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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

WAD, A PROGRAM TO CALCULATE THE HEAT PRODUCED BY ALPHA DECAY

WAD, programme permettant de calculer la chaleur produite par désintégration alpha

R.G. JARVIS and C.I. BRETZLAFF

Chalk River Nuclear Laboratories Laboratoires nucléaires de Chalk River

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Résumé

Le programme FORTRAN appelé WAD (Watts from Alpha Decay) concerne les chaînes de désintégration alpha et bêta que l'on trouve dans les cycles de combustible avancé destinés aux réacteurs CANDU. La bibliothèque des données couvre tous les nucléides nécessaires à émission alpha ou bêta et le programme permet de calculer la chaleur produite par désintégration alpha. Toute chaîne admissible peut être construite très simplement.

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ABSTRACT

The FORTRAN program WAD (Watts from Alpha Decay) deals with the alpha and beta decay chains to be encountered in advanced fuel cycles for CANDU reactors. The data library covers all necessary alpha-emitting and beta-emitting nuclides and the program calculates the heat produced by alpha decay. Any permissible chain can be constructed very simply.

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1. Introduction

WAD (Watts from Alpha Decay) is a FORTRAN program to deal with the alpha and beta decay chains that can be encountered in advanced fuel cycles for CANDU reactors.

For a given decay chain starting with a given concentration of parent nuclide at time zero, the program will calculate the concentrations of the progeny nuclides at any desired times, and will also calculate the heat produced by absorption of the alpha particles (assumed to be complete).

The data library holds the half-lives and alpha energies of all nuclides likely to be of any significance.

The four main series, involving uranium, thorium, actinium or americium, can readily be set up, or sub-sections of them can be chosen.

The input for a particular sequence is very simple - just a list of the nuclides in the path to be followed, with a simple 'alpha' or 'beta' statement for each transition.

2. Analytical solutions

Consider a decay sequence starting with nuclide r=1, with progeny nuclides r=2 through r=m. The last one of interest is not necessarily stable.

The concentration of the parent nuclide is $N_1(0)$ at time t=0, and at that time all other concentrations $N_r(0)$ are zero.

If the decay constants are (λ_1 --- λ_m), the decays are described by the well-known coupled differential equations

$$\frac{d}{dt} N_1(t) = -\lambda_1 N_1(t)$$

$$\frac{d}{dt} N_2(t) = \lambda_1 N_1(t) - \lambda_2 N_2(t)$$

$$\frac{d}{dt} N_m(t) = \lambda_{m-1} N_{m-1}(t) - \lambda_m N_m(t)$$

The solutions for r>3 were first described in 1910 by Bateman(1), but a more convenient description can be obtained in Evans(2).

The concentration of nuclide r, at time t, can be written

$$N_r(t) = N_1(0) \left[H_{r1} \exp(-\lambda_1 t) + H_{r2} \exp(-\lambda_2 t) \right.$$

$$\left. + \text{--------} + H_{rr} \exp(-\lambda_r t) \right]$$

where the H coefficients can be calculated in a systematic fashion very suitable for a computer.

Define the product: $P_r = \lambda_1 \lambda_2 \lambda_3 \text{----} \lambda_r$.

Then,

$$H_{r1} = P_r \left[(\lambda_r - \lambda_1)(\lambda_2 - \lambda_1) \text{ ---- } (\lambda_{r-1} - \lambda_1) \right]^{-1}$$

$$H_{r2} = P_r \left[(\lambda_1 - \lambda_2)(\lambda_r - \lambda_2) \text{ ---- } (\lambda_{r-1} - \lambda_2) \right]^{-1}$$

$$H_{rr-1} = P_r \left[(\lambda_1 - \lambda_{r-1})(\lambda_2 - \lambda_{r-1}) \text{ ---- } (\lambda_r - \lambda_{r-1}) \right]^{-1}$$

$$H_{rr} = P_r \left[(\lambda_1 - \lambda_r)(\lambda_2 - \lambda_r) \text{ --------- } (\lambda_{r-1} - \lambda_r) \right]$$

Thus, for the rth nuclide, there are r of the H-factors and hence r products (H.exp), and each H-factor contains r-1 of the λ-differences.

It is relatively straightforward to calculate the H's and hence the N's, but unfortunately there are extremely severe numerical problems. They are discussed in the next section.

3. Numerical problems

Numerical problems appear in the form of extreme cancellation of significant figures, when t is small or - more generally - where λt products are small. The trouble lies in the derivatives of $N_r(t)$, when t is small.

One can prove by induction, using the original differential equations, that the first (r-2) derivatives of $N_r(t)$ are zero at t=0, for all $r \geq 2$.

The behaviour of $N_r(t)$ at small t can be demonstrated by its Taylor expansion about t=0. The vanishing of progressively higher derivates for progeny lower down the chain is basically the reason for the severe numerical cancellation. Although the identically-zero terms can be identified and removed from the series, it is not possible to isolate such elements in the original calculation. In practice, it is not easy to resort to a Taylor series for small t because the expressions for the higher derivatives become very complicated.

Finally, a brute-force stratagem was adopted. The H's and the $\exp(-\lambda t)$ were computed in double precision with about 28 significant figures. Although inelegant, this allows the loss of about 22 significant figures and has proved to be adequate.

4. Nuclides involved

The required series involve elements from thallium to californium, with proton numbers from 81 through 98 and neutron numbers from 124 through 150.

5. Decay paths

Three of the four series possess alternative paths, a split occurring at a nuclide that can decay by either alpha or beta.

Consider the forked sequence:

Nuclide A decays to nuclide B which then decays either to nuclide C (a fraction f_C of the decays) or to nuclide D (a fraction f_D of the decays).

This can be replaced by the two simple paths.

1) The first starts with a fraction f_C of the original concentration of nuclide A. The decay then goes from A to B and from B to C

2) The second starts with a fraction f_D of the original concentration of nuclide A. The decay goes from A to B and then from B to D.

A sequence with two forks can be broken down into four simple paths, and so on.

6. Conversion factors

1 day = 86400 seconds
1 year = 365.256 days
= $3.155815\ 10^{7}$ seconds
1 MeV = $1.6021\ 10^{-13}$ joules
A decay constant λ in units of per-second
= (0.693147)/(half-life in seconds)

7. The basic data library, ACTDATA

ACTDATA, the basic data library holds five pieces of information for each nuclide:

Proton number Z; Element name;
Mass number M; Half-life;
Energy of emission if decay is alpha.

The data are stored on permanent file ACTDATA. Originally they were loaded in order, beginning with thallium and ending with californium. However, this is not essential because each nuclide carries complete identification and further entries can simply be added to the end of the file.

The half-lives and alpha energies were taken from Lederer and Shirley(3). They are listed in Table 1.

8. The working library, TRACDAT

The program actually calls on a working library which is arranged in more convenient matrix form. Each nuclide is located in the matrix by its proton number Z and neutron number N, where Z ranges from 81 through 98, and N ranges from 124 through 150.

In storage, to reduce un-used array space, the co-ordinates are shifted to $Z' = Z-80$ and $N' = N-123$.

9. The auxiliary program TRAC

TRAC is a simple auxiliary program that reads once through the data in serial storage in the basic library and re-orders it in matrix form for more convenient access. During the process the half-lives are converted to decay constants, λ's.

Finally, the matrix is stored on a permanent file, TRACDAT, to form the working library.

10. The main program WAD

10.1 Transfer of nuclide data

Before any work begins, the working library matrix is brought in complete, from the TRACDAT file.

10.2 Input

Each run deals with the decay chain from a particular initial nuclide. Four decay paths can be accommodated, with as many as twenty nuclides in each path. Ten time points are allowed.

The input, in card images, goes as follows.

a) A heading, to be printed in the output.

b) (Concentration of the initial nuclide), (its atomic weight), (the number of decay-paths), (the mass of initial nuclide to be used in the final wattage table).

Note that the mass does not have to correspond to the initial concentration - the final table is normalized appropriately.

c) A list of time points, in days.

The following sequences d) and e) are repeated for each path in the run

d) (the fraction of initial nuclide to be applied to the path), (the number of nuclides in the path).

e) For each nuclide in the path, starting with the parent, there is the triplet.
(element name), (nuclide mass number), (ALPHA or BETA as appropriate if it decays, or STABLE if it does not).

Inputs for different runs may be stacked one behind the other, and finally the card image 'FINISHED' will terminate calculation.

10.3 Calculations

For each path K, in turn.

Data is collected from the matrix.

The exponential factors $\exp(-\lambda_J t)$ are calculated (in double precision) and stored, for all nuclides J in the path, and for all time points.

For each nuclide J in the path K.

The H coefficients are calculated and the sum of the $H.\exp(-\lambda t)$ products (see Sections 2 and 3) is stored as $N_i(t)$.

As each path is processed the concentrations are sorted, and the corresponding rates of alpha energy emission. It is assumed that all alpha particles are completely absorbed - a very realistic assumption.

10.4 Output

For each time point, the output lists the concentration (in nuclei) and energy emission (in watts) for all nuclides. The wattage table is re-normalized to refer to the input mass of initial nuclide.

11. The sub-routine FID

FID is a very short sub-routine. It is called for each path K_i to handle the search for data in the data-matrix and to check that the nuclides in the path are in the library.

12. The sub-routine HCAL

HCAL calculates (in double precision) the J H-factors for the Jth nuclide in a path (see Section 2).

13. Program and sub-routine calls

Figure 1 shows the handling of data and calls of sub-routines.

14. A sample run

Tables 2 through 5 show a sample run. It is the forked sequence (U-233, Alpha), (Th-229, Alpha), (Ra-225, Beta), (Ac-225, Alpha), (Fr-221, Alpha), (At-217, Alpha), (Bi-213, 0.022 Alpha and 0.978 Beta)

The alpha-fork goes
(Bi-213, Alpha), (Tl-209, Beta), (Pb-209, Beta), (Bi-209, Stable)

The beta-fork goes
(Bi-213, Beta), (Po-213, Alpha), (Pb-209, Beta), (Bi-209, Stable).

The input of the two paths is shown in Table 2, and the output in Tables 3, 4 and 5.

15. Acknowledgement

We are indebted to R.S. Davis for some helpful discussion of the differential equations.

16. References

1) Bateman, H., **Solution of a system of differential equations occurring in the theory of radio-active transformations.** Proceedings of the Cambridge Philosophical Society, Volume XV, pp 423-427, (1910).

2) Evans, R.D., **The atomic nucleus, chapter 15.** McGraw HIll Book Co. Inc., New York (1955).

3) Lederer, C.M., Shirley, V.S., **Table of Isotopes.** 7th Edition. Lawrence Berkeley Laboratory, California (1978).

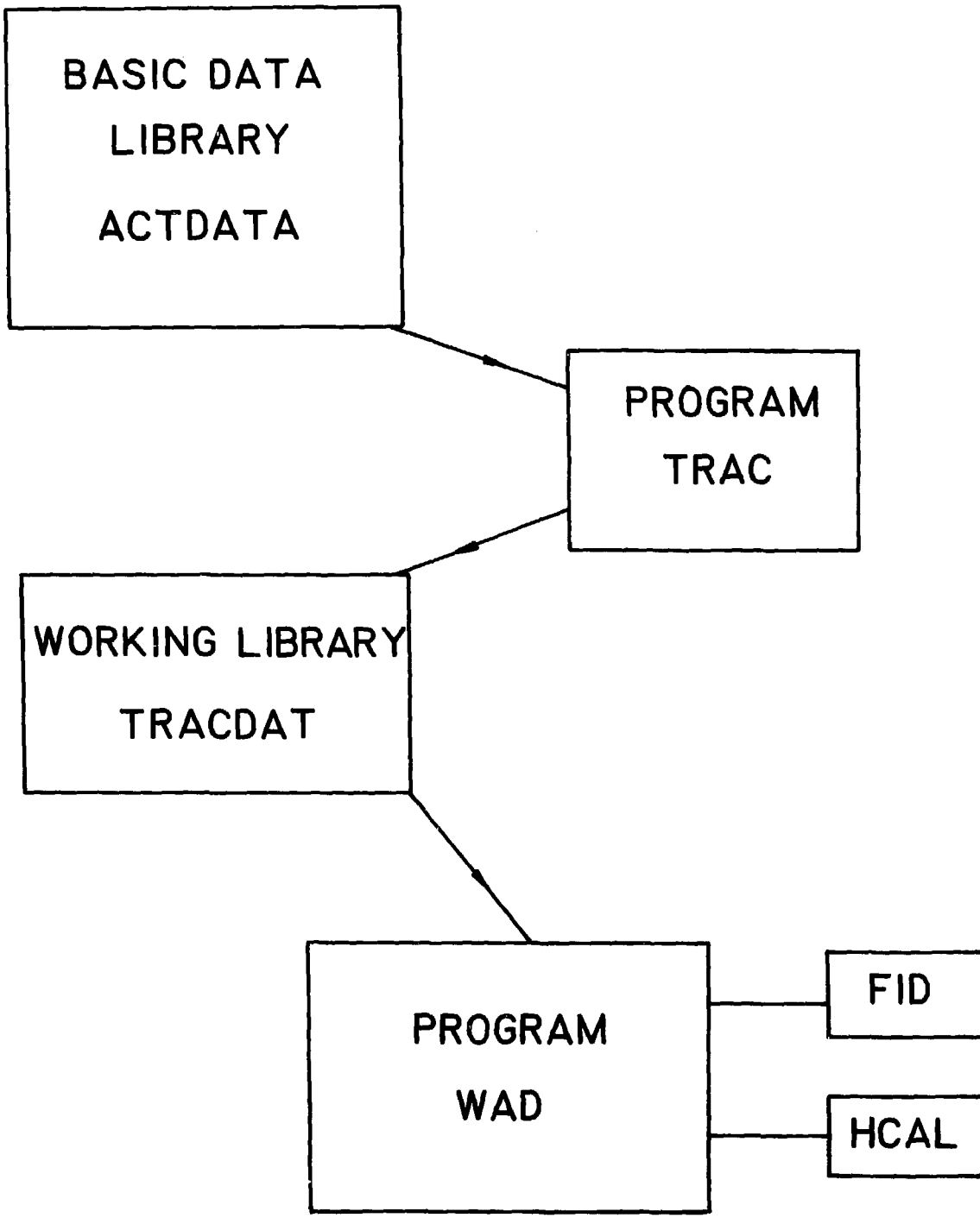

Figure 1. Program and sub-routine calls.

Table 1. Listing of basic data library

Units:

S	Seconds
M	Minutes
D	Days
Y	Years
E	Infinite ('Eons' to denote stability)

The fourth column contains alpha energy in MeV. A zero signifies beta decay if accompanied by a finite half-life, otherwise it accompanies stability.

DATA LIBRARY FOR WAD PROGRAM

Z	ELEMENT		HALF-LIFE		A-MEV
81	TL	207	4.790E+00	M	0.000
		208	3.053E+00	M	0.000
		209	2.200E+00	M	0.000
82	PB	206	0.	E	0.000
		207	0.	E	0.000
		208	0.	E	0.000
		209	3.250E+00	H	0.000
		210	2.226E+01	Y	3.720
		211	3.610E+01	M	0.000
		212	1.064E+01	H	0.000
		213	1.020E+01	M	0.000
		214	2.680E+01	M	0.000
83	BI	209	0.	E	0.000
		210	5.013E+00	D	4.690
		211	2.150E+00	M	6.568
		212	6.060E+01	M	6.984
		213	4.560E+01	M	5.869
		214	1.970E+01	M	5.459
84	PO	210	1.384E+02	D	5.305
		211	5.160E-01	S	7.442
		212	2.960E-07	S	8.784
		213	4.280E-06	S	8.375
		214	1.637E-04	S	7.687
		215	1.778E-03	S	7.386
		216	1.452E-01	S	6.779
		217	1.000E+01	S	6.540
		218	3.050E+00	M	6.003

Table 1. Part 1 Listing of basic data library

Z	ELEMENT		HALF-LIFE		A-MEV
85	AT	217	3.230E-02	S	7.067
86	RN	219	3.960E+00	S	6.772
		220	5.561E+01	S	6.288
		221	2.500E+01	M	6.000
		222	3.824E+00	D	6.440
87	FR	221	4.800E+00	M	6.301
		222	1.440E+01	M	6.000
		223	2.190E+01	M	6.340
88	RA	223	1.144E+01	D	6.030
		224	3.660E+00	D	6.674
		225	1.480E+01	D	6.000
		226	1.599E+03	Y	4.760
		227	4.220E+01	M	0.000
		228	6.770E+00	Y	0.000
89	AC	225	1.000E+01	D	6.010
		226	2.900E+01	H	6.400
		227	2.177E+01	Y	4.941
		228	6.130E+00	H	0.000
90	TH	227	1.872E+01	D	6.900
		228	1.913E+00	Y	6.360
		229	7.340E+03	Y	4.870
		230	8.000E+04	Y	4.620
		231	2.552E+01	H	0.000
		232	1.410E+10	Y	4.000
		233	2.230E+01	M	0.000
		234	2.410E+01	D	0.000

Table 1. Part 2 Listing of basic data library

Z	ELEMENT	HALF-LIFE	A-MEV
91	PA 231	3.276E+04 Y	4.980
	232	1.310E+00 D	0.000
	233	2.695E+01 D	0.000
	234	1.175E+00 M	0.000
92	U 230	2.080E+01 D	5.866
	231	4.200E+00 D	5.460
	232	7.170E+01 Y	5.302
	233	1.591E+05 Y	4.816
	234	2.446E+05 Y	4.762
	235	7.038E+08 Y	4.396
	236	2.341E+07 Y	4.478
	237	6.752E+00 D	0.000
	238	4.468E+09 Y	4.185
	239	2.354E+01 M	0.000
93	NP 236	2.250E+01 H	0.000
	237	2.140E+06 Y	4.783
	238	2.117E+00 D	0.000
	239	2.346E+00 D	0.000
94	PU 236	2.851E+00 Y	5.753
	237	4.530E+01 D	5.421
	238	8.771E+01 Y	5.487
	239	2.413E+04 Y	5.142
	240	6.570E+03 Y	5.152
	241	1.436E+01 Y	4.900
	242	3.763E+05 Y	4.890
	243	4.955E+00 H	0.000
	244	8.050E+07 Y	4.580

Table 1. Part 3 Listing of basic data library

Z	ELEMENT	HALF-LIFE	A-MEV
95	AM 241	4.329E+02 Y	5.479
	242	1.520E+02 Y	5.200
	243	7.370E+03 Y	5.249
96	CM 242	1.628E+02 D	5.101
	243	2.850E+01 Y	5.813
	244	1.810E+01 Y	5.795
97	BK 247	1.380E+03 Y	5.594
98	CF 246	3.530E+01 H	5.749

Table 1. Part 4 Listing of basic data library

```
          (4N+1) SERIES STARTING WITH U 233

  TIME PERIODS  DAYS          1.0     5.0     10.0    50.0    100.0   500.0   1000.0  5000.0 10000.0

  CONCENTRATION OF FIRST NUCLIDE IS   1.000E+20     ATOMIC WEIGHT IS  233.04


  EFFECTIVE FRACTION OF FIRST NUCLIDE IS   .022
B-CHAIN          U  233  ALPHA     TH 229  ALPHA     RA 225  BETA      AC 225  ALPHA     FR 221  ALPHA     AT 217  ALPHA
 213  ALPHA     TL 209  BETA      PB 209  BETA      BI 209  STABL


  EFFECTIVE FRACTION OF FIRST NUCLIDE IS   .978
B-CHAIN          U  233  ALPHA     TH 229  ALPHA     RA 225  BETA      AC 225  ALPHA     FR 221  ALPHA     AT 217  ALPHA
 213  BETA      PO 213  ALPHA     PB 209  BETA      BI 209  STABL
```

Table 2. Sample input - two paths

CONCENTRATION OF INITIAL NUCLIDE IS 1.000E+20 EFFECTIVE FRACTION IS .022

SUB-CHAIN NUCLIDE CONCENTRATION IN ATOMS VS TIME IN DAYS

NUCLIDES		1.0	5.0	10.0	50.0	100.0	500.0	1000.0	5000.0	10000.0
U	233	2.200E+18	2.200E+18	2.200E+18	2.200E+18	2.200E+18	2.200E+18	2.200E+18	2.200E+18	2.200E+18
TH	229	2.625E+10	1.313E+11	2.625E+11	1.312E+12	2.625E+12	1.312E+13	2.625E+13	1.312E+14	2.621E+14
RA	225	3.341E+03	7.857E+04	2.920E+05	4.448E+06	1.142E+07	6.934E+07	1.418E+08	7.206E+08	1.444E+09
AC	225	5.146E+01	5.740E+03	3.998E+04	1.923E+06	6.344E+06	4.544E+07	9.438E+07	4.856E+08	9.741E+08
FR	221	1.691E-02	1.908E+00	1.331E+01	6.399E+02	2.114E+03	1.515E+04	3.146E+04	1.619E+05	3.247E+05
AT	217	1.897E-06	2.140E-04	1.493E-03	7.177E-02	2.371E-01	1.699E+00	3.528E+00	1.816E+01	3.642E+01
BI	213	1.407E-01	1.766E+01	1.249E+02	6.070E+03	2.008E+04	1.439E+05	2.989E+05	1.538E+06	3.085E+06
TL	209	6.739E-03	8.510E-01	6.021E+00	2.928E+02	9.685E+02	6.942E+03	1.442E+04	7.420E+04	1.488E+05
PB	209	3.509E-01	6.750E+01	5.060E+02	2.575E+04	8.559E+04	6.151E+05	1.278E+06	6.577E+06	1.319E+07
BI	209	3.813E-01	4.238E+02	6.678E+03	2.081E+06	1.605E+07	7.326E+08	3.155E+09	8.357E+10	3.366E+11

Table 3. Sample output - concentrations in first path

CONCENTRATION OF INITIAL NUCLIDE IS 1.000E+20 EFFECTIVE FRACTION IS .978

SUB-CHAIN NUCLIDE CONCENTRATION IN ATOMS VS TIME IN DAYS

NUCLIDES		1.0	5.0	10.0	50.0	100.0	500.0	1000.0	5000.0	10000.0
U	233	9.780E+19	9.780E+19	9.780E+19	9.780E+19	9.780E+19	9.780E+19	9.780E+19	9.779E+19	9.779E+19
TH	229	1.167E+12	5.835E+12	1.167E+13	5.835E+13	1.167E+14	5.834E+14	1.167E+15	5.831E+15	1.165E+16
RA	225	1.485E+05	3.493E+06	1.298E+07	1.977E+08	5.078E+08	3.083E+09	6.352E+09	3.204E+10	6.418E+10
AC	225	2.288E+03	2.552E+05	1.777E+06	8.535E+07	2.820E+08	2.020E+09	4.196E+09	2.159E+10	4.330E+10
FR	221	7.517E-01	8.481E+01	5.916E+02	2.845E+04	9.399E+04	6.733E+05	1.398E+06	7.196E+06	1.443E+07
AT	217	8.432E-05	9.513E-03	6.636E-02	3.191E+00	1.054E+01	7.552E+01	1.567E+02	8.071E+02	1.619E+03
BI	213	6.253E+00	7.852E+02	5.552E+03	2.698E+05	8.925E+05	6.398E+06	1.329E+07	6.838E+07	1.372E+08
PO	213	9.777E-09	1.228E-06	8.682E-06	4.219E-04	1.396E-03	1.000E-02	2.073E-02	1.069E-01	2.145E-01
PB	209	1.572E+01	3.005E+03	2.251E+04	1.145E+06	3.805E+06	2.734E+07	5.681E+07	2.924E+08	5.865E+08
BI	209	1.715E+01	1.887E+04	2.371E+05	9.254E+07	7.137E+08	3.257E+10	1.402E+11	3.715E+12	1.496E+13

Table 4. Sample output - concentrations in second path

(4N+1) SERIES STARTING WITH U 233 1.000E+03 GRAMS

TOTAL ALPHA EMISSIONS IN WATTS VS TIME IN DAYS

NUCLIDES		1.0	5.0	10.0	50.0	100.0	500.0	1000.0	5000.0	10000.0
TL	209	BETA ONLY								
PB	209	BETA ONLY								
BI	209	BETA ONLY								
	213	8.658E-13	1.087E-10	7.688E-10	3.736E-08	1.236E-07	8.859E-07	1.843E-06	9.469E-06	1.899E-05
PO	213	5.492E-11	6.897E-09	4.877E-08	2.370E-06	7.840E-06	5.620E-05	1.167E-04	6.006E-04	1.205E-03
AT	217	5.414E-11	6.108E-09	4.261E-08	2.049E-06	6.769E-06	4.849E-05	1.007E-04	5.182E-04	1.040E-03
FR	221	4.827E-11	5.446E-09	3.799E-08	1.827E-06	6.035E-06	4.324E-05	8.983E-05	4.621E-04	9.268E-04
RA	225	BETA ONLY								
AC	225	4.521E-11	5.042E-09	3.512E-08	1.687E-06	5.573E-06	3.992E-05	8.292E-05	4.267E-04	8.558E-04
TH	229	7.199E-08	3.599E-07	7.199E-07	3.599E-06	7.199E-06	3.599E-05	7.198E-05	3.597E-04	7.189E-04
U	233	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01
TOTAL		2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.755E-01	2.757E-01	2.760E-01	2.779E-01	2.802E-01

Table 5. Sample output - total alpha emissions in watts