RADIOECOLOGICAL MODELS FOR ESTIMATING SHORT AND LONG-TERM EFFECTS OF RELEASES IN THE NUCLEAR FUEL CYCLE

Mats Nilsson and Bertil Persson

Radiation Physics Department, Lasarettet S-221 85 Lund, Sweden.

INTRODUCTION

The increasing use of nuclear energy will mainly through the production and reprocessing of nuclear fuel and the long-term disposal of the radioactive waste introduce increasing amounts of radionuclides into the biosphere. For the estimation of the radiological consequences when these radionuclides,sooner or later,reach man it is necessary to know not only how the nuclides are distributed and retained within the body,but also how the nuclides are transported in the biosphere before they directly,or through food-chains,reach man. The only way to study the transport from one environmental pool to another is the direct measurement of the activity concentration for the radionuclide of interest. Such measurements have now been carried out for a considerable time. The source of the radionuclides studied has up to now mainly been fall-out from nuclear weapons tests. Because of this,it has been possible to study only a limited number of radionuclides and still a great deal of experimental work concerning the behaviour of long-lived fission products and transuranium elements,other then plutonium,remains. The results can be used to design a more complex system for transport in the biosphere or in the human body. Such systems,normally referred to as compartmental systems can be used to predict individual and collective dose equivalent arising from the use of some nuclear energy practice.

However,due to the lack of reliable data for many radionuclides regarding their behaviour in the biosphere the use of mathematical models is at present in most cases limited to yield upper limits of the parameters studied rather than quantitative estimation of these parameters. For some applications,especially regarding the disposal of nuclear waste, calculations extending millions of years ahead are used. For static systems, the reliability of the transport factors used will in such cases be of vital importance. The need for efficient mathematical methods of solution is also evident.

A GENERAL MATHEMATICAL MODEL

For a compartment system consisting of n compartments the net transport rate of the substance y into the compartment i can be expressed using the general expression

$$\frac{dy_i(t)}{dt} = \sum_{j=1}^{n} y_j k_{ji}(t) - \sum_{j=1}^{n} y_i(k_{ij}(t) + \lambda) + \frac{ds_i(t)}{dt} \qquad \{1\}$$

where k is the transfer coefficient expressing the fraction of substance y that per unit time is transferred from compartment i to compartment j and λ is the physical decay constant. The last term corresponds to the presence of a source of the substance in compartment i. A somewhat simpler way of expressing equation {1} is a notation of the form

$$\dot{\vec{Y}}(t) = \vec{Y}(t) \cdot K(t) + \dot{\vec{S}}(t) \qquad \{2\}$$

where K is the transfer coefficient matrix and $\dot{\vec{S}}$ is the source term vector. The method of solving equation {2} will strongly depend on the time variation characteristics of the matrix K and the vector $\dot{\vec{S}}$. Some, or all,of the transfer coefficients and elements in the source vector can independently be chosen to continously vary with time following, for instance,some given function or polynomial approximation of given points. Furthermore, these terms can behave in a stochastic manner. The generation of the specific values during the calculations is preferrably done with Monte Carlo technique. The number of transfer coefficients with stochastic behaviour must,however,be limited due to the very long computer time to obtain good statistics.

In both of these cases with a dynamic coefficient matrix and source vector,the change in numerical value of the derivatives may be so great that the step in time when performing the integration of the equation system must be made very short. Iterative methods are therefore recommended, as the Kutta-Merson algorithm or the Adams-Moulton predictor-corrector scheme.

In the case of a static coefficient matrix and source term it is possible to use another algorithm to solve the equation {2}. (1) The solution to this set of equations will be of the form

$$\vec{Y}(t) = \vec{Y}(0) \cdot e^{K \cdot t} = \vec{Y}(0) \cdot B \qquad \{3\}$$

The quantity B is also a matrix, which can be evaluated from the matrix K using serial expansion. The main problem when performing this, is that a large number of terms must be included if some eigenvalues of the matrix K·t are large. This can be avoided if the resulting serial expansion matrix is multiplied with a geometrical series which is chosen in a way that only a limited number of terms in the serial expansion will be used. The inverse quotient of this series is used to convert the resulting matrix by a binomial expansion resulting in the matrix B. Using the solution {3} instead of an iterative method will make the calculations run up to 10 000 times faster on a computer depending on the characteristics of the coefficient matrix. Figure 1 represents a simple system which is a good test on the precision in the algorithm used, especially for compartments 1 and 2. Table 1 gives the amount of substance in compartment 2 after a time of 2 time-units after a pulsed injection in compartment 1 for some different mathematical methods. It is a well known fact that the Kutta-Merson and Adams-Moulton methods both are very slow giving, however, excellent precision.

Figure 1.

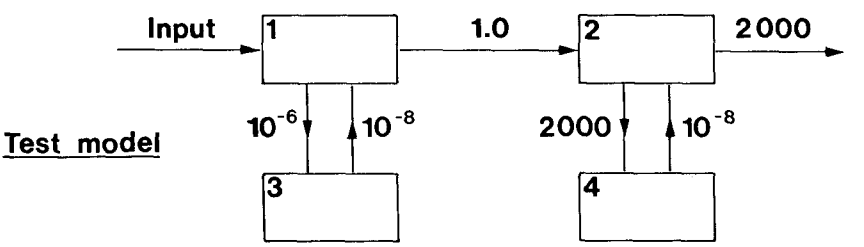

Table 1

Method of solution	Step-length	Amount of substance	Rel. error (%)	CPU time (s)
Analytical	-	$5.81506 \cdot 10^{-5}$	-	-
Kutta-Merson	h	$5.81505 \cdot 10^{-5}$	-	360
Adams-Moulton	h	$5.81506 \cdot 10^{-5}$	-	310
Matrix exponentiation	h	$5.76877 \cdot 10^{-5}$	-0.8	1.1
Matrix exponentiation	h/20	$5.81486 \cdot 10^{-5}$	-0.003	16
Matrix exponentiation	10 h	$4.96676 \cdot 10^{-5}$	-14.6	0.6

The matrix exponentiation mentioned above will, for the same step-length give a result within 1% of the exact value with a substantial decrease in computer time. It can also be seen that this algorithm must be used with caution. An increase in step-length immediately reduces the precision to an unacceptable level. For the same precision as the two iterative methods, the computing time increases but is still much shorter.

EXAMPLES OF APPLICATION

A field where mathematical models are frequently applied is on the spreading of radionuclides from the disposal of high-level radioactive waste. We have studied the two ways of disposal proposed by the Swedish Nuclear Industry (2,3).

The first alternative concerns the deposition of waste from the reprocessing of 10000 tons of UO_2 fuel. The second deals with the direct deposition of the burned-out fuel. The deposition takes place in primary rock at a depth of 500 m. The main difference between the two alternatives is the encapsulation material which for the latter mainly is copper. In this case the capsules are assumed to withstand corrosion for a certain time (100 000 years),after which the radionuclides during 500 000 years leak out into the biosphere. On their way to the first recipient in direct contact with the biosphere they are being delayed in the primary rock.

We have applied our technique on a 20-compartment global model to study the effects of earlier breakdown of the capsule and a faster leaking rate, (4). Furthermore, in order to yield the "upper limits" we have taken no notice of a possible delay in the primary rock. A combination of these factors are of particular interest for the daughters in decay chains. It is almost impossible without computers to calculate their input activity rate as a function of time due to the cooperating factors of capsule breakdown and leaking rate on the amount which at that time is built up in the capsule. An example illustrating this is the 4n+1 series. Np-237→Pa-233→U-233→Th-229. In table 2, the relative activity inflow rate of Th-229 into the primary recipient normalized to a capsule breakdown at 100 000 years after deposition and a leakage time of 500 000 years is given.

The combined effects of breakdown time and leaking rate can clearly be seen in the bottom row, which has a quite irregular pattern due to the balance between leaking rate and buildup during leaking time. These figures are also proportional to the individual absorbed doses

Table 2

Th-229	Leaking time (years)			
Capsule breakdown (year)	500 000	50 000	5 000	500
100 000	1.0(225000)	7.29(100000)	72.9(100000)	729(100000)
10 000	0.77(210000)	1.16(35000)	3.48(10000)	34.8(10000)
1 000	0.76(201000)	0.80(31000)	0.27(4000)	0.46(1000)

The figures in parenthesis indicate the time (in years) when maximum inflow occurs.

from thorium in the vicinity of the waste site. For the collective doses thorium is of particular interest due to its high sedimentation capability. The collective doses arising from thorium isotopes are not proportional to the amount that is directly injected into the shallow and deep ocean water compartments, nor to the small amount that is resuspended from the sediments, but only to the amount that is produced from the decay of its mother nuclide in these compartments.

SUMMARY AND CONCLUSION

Mathematical models are, if used with caution, an important tool for evaluating effects of releases of radionuclides. For some elements, very little is known regarding their behaviour in the biosphere, and complementary experimental work must be carried out. The non-static and stochastic nature of the biosphere must be taken into account regarding the transport of substances, thus having demands on the algorithms to be increasingly efficient.

Appendix: Brief description of algorithm used.

$$e^{K \cdot t} = \left(1 + K \cdot t + \frac{(K \cdot t)^2}{2!} + \frac{(K \cdot t)^3}{3!} + \ldots\ldots\ldots\right) = \sum_{n=0}^{\infty} \frac{(K \cdot t)^n}{n!}$$

$$\sum_{n=0}^{\infty} \frac{(K \cdot t)^n}{n!} \approx \left(\sum_{n=0}^{m} \frac{(K \cdot t)^n}{n!} \cdot \frac{1}{\beta^n}\right)^{\beta}, \text{ where } \beta^m \geq \frac{(K \cdot t)^m \cdot 10^m}{m!}$$

REFERENCES:

1) Nilsson,M.and Persson,B.: Mathematical models for simulation of processes of radioecological and biomedical transport of radionuclides. Part I: Numerical methods. To be published.
2) Kärnbränslecykelns slutsteg: Förglasat avfall från upparbetning. KBS 1978 (in swedish).
3) Kärnbränslecykelns slutsteg: Slutförvaring av använt kärnbränsle. KBS 1979 (in swedish).
4) Nilsson,M. and Persson, B.: Redogörelse avseende granskning av KBS-rapporten "Slutförvaring av använt kärnbränsle". Report to the Swedish National Institute of Radiation Protection (in swedish).