grades were more than twice those of Koongarra samples previously examined and were supposedly representative of ore to be produced under the revised mining plan for the deposit. Leaching conditions were pH 1.6, temperature 40°C and a redox potential of 450-420 mV (relative to the saturated calomel electrode).

Using Caro's acid in place of pyrolusite as oxidant had no effect on uranium extraction. Caro's acid reduced total sulphuric acid requirements in leaching by 13 per cent, and the ratio of pyrolusite to hydrogen peroxide consumption was 3.3. Both results are in good agreement with those obtained previously for Nabarlek, Ranger and Jabiluka ores.

Interox Chemicals has completed a series of plant trials using Caro's acid at the Nabarlek mill. The AAEC provided expert advice on the operation of the mill leaching circuit during the trials. The plant trials confirmed laboratory results and established that Caro's acid is a viable alternative oxidant to pyrolusite. From an operating viewpoint, Caro's acid was easier to handle, gave better control and ensured continual oxidant supply. It is clearly a better system than pyrolusite and has the advantage of almost eliminating manganese from waste effluents.

Koongarra Mines Pty Ltd has recently decided to use Caro's acid in the Koongarra mill which is now at the final design stage. Queensland Mines Ltd is also considering changing to Caro's acid at Nabarlek.

1.1.2 Waste treatment

Distribution of radionuclides in process and waste streams at the Nabarlek uranium mill

(R.J. Ring, D.M. Levins, K.P. Strong)

A survey was carried out at the Nabarlek uranium mill to determine the distribution of ^{226}Ra, ^{230}Th, ^{210}Pb and ^{210}Po in process and waste streams. The major conclusions of this work were as follows:

- Sulphuric acid leaching dissolved 0.01 per cent of the ^{226}Ra, 20 per cent of the ^{230}Th, 0.16 per cent of the ^{210}Pb, and 0.37 per cent of the ^{210}Po in the ore.
- About 0.005 per cent of the ^{226}Ra, 0.2 per cent of the ^{230}Th, and 0.002 per cent of the ^{210}Pb in the ore appeared in the yellowcake.
- Neutralisation of tailings/raffinate slurry with lime to pH 8.5 effectively precipitated almost all the ^{230}Th, ^{210}Pb and ^{210}Po.
- The concentration of ^{226}Ra increased significantly during neutralisation but decreased on ageing in the tailings pit.

Removal of radium-226 by barium chloride treatment

(R.J. Ring, D.M. Levins, K.P. Strong, R. Secomb)

Barium chloride is used extensively to remove radium from uranium mill effluents. At the Nabarlek mill, decant water from the tailings disposal pit is treated with barium chloride before discharge to an evaporation pond. The treatment circuit consists of a small mixing tank where barium chloride is added, then a clarifier where ferric chloride and flocculant carry down the fine barium-radium sulphate precipitate. Ferric hydroxide sludge from the underflow of the clarifier is recycled to aid settling.

The efficiency of radium removal is determined by the complex interaction of a number of parameters. These include rates of addition of barium chloride, ferric chloride and flocculant, hydraulic loading and the extent of sludge recycle. An experimental program was carried out to optimise the performance of the clarifier treatment circuit. Two plant tests were carried out at the Nabarlek mill as part of a collaborative research program. Radium-226 concentrations in the feed water were 75-150 Bq L^{-1} dissolved and

2-20 Bq L^{-1} suspended. Under optimum conditions, over 98 per cent of the total radium was removed in the clarifier circuit. Radium concentrations of 0.2-0.5 Bq L^{-1} dissolved and 0.5-1.0 Bq L^{-1} suspended were consistently obtained. The latter corresponded to a suspended solids concentration of about 2 mg L^{-1}. It was essential to maintain a stable sludge/liquid interface 1-1.5 m above the inlet position to achieve this clarity.

The plant tests identified a number of aspects of the process which required more detailed attention. A laboratory-scale clarifier circuit has been set up to carry out a systematic study of the process. Radium-223 is being used as a tracer in these experiments because it can be rapidly analysed by -counting. It was obtained by separation from its parent, actinium-227, in 3 M nitric acid solution using a cation exchange column. The eluate ^{223}Ra solution was passed through an anion exchange column to remove nitrate and increase the pH to 7-8.

1.1.3 Tailings management

Uranium mill tailings have a long-term environmental impact because of continual emanation of radon, possible dispersal of tailings dust and leaching of radionuclides due to water infiltration.

Radon emanation from tailings

(K.P. Strong, D.M. Levins)

Tailings piles are generally rehabilitated by covering them with earth to minimise erosion and suppress radon emanation. Measurements were made in a 107 mm diameter column to determine the effects of moisture content, porosity, and cover thickness on the radon flux. Results showed that moisture content was the most important factor affecting the radon diffusion coefficient.

Laboratory studies were augmented by field measurements on test pads on the Mary Kathleen tailings dam, the test pads consisting of varying thicknesses of waste rock and soil cover. Rain fell at the start of the field work and the radon flux from bare tailings increased with time as the tailings dried out. The effect of waste rock thickness, which was studied over the range 0-1.5 m, reduced radon flux significantly compared to bare tailings. Two methods of measuring radon flux in the field were evaluated: the accumulator method, in which a drum is sealed over the surface and the rate of build-up of radon in the air is measured; and the charcoal canister method, in which emanating radon is adsorbed onto charcoal and the γ-ray from ^{214}Bi is

counted. There was good agreement between both methods; however, the charcoal canister method required less operator attention and enabled a large number of measurements to be made simultaneously.

Collaborative research is being undertaken with Minatome Australia Pty Ltd on radon emanation from Ben Lomond tailings. This work entails measurement of the radon emanation coefficient, from both sand and slime fractions, as a function of moisture content. The radon flux from bare and covered tailings is also being measured. This information will be used to estimate the radon flux that will be produced under the proposed tailings rehabilitation scheme.

Concentration of radionuclide and sulphide fractions in tailings

(G. Dunlop*, D.M. Levins, R.J. Ring)

A research contract was awarded to AMDEL for a study of methods of physically separating the potentially hazardous fractions from tailings derived from three Australian ores. Conventional sulphide flotation removed 88-98 per cent of the pyrite in 1-5 per cent of the total mass. Removal of the pyrite eliminates the possibility that acid generation by bacterial oxidation will mobilise radionuclides and heavy metals.

Hydrocycloning was more effective than flotation for concentrating the radium into a low mass fraction. Up to 83 per cent of the radium was concentrated into only one sixth of the total mass. Most of the radium was contained in the finest particles (below 5 μm).

A combined flotation/hydrocycloning flowsheet to segregate tailings into three fractions for separate disposal was successfully tested. Possible options for disposal of the separated fractions are wet oxidation of the sulphide concentrate to produce sulphuric acid for use in the mill, backfilling of the sand fraction, and granulating of the slimes followed by disposal in trenches.

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* AMDEL

Radionuclide concentrations in Mary Kathleen tailings

(D.M. Levins, K.P. Strong)

In collaboration with Australian Groundwater Consultants Pty Ltd (AGC) present and projected levels of dissolved radionuclides in the Mary Kathleen tailings dam are being studied in connection with environmental impact studies related to rehabilitation of the tailings dam. Samples of tailings and associated water were taken at various depths in the tailings dam by AGC. The AAEC work involved measurement of dissolved radionuclides as a function of depth, radium mineralogy studies and accelerated leaching tests. Radium concentrations in the tailings water were quite low and only a minute fraction of the radium was mobilised in accelerated tests that simulated leaching by rainwater for over 100 years.

1.2 Waste Treatment and Disposal

1.2.1 Removal and conditioning of highly radioactive liquid wastes from ^{99}Mo manufacture

(J.M. Costello, E.J. Lee)

Since 1967, highly radioactive liquid wastes have been produced from the manufacture at Lucas Heights of ^{99}Mo (for radiopharmaceuticals) from irradiated uranium dioxide of up to 1.8 wt % ^{235}U enrichment.

Approximately 2 cubic metres of liquid waste containing 16 TBq (430 Ci) of fission products are in temporary storage at the Lucas Heights Research Laboratories. The aim of this research project is primarily to remove this waste from the existing storage facilities for volume reduction, waste solidification and alternative storage. A secondary purpose of the project is to provide a fission product concentrate for the manufacture of radioactive SYNROC specimens.

The waste consist of two types:

- Raffinate from the ion-exchange processing of dissolved irradiated uranium nitrate. This solution contains 1 M HNO_3, mixed fission products at 11-140 GBq L^{-1}, and uranium at 50-160 g L^{-1}.

- Washings from the loaded ion-exchange column, which consist of 1 M HNO_3 containing about 3 GBq L^{-1} of mixed fission products and 5 g L^{-1} of uranium.

TABLE 1
HIGHLY RADIOACTIVE LIQUID WASTES STORED AT LUCAS HEIGHTS

Location	Volume (L)	Total Uranium (a) Concentration ($g\ U\ L^{-1}$)	Total Uranium (b) Content (kg U)	^{235}U Content* (g)	$\beta\gamma$ Activity Content (TBq)
Bld.57 Tank No.1	372	50.3	18.7	<346	4.0
Bld.57 Tank No.3	1620	5.0	8.1	<150	5.5
Bld.54 Cell, Batch HW12	47	159.6	7.5	<139	6.6
Totals	2039		34.3	<635	16.1

*Assuming an initial ^{235}U content of 1.85 per cent.

TABLE 2
FISSION PRODUCT CONTENT OF BATCH HW12, BLD.54

Fission Product	Fission Yield (%)	Half-life	Activity (GBq)
^{89}Sr	4.75	50.4 d	35
$^{90}Sr/^{90}Y$	5.9	29 y	393
^{91}Y	5.8	58.6 d	107
^{95}Zr	6.2	65.5 d	203
^{95}Nb (from ^{95}Zr)		35 d	400
$^{103}Ru/^{103}Rh$	2.85	39.6 d	3.4
$^{106}Ru/^{106}Rh$	0.39	369 d	274
^{137}Cs	6.23	30.1 y	400
$^{144}Ce/^{144}Pr$	5.45	284 d	3640
^{141}Ce	6.0	32.5 d	1.1
^{147}Pm	2.2	2.62 y	1126
Total Activity			6.6 TBq

The locations and some details of these liquid wastes are listed in Table 1. The fission product content of the most recent waste is given in Table 2; most of the fission product activity results from ^{144}Ce-^{144}Pr (55 per cent), ^{147}Pm (17 per cent), ^{90}Sr-^{90}Y, ^{137}Cs, and ^{95}Nb (about 6 per cent of each radionuclide).

The SYNROC application requires extraction of dissolved uranium from the liquid wastes, followed by raffinate concentration. Counter-current solvent extraction in mixer-settlers has been selected for this purpose; five contactors have been designed as follows:

Extraction contactor	(1)	-	8 stages
Scrub contactor	(1)	-	6 stages
Backwash contactor	(1)	-	12 stages
Solvent wash contactors	(2)	-	1 stage each

A prototype extraction unit is currently undergoing hydraulic tests. Evaporation equipment has been designed and is awaiting fabrication. Following inactive proving trials, this equipment will be installed in a hot cell. Processing of radioactive liquor is planned to commence in 1983, subject to the availability of operational and engineering support staff.

2. ENVIRONMENTAL CHEMISTRY

2.1 Transport of Heavy Metals and Radionuclides Through the Environment (A.I.M. Ritchie)

The Environmental Chemistry Section is carrying out a number of projects to aid prediction and assessment of the impact of the uranium industry on the soil and aqueous environment. The work covers not only the full spectrum of radionuclides but also the non-radioactive materials associated with the industry. This includes such heavy metals as copper, nickel and zinc, which are frequently associated with the chemicals used during uranium treatment (such as manganese, sulphuric acid and organics) and the chemicals used in enrichment or waste processing (such as fluoride), and may exert an even greater influence in the uranium mine. The transport process may be broken down into the following subject areas: (i) adsorption/desorption; (ii) ion-

exchange; (iii) solution/precipitation; (iv) redox; and (v) aqueous speciation. Specific projects in some of these areas are described below.

Introduction of industry into a new area can cause an environmental source of pollution; for example, surface expression of saline water, conversion of aerobic waters to anaerobic waters, and the generation of acidic waters. This has been termed source chemistry. Of particular concern to the uranium industry is the generation of acid waters through the oxidation of pyrite in the waste rock heaps associated with uranium ore bodies with a high sulphur content. This problem is common to any heavy metal sulphide ore body located in tropical or temperate climates, and also occurs in high sulphur coal mines. A project on this subject is also discussed below.

The creation of the CSIRO Division of Energy Chemistry at LHRL resulted in a move by the Environmental Chemistry Section to the new top floor of Bld.21. Unfortunately, the necessary services for a chemistry section had not been included during the construction of the new floor. These services are being installed but experimental work in some areas has been interrupted for a period of up to one year.

2.1.1 Adsorption of heavy metals and radionuclides on soils and minerals

(R.T. Lowson, J.V. Evans)

The adsorption work previously reported (AAEC/ES-PR 1981) has been extended to a study of the adsorption of uranium on oxides and clays, but has been limited owing to disruption caused by the transfer of the section. However, the results indicate that the adsorption/titration curves follow the general relationship observed with the other metals. The previous results are in the final publication stages.

During 1980/81, a 48-column automatic rig was constructed to enable transport through soils to be measured by monitoring the position of a radiotracer of the metal under test. Transfer of the section has delayed commissioning of the rig by approximately one year. The rig is now in the final stages of being rebuilt and will be operational shortly. It will be used to study the transport of radionuclides of interest under a variety of conditions using a selection of typical Northern Territory soils which have already been collected.

2.1.2 Radium anomalies in the Northern Territory

(J.V. Evans, M.S. Giles)

Airborne scintillometry investigations in the Alligator Rivers region revealed some areas of anomalously high radioactivity which were later found to have little associated uranium. Presumably radioactive nuclides leached from uranium-bearing areas by spring waters have been concentrated in soils; the processes of build-up are being studied so that the mechanisms involved can be better understood. The objective is to investigate the nature of the concentration mechanisms and relate this to possible concentration areas in future mine drainage systems.

Initial field studies revealed that the excess radioactivity was associated with the acid swamp soils of a paper bark forest and a grass-sedge plain situated on the drainage of a constantly flowing spring.

Analysis of various fractions of the soils is continuing. Data to hand reveal:

- a disequilibrium ratio between U and ^{226}Ra of 140 (near the spring head) to 6 (five kilometres downstream),
- a disequilibrium ratio between U and ^{230}Th of 0.6 (at the spring head) to 3.3 (five kilometres downstream),
- a strong correlation between ^{226}Ra concentration and organic matter content of the soil, and
- the presence of ^{228}Th not supported by ^{232}Th, particularly near the spring head (disequilibria ratios of >540 to 1.3, zero and five kilometres downstream).

A technique for separating organic and mineral soil fractions without altering their physical characteristics is being refined.

2.1.3 The hydrolysis of metal ions

(R.N. Sylva, P.L. Brown*)

The distribution of a dissolved metal, between the free ionic state and the complexed and hydrolysed state, plays an important role in the behaviour and transport properties of the metal in the natural environment. The method of accurate pH titration coupled with computer calculations has been used to

* AINSE student, Univ. of Wollongong

determine the range of hydrolysed species present for a given metal, together with the stability constants for these species. The distribution and stability constants for the hydrolysed species of copper and uranium have been reported. Over the past year, the work has been extended to indium, scandium, thorium, molybdenum and beryllium. Indium and scandium were selected because the predicted range of hydrolysed polymeric species would allow the development of a generalised model for the hydrolysis of metal ions. It was expected that there would be a useful variation in the hydrolysis behaviour with ionic strength, and that another variable could be introduced in the form of chloride complexes. The development of the overall model has yet to be fully realised.

The hydrolysis of thorium was determined as part of the section's work on geochemical transport and, in particular, on the occurrence of thorium in uranium mine tailings. The properties of molybdenum are being determined as basic input to a technetium generator project, while the properties of beryllium are being measured because beryllium, which can be highly toxic, sometimes forms part of the uranium fuel cycle. The following values for the stability constants for the hydrolysed species indium (III), scandium (III) and thorium (IV) were obtained:

Species	Indium (III)	Scandium (III)	Thorium (IV)
1,1	$InOH^{2+}$ (-4.31)	$Sc(OH)^{2+}$(-4.840)	$ThOH^{3+}$(-2.98)
1,2	$In(OH)_2^{+}$(-9.35)	-	-
2,2	-	$Sc_2(OH)_2^{4+}$(-6.096)	-
3,5	-	$Sc_3(OH)_5^{4+}$(-17.567)	-
4,4	$In_4(OH)_4^{8+}$(-7.32)	-	-
4,12	-	-	$Th_4(OH)_{12}^{4+}$(-30.55)
6,15	-	-	$Th_6(OH)_{15}^{9+}$(-34.41)

2.1.4 Chemical oxidation of pyrite
(R.T. Lowson)

A review of the aqueous oxidation of pyrite by molecular oxygen, which was summarised in the previous progress report, has been accepted for publication in Chemical Reviews. A number of key areas were identified as requiring further work to permit a proper understanding of the system. One such area is the electrochemical characteristics of pyrite. Accordingly, over

the past year the potentiostatic characteristics of pyrite in sulphate media were determined as a function of pH, Fe^{2+} concentration, Fe^{3+} concentration and saturating gas (either oxygen or hydrogen).

In general, the characteristics were a complex function of all these variables. In the absence of Fe^{2+} and Fe^{3+}, the potential at zero forward current E_o was a non-linear function of pH but was independent of the saturating gas. Under oxygen-saturated conditions, simple cathodic and anodic curves were obtained which indicated that, under these conditions, the system was dominated by a single pair of cathodic and anodic reactions. Under hydrogen-saturated conditions, the anodic curve contained a passivated region which was most pronounced at pH 0 and could be detected up to pH 7. These results indicate that a common cathodic reaction occurs but that the anodic reaction is dependent on the saturating gas. Under oxygen-saturated conditions, the reverse sweeps had a small hysteresis component, whereas under hydrogen-saturated conditions, the reverse sweeps were extremely complex, with anodic and cathodic loops occurring within the passive region. This indicates some very unstable surface conditions.

The addition of Fe^{2+} or Fe^{3+} was found to suppress certain characteristics and enhance others. Consequently, it may be possible to identify and quantify those parts of the curves which are dominated by iron redox reactions. The analysis is complicated by the presence of the as-yet unidentified reactions for the oxidation of S_2^{2-} to SO_4^{2-}. An attempt will be made to identify the contribution of this reaction to the overall potentiostatic curve by repeating the experiments on a range of thio-sulphate solutions.

A number of hypotheses have been developed to identify the rate-controlling parameters for the oxidation of pyrite in waste heaps. To test the validity of these ideas, a leach column has been constructed measuring 2 m high by 0.3 m diameter and containing sulphidic overburden. The column is extensively instrumented with thermocouples, potential probes, oxygen probes, piezometers, humidity probes and sampling ports, and is being interfaced to an Apple computer for automatic data collection. Commissioning of the rig was delayed by about one year owing to the transfer of the rig from the Chemical Engineering Bay in Bld.2 to the new Environmental Engineering Bay in Bld.21. Leaching commenced in June 1982. No significant results will be obtained until the column becomes close to saturation. The present dryness of the column is reflected by the tendency of the piezometers to break down under the

high capillary suction exerted by the column. It is hoped that this problem will be overcome automatically as the column stabilises.

2.1.5 The chemistry of uranium mill tailings
(G. Khoe)

A start has been made to produce better definition of the chemistry of uranium mill tailings by identifying those parameters which characterise the waste. These include such physical properties as particle size and distribution, and chemical properties of the solid, and the aqueous phases and mineralogical changes that occur with ageing. Experiments will be designed to investigate the geochemical processes occurring within the tailings. These include solution/precipitation reactions, redox reactions and surface processes. Both the equilibrium and kinetic components will be evaluated.

2.2 Ion Chromatography
(S.R. Isaacs)

The ion chromatograph has been used routinely throughout the year to provide an analytical service for both the radium anomalies and technetium generator projects. Analytical techniques using this instrument have been developed for a range of environmental anion compositions which have allowed the determination of the anion composition of natural waters sampled in the Northern Territory and the exchange capacity of soils. The original columns have been replaced by faster columns thus reducing the analysis time for multiple ion samples by a factor of two. An integrator has been installed which will permit enhanced peak analysis for those systems with overlapping peaks, as well as a more exact determination of the peak characteristics.

The instrument has been used for the routine determination of nitrate down to 1 mg L^{-1} in saline (0.1 mol L^{-1} NaCl).

A halide suppressor column, for use during the determination of organophosphate in the presence of chloride, has been supplied by the manufacturer of the ion chromatograph. Some initial experiments have indicated that the suppressor column interferes with the phosphate standards and that the technique will have to be modified.

Some work has been started on the quantitative determination of thio ions in the presence of sulphate. The current method has unacceptably long

residence times for the thio-sulphate moiety and the resulting thio-sulphate peak is broad and tails badly. Ways are being sought to improve the residence time and peak shape without sacrificing peak separation.

2.3 Application of Track Etch Techniques

(W.R. Ellis)

In the determination and location of radium-226 by alpha track etching, the amount of radon and daughter products present is usually unknown. If, however, the radon were removed as soon as it formed, all the tracks registered would be due to radium only. This technique was attempted by subjecting the sample to a vacuum for a few hours to remove the existing radon and allow its daughter products to decay, then placing it in contact with the track detector, and again subjecting it to a vacuum throughout the exposure time. Inconclusive results were obtained for radium in an inorganic substrate, but further work is in hand using biological materials.

A method was investigated for the estimation of radium in soils by track etching to determine the emanated radon in a closed system. The number of tracks formed and the radium content of the soil were not well correlated (even for similar soils), presumably because of variability in the rate of radon emanation.

In cooperation with Dr M. Ahsanullah, fission track and delayed neutron methods are being investigated for the determination of uranium in seawater and marine mussels which have been exposed to uranium solutions. Visualisation of the distribution of uranium in various mussel tissues will be studied by means of the fission track method.

3. PHYSICS OF THE ENVIRONMENT

3.1 Rum Jungle Environmental Studies

(B.A. Beard, M.G. Blackford, J. Casteleyn, J.A. Daniel, J.R. Harries A.I.M. Ritchie)

3.1.1 Temperature and interstitial gas composition in overburden dumps

Previous measurements of the temperature distribution within White's dump which showed that oxidation occurs at depths as great as 14 m below the surface at some locations, raised questions as to how the oxygen is

transported to such depths and whether diffusion from the surface can supply sufficient oxygen. In January 1981, nine holes were drilled in the overburden dumps to enable the interstitial gas to be sampled deep within the dumps, and also temperature and water content to be measured in Intermediate dump. Seven of the holes were fitted with liners which contained up to 12 stainless steel gas ports at various depths down to the base of the dump. Each port was linked by a nylon tube to the surface through which gas samples could be collected. An inner liner was also installed in each hole to enable the temperature distribution to be measured.

Analysis of the gas samples showed, surprisingly, that marked decreases in the oxygen concentration occurred with increasing depth in holes where uniform temperature distribution indicated that no oxidation was occurring at depth. Analysis also showed significant oxygen concentrations in all holes where oxidation was known to be occurring at depth. The hottest hole in White's dump, location A, which has a temperature exceeding 50°C at 14 m, had oxygen concentrations exceeding 15 vol.% at depths between 10 and 15 m. In this probe hole, the oxygen concentration decreased from the surface to a minimum at about 5 m before increasing again to show a maximum at about 13 m.

Four of the six holes drilled in the Intermediate dump were fitted with gas ports. At two of the locations, the temperature at depth exceeded 55°C, and the oxygen concentration initially decreased with increasing depth, before increasing to a maximum at 12 to 15 m depth. The increase of oxygen concentration at depth in probe holes which also show an increase in temperature with depth indicated that an oxygen transport mechanism, in addition to vertical diffusion from the surface, is involved and suggests that the oxygen is supplied horizontally, either from the side of the dumps or from a high porosity area of the surface.

The carbon dioxide concentrations were generally high in both dumps, exceeding 2 per cent in most holes and reaching 20 per cent at one location in White's dump. The CO_2 concentration tends to be negatively correlated with the oxygen concentration and suggests that when the oxygen concentration at depth is high, gas transfer rates to these locations from the atmosphere outside the dump are quite high. The carbon dioxide is produced, presumably, by the action of sulphuric acid on dolomite or other carbonates in the dumps.

Temperature measurements in the new probe holes also confirmed the assertion used in earlier analysis that the temperature distributions are an

integral property of the dump and represent a large volume around the probe hole. The holes drilled in this program were within 15 m of previously drilled holes, and the temperature distributions in the pairs of holes agreed, usually within 1°C, even for those holes in which high temperatures were measured.

3.1.2 Calculation of heat source distribution

The measured temperature distributions from White's dump have now been used to calculate the heat source distribution through the wet season when water percolating through the dump provides an extra heat-transport mechanism. The net effect of downward percolating water is to displace the temperature distribution vertically downwards. In a real dump, the water need not infiltrate uniformly downward and there could be considerable variation in the temperature displacements at different locations. Within the errors estimated for the heat source distributions from this and other causes, there appears to be no variation in the heat source distribution in White's dump from dry season to wet season. This implies that the oxidation process within this dump has little, if any, dependence on the wet/dry season cycle.

The heat source probe which is being developed to determine the thermal diffusivity of the dump material has now been used at Rum Jungle. There are still uncertainties in the effect of convection in the probe hole and two holes have been drilled at Lucas Heights to allow the effect of convection to be investigated.

3.1.3 Runoff and pollution data

The data collected from the weirs on White's and Intermediate dumps during the 1980-81 wet season are being analysed for indication of differences in the behaviour of the two dumps. The analysis program has been modified to provide cumulative runoffs and rainfalls so that the runoff coefficients and pollution loads in sub-events can be measured.

3.1.4 Acid mine drainage

A review of acid mine drainage has been written which compares the experience at various mine sites and shows the large range of rehabilitation techniques used.

3.2 Modelling the Pyritic Oxidation Process in Waste Rock Dumps

(G.B. Davis, A.I.M. Ritchie)

An earlier model of pyritic oxidation in a waste rock dump assumed the dump to be a homogeneous slab, and the transport of oxygen to the oxidation sites within the dump to be the rate-limiting step in the oxidation process. Further, oxygen transport to these sites was assumed to be by gaseous diffusion through the pore space of the dump from the air at the surface of the dump. It followed that oxidation proceeded at a reaction front which moved from the surface of the dump to the base with a $(\text{time})^{\frac{1}{2}}$ dependence. This simple model predicted an annual SO_4 production for White's dump that was of the order of that deduced from field measurements. The model also predicted that heat production would be confined to the region of the reaction front and that the temperature in the dump would be a maximum at that point. Neither of these predictions accorded with field measurements.

A modified model has been formulated which still assumes that oxygen transport by diffusion is the rate-limiting step, but which assumes oxygen transport to be a two-stage process - diffusion through the pore space, followed by diffusion into oxidation sites within the particles comprising the dump. A pseudo-steady-state assumption was made with respect to diffusion within the particles to assist in solving the moving boundary value problem within the particles and the other equations were solved numerically. Variables of interest are the spatial heat source distribution and the oxygen concentration of the pore space.

At this juncture, these variables are evaluated, assuming just one particle size in the dump. Some typical results are given in Figure 1 which shows that, unlike the earlier and simpler model, the present model predicts a distributed heat source distribution whose width depends on the particle size. The model also predicts an oxygen concentration distribution rather different from the effectively linear distribution predicted by the simpler model (Figure 2). Good qualitative agreement is obtained between measurements of the heat source distribution and distributions predicted by the model (Figure 3). Work is in hand to include the effects of a particle size distribution typical of that of the overburden dumps under investigation.

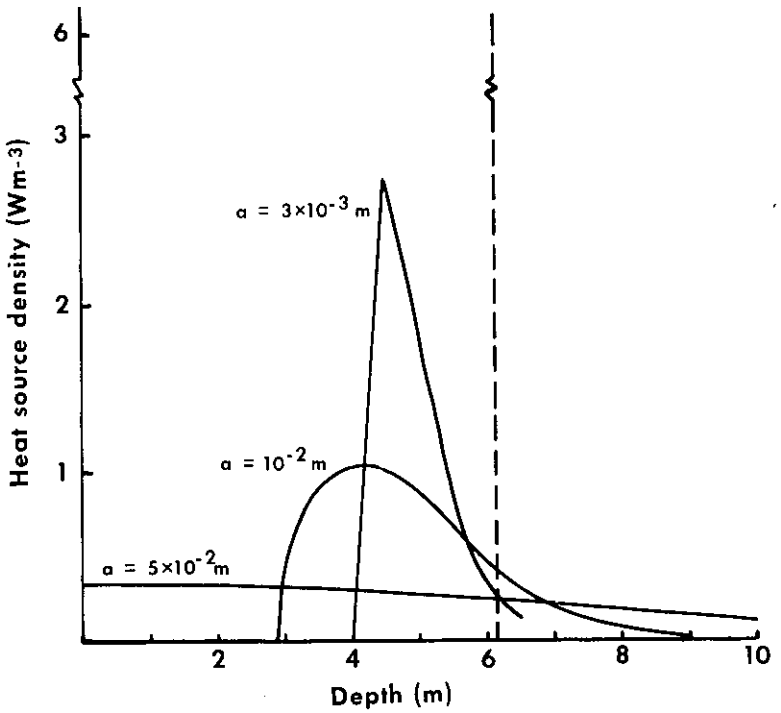

FIGURE 1 THE CALCULATED SPATIAL HEAT SOURCE AS A FUNCTION OF DEPTH FOR VARIOUS PARTICLE RADII (The dashed line is the simple model)

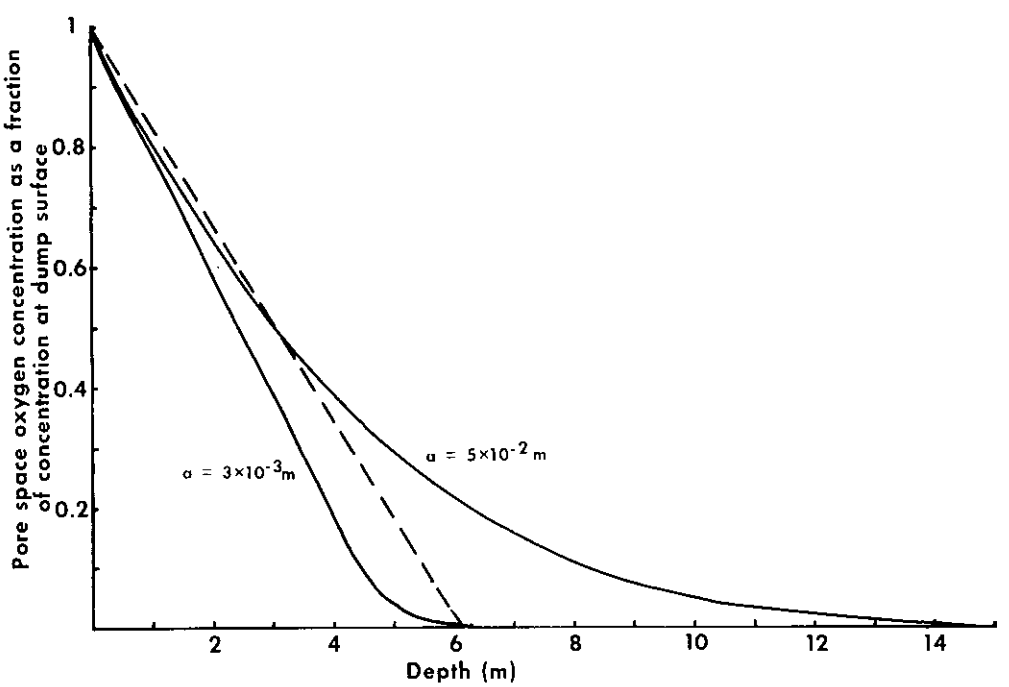

FIGURE 2 THE CALCULATED OXYGEN CONCENTRATION AS A FUNCTION OF DEPTH FOR VARIOUS PARTICLE RADII (The dashed line is the simple model)

3.3 Tritium Levels in Seawater

(J.R. Harries, G.E. Calf*)

The tritium distribution in the South Pacific has been analysed in terms of the characteristics and circulation on the isopycnal surfaces (constant density). The analysis was complicated by the elapsed time between the various sets of tritium data, but most of the tritium distribution could be explained by the horizontal mixing and advection on isopycnal surfaces. The tritium concentrations show that the upper limit for mixing on the $\sigma_t = 26$ isopycnal surface for the South Pacific must be less than 20 years. Only near the East Australian Current was there evidence of significant transport across the isopycnal surfaces.

3.4 Airborne Pollutant Transport

(M.C.E. Petersen, G.H. Clark, E.O.K. Bendun)

3.4.1 Gamma-ray dose estimates from airborne radionuclides

Many situations necessitate estimation of the dose to an individual from an airborne distribution of gamma emitters released from a source. Apart from the problem of providing an adequate description of the atmospheric transport of activity, a difficult mathematical problem arises in estimating the dose to an individual from the instantaneous distribution of airborne activity. This distribution of activity is usually described by a Gaussian plume. The problem involves integration over the distribution of gamma emitters in the plume to obtain the number of gamma-rays incident upon an individual [Gammertsfelder 1960+].

This difficulty is usually circumvented by assuming that the concentration of gamma emitters at the location of the individual is of infinite extent, and to evaluate the dose to the individual on this basis. This is called the infinite cloud approximation. Alternatively, the difficult integral arising from the plume distribution of activity can be dealt with by describing the plume of gamma-emitters as a sum of three-dimensional Gaussian puffs of activity. This also permits greater realism in the description of the atmospheric transport of the activity. A spherical approximation has been developed in which the Gaussian puff distribution of activity is replaced by a

* Isotope Division

+ Gammertsfelder, C.G. [1960] - Hazards Calculation Notebook. Aircraft Nuclear Propulsion Department, General Electric Co., USA (unpublished).

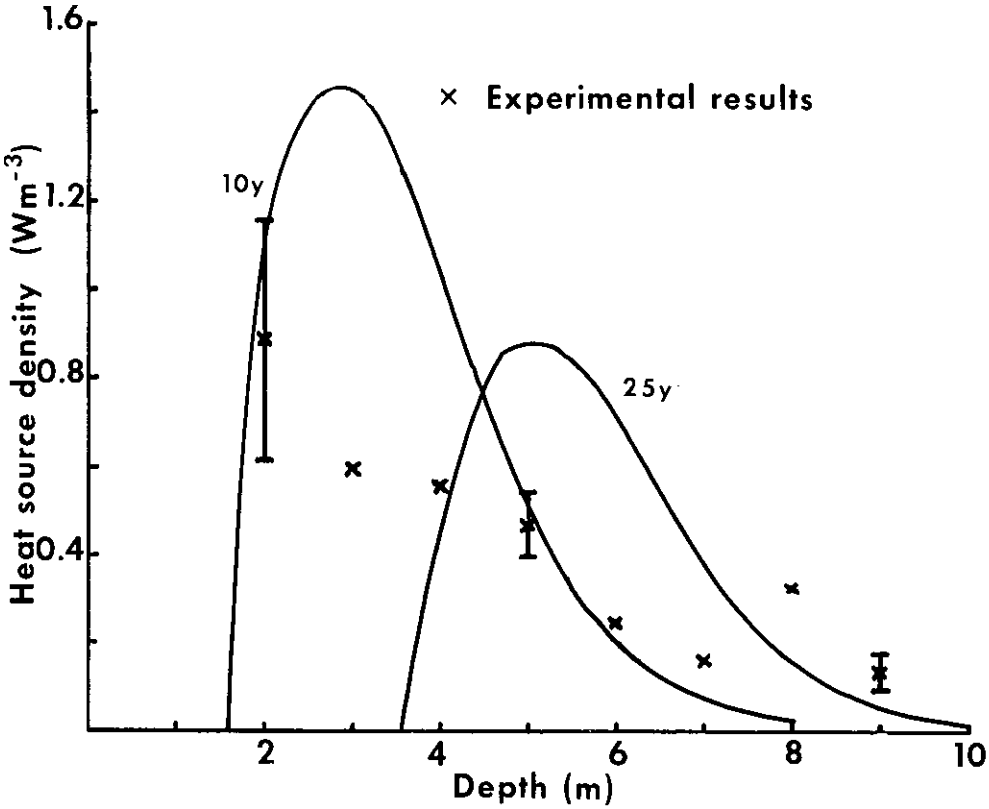

FIGURE 3 COMPARISON OF MEASURED AND CALCULATED SPATIAL HEAT SOURCE DISTRIBUTIONS

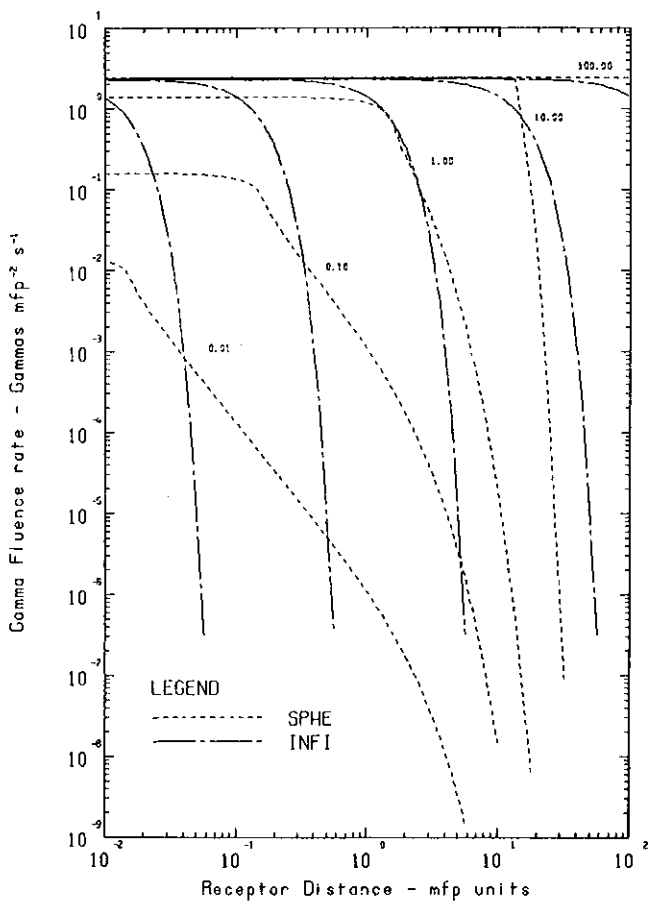

FIGURE 4 GAMMA FLUENCE RATE v. RECEPTOR DISTANCE

sphere of uniform concentration of activity whose radius, R_s, is given by

$$R_s = (9\pi/2)^{1/6}\sigma \quad ,$$

where σ is the variance of the Gaussian distribution. This approximation leads to very useful analytical expressions.

In Figure 4, the infinite cloud approximation is compared with the spherical approximation. The ordinate shows the gamma fluence rate, in gamma units of mfp^{-2} s^{-1} since dose-rate and dose are proportional to this. The unit mfp (mean free path) is used instead of m for mathematical convenience, and at the gamma energy (1 MeV) considered in Figure 4, 1 mfp ≡ 122 m. The receptor distance, shown on the abscissa, also in mfp units, is the distance of an individual from the centre of the Gaussian puff, and hence from the centre of the sphere. The figures on the graph, near the cross-over points of the two different types of curves, are the values of for the Gaussian puffs. Although not shown in the figure, the results for the spherical approximation when $\sigma \leq \sigma$ are in close agreement with the results obtained for the exact Gaussian distribution of activity in a puff.

The figure illustrates the deficiency of the infinite cloud approximation relative to the spherical approximation, when is less than 10. When an individual is close to the centre of a Gaussian puff, the infinite cloud approximation can overestimate the gamma fluence by as much as two orders of magnitude, whereas beyond the cross-over point for each pair of curves, the infinite cloud approximation can underestimate the fluence by many orders of magnitude. Again, although not shown, the spherical approximation is in close agreement with the result for the exact Gaussian distribution of activity. These results provide a criterion for the use of the infinite cloud approximation in gamma-ray dosimetry and an analytical alternative when this approximation is inadequate.

3.4.2 Improvements in meteorological instrumentation

To minimise the manual digitisation of analogue meteorological records, a microcomputer-based data acquisition system will be used to store 20-30 minute averaged wind, temperature and solar radiation data on floppy disks before transferring them to the main AAEC computer. At present, an interface between the sensors and microcomputer is being developed.

3.5 Environmental Radon Measurements

(S. Whittlestone, B. O'Brien, K. Mears)

3.5.1 Radon calibration facility

The 8 m^3 chamber for the radon calibration facility has been completed. Plumbing for the water jacket temperature control and for the chamber air supply is almost ready. Installation of the exhaust air duct should enable radon atmospheres to be created in the chamber, and determination of the uniformity of the radon concentration, the residence time of the radon daughters before plate out, and other parameters relevant to the operation of the facility.

3.5.2 Radium analysis rig

A rig for the routine analysis of radium using direct transfer of radon from sample to scintillation cell has been completed to the stage where manual operation is possible. Preliminary trials have demonstrated a degree of reproducibility which is steadily improving. It is hoped to reduce errors from the present level of about 30 per cent to better than 5 per cent.

3.5.3 Contract field measurements

The radon emanation rate has been measured at about a hundred points over the tailings from two abandoned uranium mills - Moline and South Alligator in the Northern Territory - to provide data relevant to the rehabilitation of the sites. Substantially elevated radon emanation was found in areas where the gamma dose was high, and detailed analysis of the results is in progress.

3.5.4 Radon tracer measurements

Radon can be used to indicate large-scale movement of air masses because it is emitted from continents but not from oceans, and it decays to background levels in about ten days. The concentration of radon is, therefore, useful for the interpretation of measurements of atmospheric pollution levels at such installations as the Cape Grim Baseline Air Pollution Monitoring Station, on the west coast of Tasmania, which was established to monitor contamination of the atmosphere as a whole to enable this to be compared with that which can be attributed to a source, such as continental Australia.

The radon detector at Cape Grim comprises a 2 m^3 chamber in which radon daughters are allowed to build up and become attached to particles generated by a hot wire dipped in oil. The air is drawn at 40 L min^{-1} through a filter

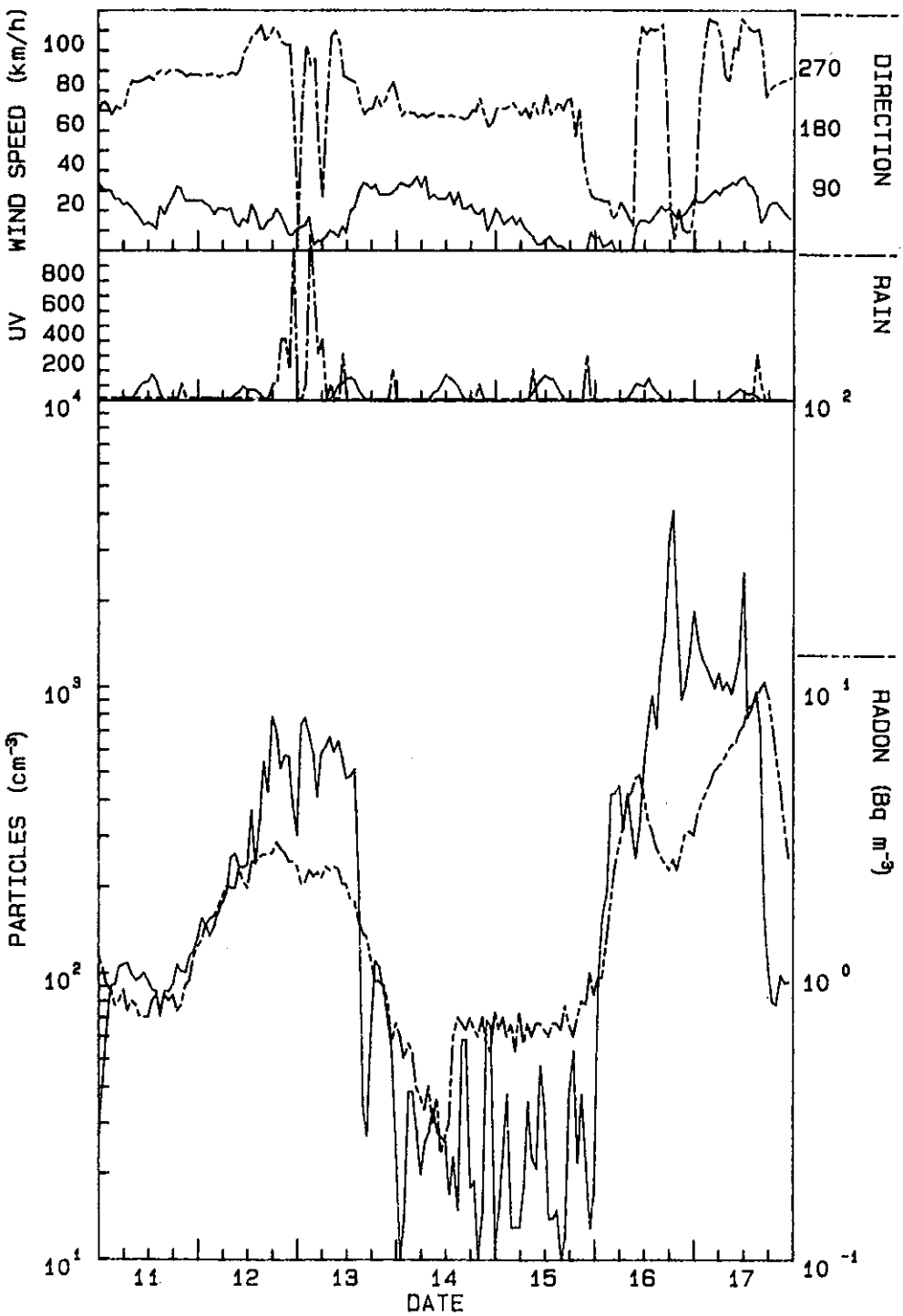

FIGURE 5 RADON, PARTICLE COUNT AND METEOROLOGICAL PARAMETERS AT CAPE GRIM, JUNE 1981

upon which the radon daughters are deposited. The α-particle activity of this filter, then, is proportional to the concentration of radon in the air. A sensitivity of 30 counts min^{-1} Bq^{-1} m^{-3} has been achieved. By way of comparison, the concentration of radon in Sydney air is ∿ 1000 Bq m^{-3}.

Some data acquired during a week when winds came from the Australian continent (NW to NE) and from the Antarctic (S to SW) are shown in Figure 5; meteorological data include wind speed and direction, ultra-violet radiation and rainfall. It can be seen that during 'baseline' conditions, with the wind from the south to south-west, radon levels are less than 1 Bq m^{-3}, whereas during a northerly wind the levels reach 10 Bq m^{-3}. The trend of the particle concentration is much the same as that of the radon, although there are dramatic increases not seen in the radon, such as the one just before midnight on the 16th, when the wind came from the direction of Melbourne.

3.6 Radiation Dosimetry Physics

(D.K. Gibson, I.D. Reid*, M. Hyde)

This project is concerned with measurement of the ejection of electrons from atoms by the impact of protons. We are attempting to measure the energy spectra of electrons ejected at various angles in order to obtain the double differential cross sections. In particular, we are interested in the cross section near zero degrees (where the electrons are ejected in nearly the same direction as the incident proton) because it is made up of two components, namely:

(a) the electrons ejected by electromagnetic energy transfer, with cross sections varying slowly with angle; and

(b) the electrons ejected by charge exchange into the continuum (CEC), with cross section sharply peaked at zero degrees and electron energy such that the electrons have the same velocity as the incident protons.

Comparison of ionisation by electrons and protons suggests that the CEC component could contribute up to 30 per cent of the total ionisation cross section by protons at certain energies. We have attempted to make the first direct measurement of the CEC cross section by measuring the electron ejection between 0 and 10° and over the energy region appropriate to the proton velocity. Proton energies of 30, 40 and 50 keV were used. Graphical and

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* AINSE

numerical means were used to extrapolate the electromagnetic component from 10 to 0° and the results of Rudd and Madison [1976][1] used to normalise them to absolute values. Subtracting these estimates from the total yielded the double differential CEC cross sections, $\sigma_c(\varepsilon,\theta)$. To obtain the total CEC cross section, a value for

$$\sigma_c(\varepsilon,\theta) \quad \sin\theta \; d\theta \; d\varepsilon$$

was estimated by numerical integration.

Table 3 shows that our estimates of the CEC cross section are a few per cent of the known total ionisation cross section rather than the above-mentioned 30 per cent.

TABLE 3

Proton Energy (keV)	σ_{CEC} (10^{-18} cm^2)	σ_{ion} (10^{-18} cm^2)	Ratio $\sigma_{CEC}/\sigma_{ion}$ (%)
30	1.3	48.0	2.7
40	1.3	63.3	2.1
50	2.0	74.2	2.7

This work cannot be described as final because of a perturbing effect which came to light during the measurements; the detectors appeared to have counting efficiencies which depend on the average electron flux received over the preceding few minutes. A later study of the pulse height distribution of the channeltron output pulses showed that the spectra distribution had degraded so far that it was difficult to separate the signal and noise reliably. These faults led us to suspect that the channeltrons were impaired by an insulating layer deposited over their surfaces. The performance of the channeltrons was so poor that a radical idea was tried, and the detectors were cleaned with a solution of NaOH which generally removes hydrocarbon deposits. This treatment produced a very marked improvement in the performance of the detectors. Performances of five cleaned detectors have been compared and shown to have efficiencies within 10 to 20 per cent of one another and no

1. Rudd, M.E. and Madison, D.H. [1976] - Phys. Rev. A, 14:128.

apparent intensity dependence. We are awaiting delivery of two new channeltrons to compare with the cleaned ones.

The statistical accuracy of the results has also been improved by increasing the pumping speed in the target chamber. This was achieved by replacing the turbomolecular pump by a fast diffusion pump with a liquid nitrogen cold trap. An improvement of signal-to-background count rate of about five times has been achieved.

3.7 Cost Analysis for a SYNROC Production Plant
(M.C.E. Petersen)

A computer code has been written to estimate the unit levellised capital and operating costs of a SYNROC (or alternative) high level radioactive waste immobilisation plant. In the US Dept. of Energy report 'Technology for Commercial Radioactive Waste Management'*, a similar code called UNICOST was used; however, this code has not been documented and is not generally available.

The essential feature of the AAEC and DOE codes is that the price of the output product or goods is estimated from the known prices of such input goods as labour, materials and services, but also includes the cost of borrowed and equity capital. Taxes are paid to the appropriate authorities on net profits after taking into account both the carry-forward losses, which usually occur during the construction and start-up phases of an operation, and the carry-back losses, which are usually associated with decommissioning and shut-down phases of an operation.

This type of project cost-benefit analysis differs from the conventional one where the market price of the output goods is known and the present discounted value (PDV) of the net cash flow and the internal rate of return are estimated. The present computer code deals with the reverse problem; it chooses that price for output goods at which all suppliers are paid a competitive market price for their services and goods and where the return on equity capital is equivalent to that from alternative types of investment. The computer code achieves this by solving the PDV versus price (of output goods) for price at zero PDV. This process ensures that an economically justified price of output is determined.

* DOE/ET-0028, May 1979.

4. ENVIRONMENTAL BIOLOGY

4.1 Marine Pollution Studies

(M. Ahsanullah)

Short-term experiments (acute toxicity tests) were carried out using arsenic, chromium, selenium and uranium on the amphipod Hyale longicornis, newly hatched crab larvae Paragrapsus, and a marine copepod species (yet to be identified). In general, the duration of the experiments was 96 hours. This was extended to 168 hours for the amphipods and reduced to 24 hours for copepods. At present, the data are being subjected to probit analysis.

During the investigation, H. longicornis exhibited aggressive behaviour, and attacked and ate weak individuals. This cannibalistic behaviour was not seen before in the short-term experiments with Allorchestes compressa, a similar species belonging to the same family Hyaliidae. Further, because of the obscure reproductive behaviour of H. longicornis it was decided to use A. compressa, whose reproductive biology is well known. This species will be used for long-term (chronic toxicity) experiments on growth over multiple generations at various temperatures and salinities.

Preliminary experiments indicated that uranium concentrations greater than 40 mg L^{-1} precipitated in seawater and, when a copepod species was exposed to 30 mg L^{-1} U for 24 hours, the survival rate was 100 per cent, indicating that this element is not toxic even at high concentration. As a result, the decision was made to conduct long-term experiments on accumulation and depuration of uranium at low concentrations (including 0.1 mg L^{-1} U because levels less than this present minimal risk of deleterious effects) by the common mussel Mytilus edulis. Arrangements were made to obtain mussels of known age from the Marine Science Laboratories of the Ministry for Conservation, Victoria. In experiment 1, mussels were starved and in experiment 2 they will be fed regularly with the alga Dunaliella tertiolecta. The algal culture has already been established.

Experiment 1 has been completed. The mussels of two age groups - 8 and 20 months - were exposed to 0.1, 1 and 2 mg L^{-1} U plus control (seawater only) under a continuously-flowing seawater system for eight weeks. The data have yet to be analysed.

4.2 Role of Aquatic Macrophytes in Radium Cycling on the Magela Floodplain

(A.R. Williams, J.R. Twining)

4.2.1 Objectives

A biogeochemical survey of the Magela floodplain has identified foliar uptake of radium by aquatic macrophytes as a potential mechanism of radium accumulation in soil in the region where drainage from the uranium developments enters the floodplain.

To predict the fate and long-term cycling of effluent radium, both during and after the completion of the mining operations, we have set up laboratory facilities at Lucas Heights to investigate radium uptake in two major macrophytes from the beginning of the Magela floodplain - the water lily Nymphaea gigantea and the grass Pseudoraphis spinescens; also we have initiated some computer-modelling studies of plant productivity and radium cycling in the floodplain soil-plant-water system.

4.2.2 Progress

Biological uptake of radium

In animals, the distribution of radium is correlated with that of calcium, but in plants there appears to be no such correlation, although plants discriminate against radium, relative to calcium, in the uptake process.

The concentration of such major ions as Ca in the ambient water can dramatically affect the extent of radium uptake and, although this effect has not been studied in macrophytes, Ca concentrations in the order of 10^{-4} to 10^{-3} M have been found almost entirely to suppress radium uptake in fish and mussels. Since uranium mill raffinate contains high concentrations of Ca and Mg, this effect also needs to be investigated for macrophytes. The mechanism of Ca interference in Ra uptake may be saturation of the membrane uptake sites, although Ca also has a nutrient role in membrane structure and it is active in increasing the selectivity of the membrane in plants.

Algae accumulate radium rapidly from water with a concentration factor of $\sim$ 440 (fresh weight) in one to three days; macrophytes accumulate radium somewhat more slowly, with a concentration factor of $\sim$ 55 in 12 to 15 days.

This difference may be important where the macrophytes support a significant growth of epiphytic algae. The contribution of radium from water and sediment to macrophyte foliage was roughly equal at background concentrations; however, in polluted waters, the contribution from water to foliage increased, as was expected. There is some evidence to suggest that the presence of macrophytes may increase metal uptake by sediment from polluted waters.

Radium uptake in Nymphaea gigantea

Two static experiments on whole plants of Nymphaea gigantea and a morphologically similar commercial species, N. capensis, have been carried out. Both species gave similar results and, in 400 L of water over a two-week exposure period, 25 per cent of the radium was taken up within the first day, with an exponential half-time of 0.5 hours, indicating adsorption onto surfaces; a further 25 per cent was taken up, with a half-time of 2.5 days, equivalent to the uptake rate of macrophytes, although only about 1/5 of this was found in the plant; most of the remaining 50 per cent was still in solution after 15 days, disappearing with a half-time of $\sim$ 40 days. Even though a large volume of water was used in these experiments, much of the radium was still lost very rapidly, thus indicating the need to use a continuous flow system. This is currently under construction, and has a planned maximum turnover rate of one complete change every three days.

To measure the radium concentrations in these experimental samples, we have developed a solvent extraction method for counting ^{222}Ra + daughters by liquid scintillation. A specially designed (Twining) flask achieves 70 per cent radon extraction into toluene, with exclusion of radium, and for each radon atom, three alpha and two beta particles are detected with 90-100 per cent efficiency. The lower limit of determination of the method lies between 1 and 10 mBq.

Computer studies

A morphometric growth model of Nymphaea gigantea was developed using field and laboratory measurements of six plant parts (laminae, petiole, fruit, peduncle, stem and roots). The model may be used to estimate the biomass and productivity of foliage and whole plants, from a count of the number of floating leaves and emergent flowers, and the depth of the water; this greatly facilitates field survey of the annual growth cycle and population dynamics of the species. The estimated annual productivity is quite low (100 to 500 g m^{-2}), increasing with depth of the water.

A simple compartment model of the floodplain soil-plant-water system was also set up, using data from the literature, to simulate the passage of water down the floodplain and so investigate the role of the aquatic macrophytes. Although this model is still under development, it indicates a potential for accumulation in soil at the beginning of the floodplain, as was suggested by the biogeochemical survey work.

4.3 Primary Productivity of Estuarine Benthic Microalgae and Element Concentrations in Sediment Interstitial Water and Sediment

(M.S. Giles)

Three field excursions to the Alligator Rivers region have been made during the year. Photosynthetic productivity of benthic microalgae under saturated light conditions was found to vary seasonally over about one order of magnitude (0.015-0.17 mg C g^{-1} h^{-1}), with the highest readings occurring at the end of the dry season. Such a large variability probably rules out the use of this measurement as a monitoring criterion for heavy metal pollution studies.

Interstitial waters from estuary sediments were generally similar to free estuary water, whereas interstitial waters from freshwater sediments were higher than the water column above them. In estuary sediment interstitial water, ^{226}Ra was relatively high (270 mBq L^{-1}) and one interstitial water sample from the freshwater section gave anomalously high concentrations of metals of up to two orders of magnitude higher than other freshwater sampling sites.

Acid digests of estuary sediment particles revealed relatively high levels of Zn, Cu, Pb and Mn, averages being 15.1, 9.9, 7.3 and 167 µg g^{-1} (dry weight) respectively. Analysis of samples and collation of data is continuing.

4.4 Uptake and Loss of Radium-226 by Freshwater Mussels

(R.A. Jeffree)

Mussel populations inhabiting billabongs close to the Ranger uranium mine in the Northern Territory are a source of traditional food for Aboriginals in that region. As a result of mining operations, mussels may become exposed to increased levels of ^{226}Ra and to levels of other alkaline earth metals, calcium and magnesium that vary during the mining and post-mining phases.

Thus, the rates of uptake and loss of radium in mussel flesh and the effects of calcium and magnesium on these rates must be known.

Previous experimental studies have demonstrated that an elevation of calcium water concentration inhibits the uptake of radium. Further laboratory experiments have determined the effects of elevated magnesium water concentration on radium uptake and loss. A magnesium level increased by a factor of 10 above background does not inhibit the uptake of radium, and the radium accumulated is not subsequently lost when mussels are exposed to radium-free conditions. However, an elevation of the magnesium level by a factor of 50 does inhibit the uptake of radium, apparently by disruption to the normal physiology of the mussel. This hypothesis is being investigated.

Field-collected and laboratory-exposed mussels are characterised by great variation in their radium tissue concentrations. This limits the value of such data for both the accurate determination of significant increases in radium tissue concentration caused by mining activities, and the prediction of tissue concentrations resulting from elevated radium water concentrations. Current studies have aimed at explaining this variation.

It has been found that radium concentration in the tissue of field-collected mussels increases significantly with size and age. The concentrations of the alkaline earths, calcium and barium, but not magnesium, also increase significantly with size and age; calcium concentration is the best predictor of radium concentration, explaining nearly 90 per cent of the variation amongst individuals. It is likely that the variation in radium concentration in laboratory-exposed animals is related to short-term variation in calcium uptake rates between individuals. Sites of accumulation of radium within the tissues are being investigated to elucidate the mechanism of accumulation.

The CR-39 nuclear track recorder has been used successfully to determine the sites of accumulation of radium in field-collected mussels; exposures of the CR-39 to 5 μm sections of mussel tissue have shown that the highest densities and greatest number of α tracks are associated with granular deposits, with lower densities in gills and renal tissue. Further studies with the CR-39 have determined the sites of accumulation of radium in laboratory-exposed mussels. Preliminary observations indicate that most of the accumulated radium is also located in granular deposits. Particularly high densities of α tracks are associated with granules in the edge of the

mantle, the region that secretes the shell.

Results from the experimental studies and data gathered from field-collected mussels all support the hypothesis that radium is treated metabolically as an analogue of calcium; from these results, several conclusions can be drawn with respect to the effects of mining effluent on radium concentrations in mussel flesh.

During the operational phase of mining, releases of effluent are predicted to elevate the radium and calcium levels in the receiving waters; radium uptake will be inhibited by the elevated calcium level. However, after the abandonment of the mine site, calcium will, in the long-term, leach from the tailings material at a greater rate than radium. Calcium levels in the receiving waters will revert to the low background levels; radium levels will continue to be elevated. Under these conditions, radium will be readily taken up in mussel flesh and retained in the granular deposits.

4.5 Toxicity of Copper to Freshwater Fauna

(N.J. Williams, K.M. Kool*)

Copper is one of the most toxic contaminants expected to enter the Magela Creek system (Alligator Rivers region, NT), in small quantities, via seepage from the Ranger uranium mining and milling area. The general ionic composition of some creeks peripheral to that area is also expected to change. Copper and other transition elements, while mainly essential at trace concentrations or in the diet, become toxic at greater concentrations which can profoundly alter the faunal composition of fresh waters.

Short-term toxicity experiments were run using water of the general ionic composition predicted to result from seepage. Temperatures were set at 27°C routinely owing to the high natural water temperatures of northern NT. The species studied were acclimated in the laboratory to the immediately pre-experimental conditions.

Two species of fish and one shrimp were studied which showed great differences in susceptibility: Neosilurus sp. was less susceptible than Melanotaenia sp. which was much less susceptible than shrimp Caridina sp. Other species of fish were not available in sufficient numbers or of sufficient quality. The shrimp's four day LC_{50} was approximately 15 µg L^{-1}

* Univ. of New England

free cupric ion at approximately 20 μg L^{-1} total copper.

A critical thermal maximum in nominally copper-free water at 38°C was found for Melanotaenia which was higher than many other tropical species. When copper was added in sub-lethal concentrations (negligible mortality at 27°C over the duration of exposure), this critical thermal maximum was reduced to the values (32-35°C) occasionally occurring naturally in the Magela Creek system. A short-term toxicity experiment, directly comparing toxicity at temperatures well within the range of viable temperature, yielded a Q_{10} of approximately 2.

Melanotaenia sp. was more susceptible to copper in a naturally occurring water type than in the modified type predicted. A possible reason for this is that contamination by seepage (mainly Mg and SO_4) ameliorates the acute effects of the toxic components, apart from any other direct or indirect effects that the alterations in water quality will have on the composition of the aquatic fauna in the peripheral creeks.

This project was supported by funds from the Office of the Supervising Scientist.

5. RADIATION BIOLOGY

5.1 Effects of Phomopsin on Chinese Hamster Cells In Vitro

(J.K. Brown, M. Mountford)

In conjunction with Dr Y.A.E. Bick, University of Tasmania (AINSE Grant), a study was carried out of changes in the cellular kinetics of a mammalian cell line in the presence of the radiomimetic agent phomopsin (P). Because there is some evidence that P inhibits cell proliferation by acting as a metaphase-blocking agent, the well known spindle poison colcemid (C) was used as a comparative index in the present study.

Figure 6 shows that the cell count of sequentially harvested, untreated (control) cultures showed a steady increase in cell numbers with time in culture. In contrast, the proliferation index in cultures exposed to P and C at 1 μg mL^{-1} was depressed and, at 10 μg mL^{-1}, there was little or no proliferation at all. Hence, P is an inhibitor of cell growth in vitro.

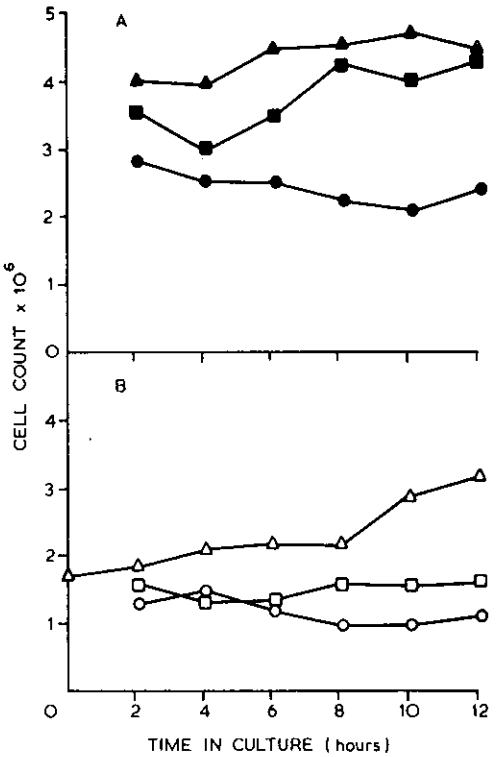

FIGURE 6 CELL COUNTS IN TWO-DAY CULTURES OF CHINESE HAMSTER CELLS EXPOSED FOR VARYING PERIODS OF TIMES TO PHOMOPSIN (P) AND COLCEMID (C) COMPARED WITH UNTREATED CELLS (CONTROL) (Panel A, 1 μg mL^{-1} P, ■ ; 1 μg mL^{-1} C, ● ; control, ▲ . Panel B, 10 μg mL^{-1} P, □ ; 10 μg mL^{-1} C, ○ ; control, △ .)

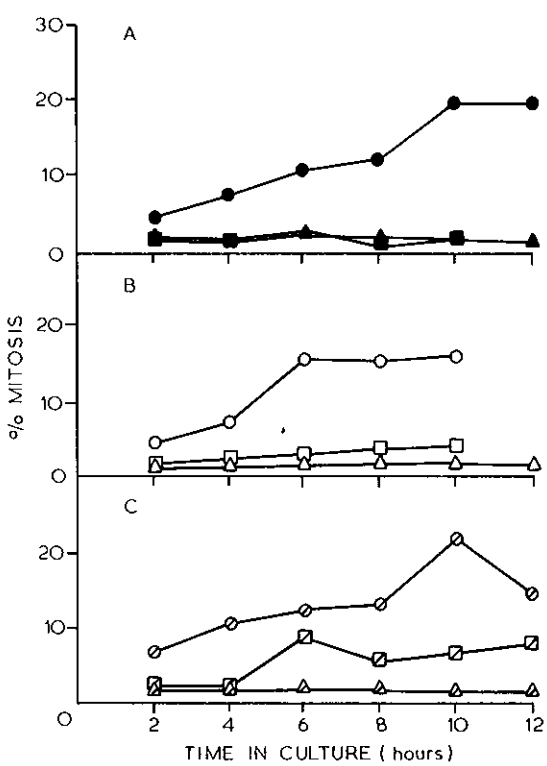

FIGURE 7 PERCENTAGE OF MITOTIC CELLS IN CULTURES OF CHINESE HAMSTER CELLS EXPOSED TO PHOMOPSIN (P) AND COLCEMID (C) COMPARED WITH UNTREATED CELLS FOR VARYING PERIODS OF TIME (Panel A, 1 μg mL^{-1} P, ■ ; 1 μg mL^{-1} C, ● ; control, ▲ . Panel B, 5 μg mL^{-1} P, □ ; 5 μg mL^{-1} C, ○ ; control, △. Panel C, 10 μg mL^{-1} P, ⊠ ; 10 μg mL^{-1} C, ⊘ ; control, ◬ .)

Figure 7 illustrates that, at 1 μg mL^{-1}, P had no significant effect as a metaphase-arresting agent, a slight effect at 5 μg mL^{-1} and, at 10 μg mL^{-1}, an approximately four to five times increase in mitosis from six hours onwards. Comparatively, cultures exposed to C, even at 1 μg mL^{-1}, showed a massive increase of cells in mitosis. It would appear, therefore, that the inhibitory effect of P on the cells is due, in part at least, to the blocking of cells in mitosis.

The pattern of DNA synthesis was determined by pulse-labelling cultures with tritiated (^3H) thymidine. Liquid scintillation counting (Figure 8) on serial cultures, harvested two-hourly over one cell cycle, showed that, while control cultures showed a steady rate of DNA synthesis, there was a marked uptake of ^3H-thymidine in cultures exposed to P and C at 1 and 10 μg mL^{-1}. Similarly, the percentage of cells in the S phase of DNA synthesis (labelling indices determined by autoradiography) also showed an increase in sequentially harvested cultures exposed to 1, 5 and 10 μg mL^{-1} of P or C (Figure 9). An elevated grain count was also observed, lasting four to six hours and then decreasing at ten hours to a value below that of the control cultures. Thus, there is evidence of increased DNA synthesis in hamster cells exposed to P, both in the number of cells in DNA synthesis in S phase (increased labelling index) and in the rate of DNA synthesis per cell (increased grain count).

Unscheduled DNA synthesis (UDS), i.e. cells in G1 and G2 of the cell cycle, was undertaken by exposing density-inhibited cultures in arginine-free medium to P at a concentration of 10 μg mL^{-1} for two and a half hours. Results obtained so far, by liquid scintillation counting and autoradiography, have been equivocal, with one experiment demonstrating a statistically significant increase in UDS in exposed cultures, and a second identical experiment showing no increase. Further experiments are in progress.

Cells exposed to C at 10 μg mL^{-1} for ten hours showed a chromosomal aberration rate of 0.17 breaks per cell; this value is similar to the spontaneous aberration rate found in hamster cells routinely exposed to a two-hour block with 0.05 μg mL^{-1} of C. On the other hand, cells exposed to 10 μg mL^{-1} of P showed an aberration rate of 0.37 and 0.43 breaks per cell at six and ten hours, respectively, and values of 0.47 and 0.52 at similar times in cultures exposed to 20 μg mL^{-1} of P. All the aberrations observed were of the chromatid type. Thus P is a clastogenic agent like radiation.

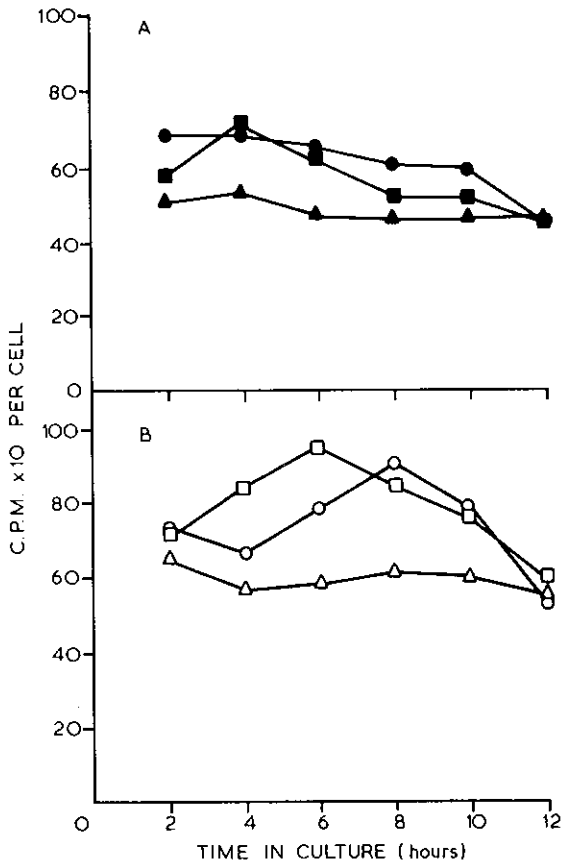

FIGURE 8 UPTAKE OF TRITIATED THYMIDINE, EXPRESSED AS C.P.M. PER CELL, IN CULTURES OF CHINESE HAMPSTER CELLS EXPOSED TO PHOMOPSIN (P) AND COLCEMID (C) COMPARED WITH UNTREATED CELLS FOR VARYING PERIODS OF TIME (Panel A, 1 μg mL^{-1} P, ■ ; 1 μg mL^{-1} C, ● ; control, ▲ . Panel B, 10 μg mL^{-1} P, □ ; 10 μg mL^{-1} C, ○ ; control, △ .)

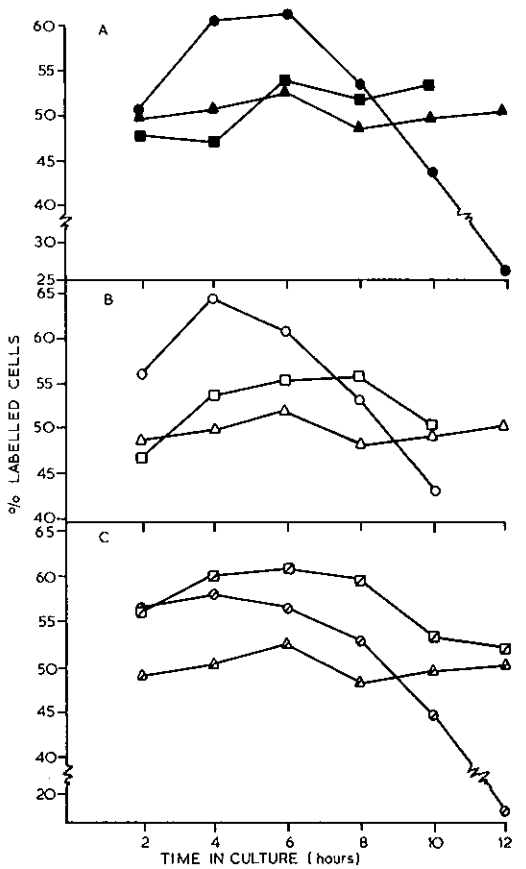

FIGURE 9 PERCENTAGE OF ^{3}H-LABELLED CELLS FOUND IN CULTURES OF CHINESE HAMSTER CELLS EXPOSED TO PHOMOPSIN (P) AND COLCEMID (C) COMPARED WITH UNTREATED CELLS FOR VARYING PERIODS OF TIME (Panel A, 1 μg mL^{-1} P, ■ ; 1 μg mL^{-1} C, ● ; control, ▲ . Panel B, 5 μg mL^{-1} P, □ ; 5 μg mL^{-1} C, ○ ; control, △ . Panel C, 10 μg mL^{-1} P, ⊠ ; 10 μg mL^{-1} C, ⊗ ; control, ⊗ .)

Amongst the drugs commonly used today in cancer chemotherapy are the metaphase-blocking agents, vincristine and vinblastine. In view of the similarity of action of P found in the present study, it may be worthwhile to investigate the potential usefulness of P as an anti-cancer agent.

5.2 Chromosome Aberrations in Peripheral Blood Lymphocytes after Partial Body Exposure to Radiation

(J.K. Brown, M. Mountford)

After an acute exposure to penetrating radiation, accepted practice in biological dosimetry has been to collect a blood sample 24 hours after the exposure. It has been assumed, but not experimentally confirmed, that by this time the lymphocytes of the lymphoid tissue in the irradiated area and those of the peripheral blood will be homogeneously mixed and sampling will then be representative.

To determine the optimum sampling time, we propose to determine the incidence of chromosome aberrations in serial blood samples taken over the 24-hour period post-irradiation (at five minutes and 1.5, 3, 6 and 24 hours). Samples will be taken from radiotherapy patients undergoing 2 Gy daily doses of 6 MeV electrons to four areas of the body, namely, breast, head-neck, pelvis and sacroiliac region. Samples will be collected after the fifth dose of radiation by which time the number of chromosome aberrations, such as dicentrics, will be sufficiently high to give some sort of statistical accuracy.

Figure 10 shows the results on the first three patients. Parallel blood counts have revealed that there is considerable perturbation in the absolute lymphocyte count during the three hours after irradiation. Consequently, it is not surprising to find an oscillation in the frequency of dicentric chromosome aberrations reaching a peak at three to six hours and then falling to a 24-hour value not very different from that found at five minutes.

Confirmation of these preliminary results would indicate that samples taken 24 hours after partial body radiation exposure will give an accurate estimate of the biological dose.

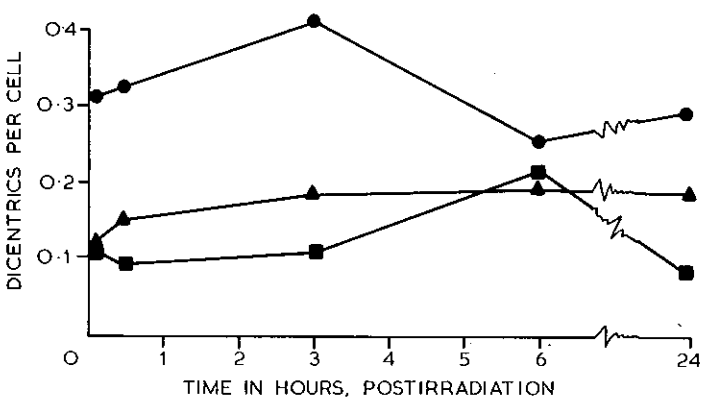

FIGURE 10 NUMBER OF DICENTRICS FOUND IN BLOOD SAMPLES FROM THREE MALE SUBJECTS G.D. (●), T.N. (■) and R.B. (▲) COLLECTED AT VARIOUS TIME INTERVALS AFTER THE FIFTH DOSE OF RADIATION (5 x 2 Gy) TO THE SACRO-ILIAC REGION (●,■) OR THE HEAD-NECK AREA (▲). THREE HUNDRED METAPHASES SCORED EACH POINT, 48-HOUR CULTURES.

5.3 Radiation Dose-Response of Amphibia

(H.C. Panter, J.E. Chapman)

A species of frog (Limnodynastes tasmaniensis) which can be reared in the laboratory is being used in radiation studies. As this frog will breed year-round, given favourable environmental conditions, it is well suited for experimental studies. The collaborating worker in the project, Mr M.J. Tyler of the University of Adelaide, supplied wild-caught adults, which have been supplemented by frogs reared at AAEC. A fibreglass breeding chamber with recirculating water and a diurnal lighting system houses the 40 or so breeding frogs, which are fed on mealworms (larvae of Tenebrio molitor).

The early developmental stages have been subjected to acute X- or γ-rays and survivals determined. Preliminary results indicate a steady decrease in radiation sensitivity from the four-cell stage to the stage when gill circulation begins. The LD_{50} increase from around 50 rads at the four-cell stage to between 1000 and 1500 rads at the gill circulation stage. Sensitivity does not appear to change much after this stage. However, irradiation at the gill circulation stage has produced abnormalities, such as polydactyly and ectromely in subsequently formed digits, even though the limb rudiments were not formed at the time of irradiation. An experiment to test the relative effectiveness of γ-rays from cobalt-60, and X-rays, is in progress.

Two experiments have been carried out using chronic γ-irradiation from a caesium-137 source. These experiments have shown a higher than expected effect from chronic γ-irradiation, but will have to be verified by further work.

Methods for skeletal staining (bone and cartilage) in froglets and frogs have been developed, and techniques for preparing numbers of chromosome spreads from irradiated and normal tadpoles and frogs are being improved. Methods used by other workers for amphibian chromosomes have to be modified to yield the large numbers of metaphases required for radiation studies. Cell cultures from various organs have proved quite successful, especially those derived from the heart.

6. PUBLICATIONS AND CONFERENCE PRESENTATIONS

6.1 Journal Papers

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