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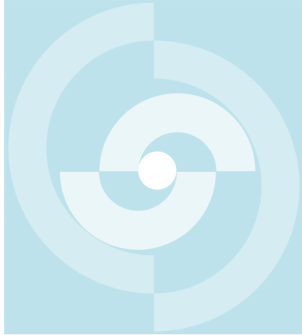
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# TECHNICAL REPORT 85-16

CONCENTRATION RATIOS FOR BIOPATH:  
SELECTION OF THE SOIL-TO-PLANT  
CONCENTRATION RATIO DATABASE

H. A. Grogan

January 1985

Swiss Federal Institute for Reactor Research, Würenlingen



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Der vorliegende Bericht wurde im Auftrag der Nagra erstellt. Die Autoren haben ihre eigenen Ansichten und Schlussfolgerungen dargestellt. Diese müssen nicht unbedingt mit denjenigen der Nagra übereinstimmen.

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SUMMARY

In Switzerland, high-level radioactive waste is planned to be deposited in deep crystalline formations. "BIOPATH" is the computer code which has been used to simulate the movement of radionuclides in the biosphere, in order to calculate the doses to man. The key parameter used in the model on which the code is based is the concentration ratio (CR). This specifies the transfer of individual radionuclides via the various foodchains, ultimately to man. Five types of CR are required for the model, namely soil to plant, intake by cattle to milk, intake by cattle to meat, intake by poultry to eggs and from water to fish. Each of these CR are discussed in turn; it is shown that soil to plant CR exhibit a variability that greatly exceeds that observed in the other types of CR and for this reason the report goes on to consider these in much closer detail. The different methods and experimental techniques used to derive soil to plant CR are examined and assessed, to allow evaluation of a meaningful data base for use in BIOPATH. The report concludes by examining the data available for each radionuclide in turn and listing a recommended CR for it, to input into the BIOPATH model.

ZUSAMMENFASSUNG

Die Endlagerung hoch-radioaktiver Abfälle ist in der Schweiz in tiefen kristallinen Formationen vorgesehen. "BIOPATH" nennt sich das Computer-Programm zur modellmässigen Simulation der Radionuklid-Wanderung in der Biosphäre, das dazu dienen soll, die Personendosen zu berechnen. Der Schlüsselparameter, der im Modell eingesetzt wird auf welchem das Programm beruht, ist das Konzentrations-Verhältnis (CR). Dieses beschreibt den Transfer einzelner Radionuklide durch die verschiedenen Nahrungsmittel-Ketten bis sie schliesslich zum Menschen gelangen. Fünf verschiedene CR-Werte sind für das Modell notwendig; nämlich: Boden zu Pflanze, Futteraufnahme des Viehs zu Milch, Futteraufnahme des Viehs zu Fleisch, Aufnahme durch Geflügel zu Eier und Wasser zu Fisch. Jedes dieser Konzentrations-Verhältnisse wird der Reihe nach definiert. Die CR-Werte für Boden zu Pflanze zeigen eine Bandbreite, die diejenige, welche für die anderen Typen von CR beobachtet werden, massiv übertrifft. Deshalb werden die Boden-Pflanzen CR-Werte im vorliegenden Bericht detaillierter untersucht. Die verschiedenen Methoden und experimentellen Techniken die eingesetzt werden, um Werte für das Verhältnis Boden zu Pflanze zu bestimmen, werden untersucht, geprüft und bewertet, um die Evaluation einer belastbaren Datenbasis für den Einsatz in "BIOPATH" zu ermöglichen. Der Bericht schliesst mit der Ueberprüfung der vorliegenden Daten und der Auflistung eines empfohlenen CR-Wertes für jedes einzelne Radionuklid als Eingangsdaten für das BIOPATH-Modell.

RESUME

En Suisse il est prévu de stocker définitivement les déchets hautement radioactifs dans des formations cristallines profondes. "BIOPATH" est le code d'ordinateur qui a été utilisé pour simuler le transport des radionucléides dans la biosphère, afin de calculer les doses d'irradiation individuelles. Le paramètre-clé du modèle sur lequel le code est basé est le quotient de concentration (CR). Ce dernier détermine le transfert des radionucléides individuels via diverses chaînes alimentaires pour aboutir finalement l'homme. Cinq types de CR sont nécessaires au modèle, / savoir les quotients: plantes/sol, lait/fourrage (pour le bétail laitier) viande/fourrage (pour le bétail de boucherie) oeufs/becquée (pour la volaille) et enfin poisson/eau. On discute les CR un par un; on montre que plante/sol offre une variabilité qui dépasse largement celle que l'on observe pour les autres types de CR, sa pourquoi le présent rapport le discute en détail. Les diverses méthodes et techniques expérimentales utilisées pour déduire le CR plantes/sol sont examinées et discutées critiquement, afin de permettre l'évaluation d'une base de données significative en vue de son utilisation dans BIOPATH. Le rapport se termine sur l'examen des données disponibles ainsi que sur la présentation d'un CR approprié pour chacun des radionucléides, en vue de son utilisation dans le modèle BIOPATH.

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## 1. CONCENTRATION RATIO

A key parameter in the foodchain calculations with the BIOPATH code for the biosphere transport is the concentration ratio (CR). It specifies the transfer of individual radionuclides in foodchains, from soils to plant products, and from feed or forage and drinking water to animal products and ultimately to man. The model requires five types of concentration ratio:

- i) soil to plant
- ii) intake by cattle to milk
- iii) intake by cattle to meat
- iv) intake by poultry to eggs
- v) water to fish.

Because the definition of the CR to all animal products is basically similar, only three types of ratio need to be defined:

- i) soil to plant
- ii) animal feed to animal products
- iii) water to fish

Soil to plant CR exhibit a variability that far exceeds that observed in CR for animal products (49). The main reason for this, is that the uptake of an isotope from soils by plants depends on numerous interrelated soil properties, such as clay content, dominant clay mineral, organic matter content, cation exchange capacity, pH etc. It also varies with chemical and physical form of the radionuclide, plant species, plant part and stage of growth, as well as with experimental conditions.

In view of this soil to plant concentration ratios shall be discussed in considerable detail in section 2 in order to allow a clearer understanding of them.

### 1.1 Soil to Plant

The CR for a particular radionuclide from soil to plant is expressed as the quotient

$$CR = \frac{\text{Bq}/(\text{kg plant fresh weight})}{\text{Bq}/(\text{kg dry soil})} \quad (1)$$

For this, the radionuclide activity in the edible portion of the plant is taken and the CR is independent of units as it is a simple ratio of concentrations. The soil and soil solution are assumed to be in equilibrium as are plant concentration and soil concentration. In general CR are based on the radioactivity present in the 0-10cm layer for pasture and leys (representing rooting depth) and in the 0-20cm layer for arable land (ploughing depth).

It should be noted that many authors have derived the CR for dry plant material to reduce the variability in the measured results. The CR for dry weight is frequently an order of magnitude higher than for fresh crop weight, depending on the crop moisture. These dry weight values are converted to fresh weight for BIOPATH assuming the water content for specific crop types, when used for consumption, as shown in table 1.

Table 1: Moisture Content of Edible Crop Part

<u>Crop</u>	<u>% Water</u>
Green vegetables	90
Root vegetables	85
Cereals	11
Pasture	75

Similarly, concentrations based on wet soil have been converted to dry soil values by assuming a water content of 20%.

## 1.2 Animal Products

The definition of animal concentration ratios is best exemplified by the concentration ratio to milk. These CR are described as distribution factors (DF) in the BIOPATH code and for a given radionuclide given as:

$$DF_m = \frac{\text{Bq/(1 milk)}}{(\text{Bq/(kg feed)}) (\text{kg intake of feed/day})} \quad (2)$$

The DF has the units day l<sup>-1</sup>.

The activity of the radionuclide in the milk is assumed to be in equilibrium with concentrations in the feed. The activity in the feed and daily intake of feed must both be based on either dry or wet weight.

The distribution factors to meat and eggs are similar to the above definition however they must be based on the average activities in the animal parts or products used in man's diet. The units of these DF are day kg<sup>-1</sup>.

### 1.3 Fish

The CR for fish is defined as the activity of the radionuclide per unit fresh weight of the organism divided by the activity of the same radionuclide per unit volume of water:

$$\text{CR} = \frac{\text{Bq}/(\text{kg fish fresh weight})}{\text{Bq}/(\text{l water})} \quad (3)$$

The CR has the units l kg<sup>-1</sup>.

## 2. SOIL - PLANT CONCENTRATION RATIOS

A number of other expressions also exist in the literature to describe the concentration ratio as shown in equation (1), these include transfer factor (TF), transfer coefficient (TC) and concentration factor (CF) and appear freely interchangeable. The CR for a given radionuclide is an empirical relationship and is therefore not directly related to any of the many processes that effect the transfer of the radionuclide from soils to plants; it simply relates the concentration of accumulated activity in the soil at a particular time. However, these processes are included implicitly in estimates of the CR. Consequently it is difficult to predict the direction and the extent of changes in the CR if circumstances vary (e.g. different soils, plants and climate). In order to overcome this black-box approach Van Dorp et al, (75) suggested a new method for soil-plant transfer calculations in which the transfer is described as a function of plant and soil parameters all having physical meaning (he notes that the model is still an oversimplification of reality). Using this technique all the parameters can be experimentally determined, but a realistic estimation of them is also possible.

Unfortunately, the relative weakness of the existing literature soon becomes apparent as all the necessary parameters, such as root and shoot production, transpiration and adsorption properties of the soil, are rarely mentioned for any single experiment, thus making the implementation of this new approach somewhat difficult. The problem therefore remains to make the best of the existing data in the literature for concentration ratios. Table 2 shows the range of these for a number of radionuclides relevant to nuclear waste management. The values are taken from Ng et al, (49). The enormous variability is striking, with individual CR's commonly varying over two or more orders of magnitude.

Table 2: Range of CR reported in the literature for a number of radionuclides

<u>Element</u>	<u>CR Range</u>	<u>Mean</u>
Ni	7.0 E-3 - 1.5 E-1	3.3 E-2
Sr	1.0 E-3 - 1.7 E0	1.6 E-1
Cs	1.5 E-5 - 5.9 E-2	9.5 E-3
I	2.0 E-4 - 1.2 E-1	1.4 E-2
Np	2.8 E-2 - 1.3 E-1	7.9 E-2

These values have been obtained from radioisotope experiments on plants grown in pots or other containers in laboratories and greenhouses, or in containers or field plots outdoors. In the absence of radioisotope data, CR values are estimated from the concentrations of stable isotopes in plants and associated soil.

In order to determine the most applicable CR (or range of CR) for an assessment model a number of factors have to be considered. Firstly, the purpose for which the CR will be used e.g. continuous low-level release, accident situation etc., and secondly, the way in which the specific literature values have been determined e.g., water culture, pot experiments, field measurements, or observations.

## 2.1 Available Data and its Applications

The early data for concentrations of radionuclides accumulated in crops and soils were derived from studies following fallout from weapons testing. Of primary interest at that time were isotopes of strontium, caesium and iodine, because they contributed most to the radiation dose received by man. These investigations have several limitations; not all the transfer pathways were distinguished

i.e., root uptake, resuspension, interception of airborne radionuclides by foliage etc.; the emphasis was placed on regional rather than local dose assessments, so that data for soils from one location were combined with data for plants from another location. In these early investigations only a restricted number of radionuclides were considered (I-131, Pu-239, Sr-89/90, Cs-137) for a narrow range of crops, mainly grass and cereals (36). An initial difficulty found with plant uptake experiments was that because radioisotope detection techniques were not very sensitive (11), experiments had to be carried out with soil concentrations of nuclides that were often high enough to have toxic effects on the plants. This inevitably affected uptake apart from any other limitations.

The development of nuclear power programmes has directed the attention towards many additional radionuclides which may potentially be released into the environment, such as Mn-54, Co-60, Zn-65, Ru-106, Sb-124, Cs-137, Ce-144 and Th-229 to name a few. In addition the reprocessing of nuclear fuel and waste disposal have led to an interest in the transuranium elements, mainly Np, Pu, Am and Cm as well as in the fission products Tc-99 and I-129 (36). However, a survey of the literature reveals a wealth of information on the fallout radionuclides especially Sr and Cs but data on the remaining radionuclides are less abundant.

In addition to the limitations of the early work described in the previous paragraphs experimentally determined CR may be inappropriate simply because essential information describing the experimental conditions were not recorded. For instance, the climate, fertilizer application, time of radionuclide application, chemical form, depth of contaminated soil layer and time of harvest.

During the last decade many computer models have been developed to predict doses to man resulting from radioactive releases (either routine or accidental) and the shortage of relevant data on soil-to-plant CR under realistic field conditions for these has become apparent. This has resulted in various groups being set up in Europe to provide CR for european field conditions. In 1982, the International Union of Radioecologists (I.U.R.) instigated a programme to co-ordinate the research on soil-to-plant CR measurements (36).

It aimed to improve estimates of risks from the applications of nuclear technology in Europe and began by compiling the present available reliable data for european conditions in order to assess areas where more fundamental research is required and formulate guidelines for this. At present a large part of the data covers just a

small number of radionuclides e.g., Cs, Sr, Mn, Co, but the data are being continuously added to as results are produced and at the same time the range of radionuclides is being extended to those that are less clearly understood e.g. Np and Tc.

## 2.2 Greenhouse versus Field Studies

Field experiments with radionuclides tend to be less common than experiments in greenhouses using small pots or culture solutions. This is mainly a consequence of external contamination problems with field work and also the relative speed and ease at which greenhouse experiments can be conducted allowing control over variables such as light, temperature and humidity. Recent work however, has highlighted problems in extrapolating from laboratory to field conditions (68). Sartor et al, (57) noted that concentration ratios derived from pot experiments in greenhouses were higher than those from comparable studies conducted in large containers outdoors.

Steffens et al, (68) demonstrated that Co-60 uptake by lettuce grown in small pots (400g soil) in growth chambers was sixteen times higher than in a lysimeter experiment conducted outdoors with the same soil (sandy). The greater uptake by potted plants is attributed to the continued contact of the roots with contaminated soil throughout the growth period and better growing conditions in greenhouses where temperature, light, water and fertilizers are controlled. The increased CR observed for plants grown in culture solutions are explained on similar grounds. In 1968, Sartor et al. (58) compared Sr-85 CR's for tomato and wheat grown in large outdoor containers and field plots. The CR was sometimes greater for the crops grown in the field plots and sometimes greater for the crops grown in the containers. From these limited experiments he concluded large outdoor containers may simulate field conditions reasonably well.

A similar conclusion was arrived at by Grogan (30) who grew a range of crops in pots outdoors which allowed growth to full maturity. The CR's obtained were comparable with those from lysimeter experiments.

These conclusions do not invalidate greenhouse experiments, as they are very useful in determining relative orders of radionuclide uptake (51), (66) and their mobility and subsequent distribution within a plant (43), (33).



### 2.3 Distribution of Radionuclides in Plants

Many studies have shown that radionuclides are not uniformly distributed within a plant once they have entered the transpiration stream but tend to concentrate in certain organs. Cline and Hinds (13) found in a field experiment that pea leaves contained higher concentrations of Pu-238 than the stem and pod tissues, with the lowest levels in the seed (edible portion). Grogan (30) found a similar pattern of uptake, with Ru-106, Ce-144, Cs-137 and Tc-99 all occurring in the highest concentrations in the leaves and the lowest in the seeds.

Many workers have noted significantly lower CR for radionuclides to the grain of cereal compared to the straw (32), (69), (31). Schreckhise and Cline (59) found 30 to 50 times less plutonium, americium and curium in barley seeds than in the remainder of the above ground parts, and correspondingly 5 times less neptunium. Pea seeds contained 70 to 230 times less Pu, Am and Cm, and 30 times less Np than the other aerial tissues. Similarly Grogan (31) found that radionuclide concentrations in potato tubers (edible portion) were significantly less than in the aerial shoots. These results demonstrate the importance of using a CR relevant to the edible portion of a crop for dose assessment models, rather than to the more easily measurable non-edible plant part (e.g., pea leaves, stem).

All crops do not take up a given radionuclide to the same extent. Evans and Dekker (23) found that Cs-137 was taken up by certain categories of crop in the following order:

Vegetable > forage crops > cereal crops.

Bean plants (*Phaseolus vulgaris*) were shown by Szabo (72), (73) to accumulate both stable and radioactive caesium and strontium more than either wheat or maize. In recent work conducted by Stoutjesdijk et al (71) with outdoor lysimeters, Cs-137 uptake was greatest in spinach (*Spinacea oleracea*) followed by cabbage (*Brassica oleracea*), potatoes (*Solanum tuberosum*) and beans (*Phaseolus vulgaris*) all having similar Cs-137 CR's and barley (*Hordeum vulgare*) a cereal, having the lowest uptake of caesium. This again supports the evidence that vegetable crops have greater uptake of radionuclides than cereals.

The results from Grogan (30) for Cs-137 also indicated greatest uptake occurred into vegetables (lettuce, peas, potatoes) compared to the cereals. For this reason four broad categories of soil-to-plant CR are input into the BIOPATH code for each radionuclide considered. These are CR for:

- 1) pasture
- 2) cereals
- 3) leaf vegetables
- 4) root vegetables

The CR for pasture covers the movement of radionuclides in the foodchain via animals, to man whilst the remaining three CR describe the major food types consumed by man. Although further divisions may be desirable for example between leafy (cabbage, lettuce) and leguminous (peas, beans) vegetables because an essentially different plant organ is being consumed (leaf vs. seed) in practise this is perhaps an unnecessary sophistication in view of the limited data base at the present time. The category of root vegetables includes all subterranean plant parts that are consumed, this is a convenient classification and gives scope to model the impact of situations such as surficial contamination via intimately bound soil on the crop.

#### 2.4 Soil factors

Many workers have shown that soil-to-plant concentration ratios are strongly influenced by soil processes and parameters (50), (25), (4), (70) and that the exact relationships between these are complex. The major factor governing the availability of elements in soils to plants is their solubility, since in general plants absorb only dissolved elements (82). Soil properties, soils processes and the form in which the radionuclide arrives in the soil all interact to determine solubility.

Soils with large particles and a coarse texture (e.g. sand) are associated with higher CR and medium and fine textured soils (silts, clays) with lower CR. However, these relationships are not simple and depend on the specific radionuclide in question. Considerable research has been carried out on the behaviour of caesium in soils and it is generally considered to become fixed by inorganic colloids or clays in a non-exchangeable form (39) especially in soils containing 2:1 clays such as illite and vermiculite. In this fixation the Cs enters the crystalline lattice of the clay minerals and is irreversibly fixed between the aluminium and silicon layers. This process also occurs with the potassium ion, although it is held less tightly due to its slightly smaller ionic diameter (37).

Nishita et al, (52) demonstrated that Cs uptake by plants decreased with increasing concentrations of exchangeable potassium in the soil, however, other exchangeable cations also influence caesium uptake from soil (6), highlighting the complexity of the situation.

The chemistry of the transuranic radionuclides in soils is extremely complex and only partially understood (80) and of these much less data is available for Am, Cm and Np than for Pu. The complexity can be appreciated, as four different oxidation states of Pu (III-VI) can coexist in the environment and this state influences its behaviour (16). Transuranides are strongly absorbed by the finer soil particles and colloids, Pu being more strongly sorbed than Am, Cm and Np (16).

In addition to soil texture, clay mineral content, cation exchange capacity, other soil factors such as the chemical form and concentration of the radionuclide, organic matter content, soil moisture content and redox potential will all interact to determine the behaviour of a radionuclide in the soil. However, these topics can only be meaningfully discussed in terms of specific radionuclides, but the magnitude of their effect on soil-to-plant CR should be appreciated (82).

## 2.5 Climate

Considerable field work has been conducted in the USA on radionuclide uptake from soils by crops at a number of locations principally Los Alamos National Laboratory, Los Alamos, Savannah River Laboratory, Aiken and Pacific Northwest Laboratory, Richland. The first two are in arid or semi-arid environments often with very little vegetation cover resulting in ecological conditions that are quite different to Europe (Switzerland). The soils in these sites are predominantly coarse-textured sands, low in organic matter content and quite different to those encountered in Switzerland which tend to be normally moist and highly vegetated.

## 2.6 Improvements in Data

Uptake experiments for the transfer of radionuclides from soils to crops require a considerable input of time, cost and expertise, before meaningful realistic data are yielded. Considerable technical difficulties arise in deciding on the precise chemical form and state in which the radionuclide under consideration should be added to the

soil and the depth and manner in which it should be applied. To date most experiments have been limited to the form that is commercially available and literature on the long-term speciation of radionuclides in the soil system is decidedly sparse. This is an important consideration as it will effect the mobility in the soil and rate of transfer to the plants. Many research groups specify an equilibration period after soil contamination and prior to sowing (70), (71) in order to overcome this problem and this process appears to take between one and three months (36).

In view of the problems involved in setting up an experimental system to provide numbers for less studied radionuclides, it may be worthwhile to consider natural analogue systems. For example if an area was located having a natural vein of an element in the groundrock, which yielded locally high concentrations of the element (e.g., thorium, uranium) then the vegetation in this region, if selectively sampled, could provide CR's as well as being a useful check for existing data. In such a system problems of speciation are not encountered as presumably the system will have existed for a large number of years resulting in an equilibrium situation.

### 3. CONCENTRATION RATIOS IN THE LITERATURE

The following section considers each radionuclide, for the biosphere modelling, in alphabetical order. The available data are briefly reviewed and assessed for its applicability to the modelling of a waste repository. At all times, careful consideration was given to the origin and method by which the literature values had been obtained due to the inherent problems associated with the interpretation of CR, discussed in the previous section of this report. In some instances it has been more appropriate to discuss the CR in the form they are reported in the literature i.e., dry plant/dry soil. In these cases this has been clearly indicated in the text, the CR used for BIOPATH are converted to wet plant, using the appropriate values listed in table 1.

In selecting specific CR for use in BIOPATH emphasis has been placed on the more recent experimental work conducted under well defined field conditions. When no data could be found for a particular radionuclide the CR was determined on the basis of the behaviour of an analogous element. It is appreciated that certain processes, such as the preparation of food prior to consumption may affect the calculated CR, but as little data are available on this such considerations are not included.

A complete table listing all the CR recommended for each radionuclide and crop type is given in section 4 (table 3).

### 3.1 Radionuclide specific Data

#### 3.1.1 Actinium

In the absence of any relevant experimental data it is assumed that Ac behaves similarly to Am, whose dominant oxidation state is also +3. For Am, the CR reported in the literature for root uptake (i.e., no excessive resuspension, surface deposition) range from  $10^{-7}$  -  $10^{-4}$ . A value from the upper end of this range equivalent to ca.  $2 \times 10^{-3}$  (dry plant) was selected for use in BIOPATH.

#### 3.1.2 Americium

See transuranium elements.

#### 3.1.3 Caesium

A large amount of data are available on soil to plant CR for caesium isotopes but these are ranging over four orders of magnitude. It has not been possible to account for much of this variation with a single major plant or soil factor. The main soil factors influencing CR are clay mineral content, organic matter, moisture content, cation exchange capacity, pH and soluble and exchangeable potassium contents. Coughtrey et al, (15) reviewed the literature relating to Cs and observed that under most conditions the uptake of Cs from the soil is limited by soil supply rather than the potential or capacity of plants to take up Cs from soil. For this reason when the CR were assessed from the literature, to determine suitable CR for BIOPATH, many of the experiments made in greenhouses or laboratories were excluded, so that only values derived from outdoor field experiments were utilized. In addition to this, work before the early 1960's on this radionuclide, was also discounted. The two main sources of data were the I.U.R. (36) compilation of CR determined under European conditions and Ng's (49) review of CR (global). The CR reported range from  $6 \times 10^{-4}$  to  $7.3 \times 10^{-2}$ . CR values of  $2.0 \times 10^{-2}$ ,  $1.3 \times 10^{-2}$ ,  $1.3 \times 10^{-2}$  and  $8.0 \times 10^{-3}$  for pasture, cereals, leaf vegetables and root vegetables, respectively were selected.

#### 3.1.4 Calcium

Very few data for radioactive Ca exist in the literature, instead Ca is usually referred to in association with its effect on Sr uptake by plants. The uptake and metabolism of these two elements by plants appears to be very similar and interrelated. In the absence of any appropriate data for Ca, the Sr values were taken for use in BIOPATH assuming analogous behaviour. The range of Sr values is from  $1.6 \times 10^{-3}$  - 1.1 for all the crop types. The individual CR values  $5.0 \times 10^{-1}$ ,  $1.2 \times 10^{-1}$ ,  $1.5 \times 10^{-1}$ ,  $1.4 \times 10^{-1}$  were used in BIOPATH for pasture, cereals, leaf vegetables and root vegetables, respectively.

#### 3.1.5 Curium

See transuranium elements.

#### 3.1.6 Iodine

Physical and biological characteristics of I-129, frequently have to be based upon observations of other iodine isotopes, in the absence of sufficient experimental data. I-131 is used to model its short term behaviour (8 days half-life) whilst stable iodine is used to model it in the long term (47).

There are very few data relating to the chemistry and mobility of iodine in soil. Coughtrey et al, (15) concluded approximately 10% of an addition of soluble I-131 to soil remains available for plant uptake. It appears that only a small proportion of iodine in soil will be present in an available form in the soil solution for plant uptake. This agrees with culture solution experiments that demonstrate root uptake of iodine (especially iodide) occurs readily, but when field studies are carried out the iodine uptake is much reduced, implicating the soil and its related processes as the limiting factor.

Soil contamination studies indicate I-129 is transported down a soil profile at a very slow rate (62). Higher iodine concentrations in the soil (ca.  $5 \mu\text{g g}^{-1}$ ) are often associated with organic fractions of the soil and Brauer + Strebin (9) verified that I-129 accumulates in the top soil and litter layer.

The extent of root uptake depends on the chemical form of the iodine and greenhouse experiments have demonstrated that the iodide form is taken up more readily than the iodate (74), (79). However, because of the limited knowledge of I-129 behaviour and chemistry in soil systems it is difficult to extrapolate the laboratory measurements to the field system with any certainty.

Menzel (42) classified iodine as one of the elements that are not concentrated having CR in the range 0.1 to 10 (dry plant). Coughtrey et al, (15) when reviewing the literature, found no evidence for CR to ever exceed unity and indicated that CR in the range 0.01 - 0.8 (dry plant) were more representative. A CR value of 0.4 (dry plant) was suggested for vegetation in general in the absence of any data to the contrary. Soldat (64) calculated a CR of 0.02 (wet plant) based on the behaviour of I-127, this is equivalent to about 0.1 - 0.2 on a dry plant basis. In view of the more conservative nature of Coughtrey's value, this was selected for the two categories pasture and cereals, in BIOPATH. Somewhat more experimentally determined CR for leaf and root vegetables were reported in Ng's (49) literature review and the mean of these values was selected for each. Thus  $1.0 \times 10^{-1}$ ,  $3.6 \times 10^{-1}$ ,  $1.9 \times 10^{-2}$ ,  $5.6 \times 10^{-3}$  for pasture, cereals leaf and root vegetables, respectively are used in BIOPATH.

### 3.1.7 Lead

The uptake of Pb-210 by plants has not been studied extensively, by far the greatest amount of research has been conducted on the stable element in relation to its toxicity to plants as a heavy metal. Plants do not require Pb and at elevated soil concentrations it is toxic to plants. Pb uptake by plants is a function of its lead tolerance as well as the hydrogen ion concentration in the soil. Investigations (65), (38) have shown that Pb-210 accumulation occurs predominantly in the roots, with only a small amount being translocated to the shoots. There is also evidence for Pb-210 exclusion from roots, Kalin + Sharma (38) found relatively uniform CR irrespective of the soil concentration. It has been noted that Pb-210 uptake does not appear to be a function of its solubility in soils (65).

The values for BIOPATH were taken from Schüttelkopf + Kiefer (61) who had analysed a range of vegetation growing in the Black forest in Germany in the proximity of uranium mines (see Radium). The reported CR ranged from  $4.0 \times 10^{-3}$  -  $2.6 \times 10^{-2}$ . They thought that somewhat higher CR for Pb-210 could be attributed to surficial

deposition of resuspended Pb-210. It was not considered necessary to reduce these values for BIOPATH since for the long time-scales involved, this process (resuspension) may also be of significance. Therefore for BIOPATH a value equivalent to  $1.8 \times 10^{-2}$  dry weight was taken for each crop type.

### 3.1.8 Molybdenum

Mo in soils is generally adsorbed on iron oxides and clay minerals. It appears that less than 10% of the Mo is available in the soil solution for plant uptake, either directly or via soluble organic complexes (15).

Mo is considered an essential element for plants, it is included in the catalytic group of elements and is incorporated in several metalloflavin enzymes, including those concerned with nitrogen fixation and the reduction of nitrate (19). It can be assumed that plants "scavenge" any available molybdenum from the soil and absorb it readily in an ionic form. Under normal conditions the supply of Mo from the soil limits the extent of plant uptake. The availability of Mo in soil is affected by soil pH and deficiency is usually associated with acidic soils. For these reasons, a wide range in CR are reported for Mo. For example, Cataldo + Wildung's data (10) for Mo uptake by Glycine max grown under normal conditions suggest CR in the range  $2,5 \times 10^{-2}$  -  $2,5 \times 10^{-1}$ , however, when Glycine max was grown in an amended soil a CR ca. 5 was measured.

From this it is clear that the soil-plant relationships are complex and a single CR cannot truly be used to represent most situations.

For use in BIOPATH the mean CR value compiled by Ng (49) from experimental data was selected. This value, which is equivalent to CR 4.5 (dry plant) was used for all four groups in the absence of sufficient data to indicate differences between the groups. This value overestimates the uptake of Mo under normal conditions since many of the experiments were carried out in laboratories where Mo was frequently applied to the soil in large amounts or unusual forms. However, because it is readily taken up by plants and considerable uncertainties are involved to predict the chemical form in which it will arrive to the biosphere, this more conservative value was chosen.



### 3.1.9 Neptunium

See transuranium elements.

### 3.1.10 Nickel

The data in the literature regarding the behaviour of Ni in the soil and its uptake by plants are limited and are largely based on studies of the stable element. There have been a number of experiments examining the uptake of Ni-63 by plants, but these were performed in culture solution (15).

From the few data available, it would appear that most Ni becomes firmly bound in soils, being adsorbed to or occluded by the mineral lattice. As a result ca. 3% (17) of the soil Ni is in an available form for plant uptake, this value increases to ca. 5-10% under anaerobic or waterlogged conditions due to its association with the soil microflora.

The CR for plant uptake appears to be low (ca. 0.1 dry weight) with the uptake being limited by its availability in soil. In the absence of any better data CR were taken from stable element calculations made by Furr et al (26) and Andersson + Nilsson (3). These CR ranged from  $7.8 \times 10^{-3}$  to  $1.5 \times 10^{-1}$  for cereals,  $1.1 \times 10^{-3}$  to  $2.3 \times 10^{-2}$  for leaf vegetables and  $7.2 \times 10^{-3}$  to  $4.0 \times 10^{-2}$  for root vegetables. The mean values for each group were taken and the CR for pasture ( $5 \times 10^{-2}$ ) was assumed to be similar to that measured for leaf vegetables, since no direct experimental data were available.

### 3.1.11 Palladium

No experimental data could be found concerning the behaviour of Pd in soils and its uptake by crops. The same CR which were selected for Ni were taken assuming analogous behaviour of these two isotopes. Pt is considered to be a better analogue for Pd, (44), however, no data were available for this element either.

### 3.1.12 Plutonium

See transuranium elements.

### 3.1.13 Protactinium

In the absence of relevant data Pa is assumed to be chemically similar to Np (14), so that the same CR were used for both elements. The range of CR for Np found in the literature is  $5 \times 10^{-5}$  to  $1.3 \times 10^{-1}$  and CR values of  $9.4 \times 10^{-3}$ ,  $1.7 \times 10^{-2}$ ,  $2.7 \times 10^{-2}$ ,  $6.0 \times 10^{-2}$  for pasture, cereals, leaf vegetables and root vegetables, respectively, were taken for calculations with BIOPATH.

### 3.1.14 Radium

The literature relating to Ra-226 uptake by crops is rather limited, although in recent years an increasing number of field and laboratory studies have been made on this element (61), (67), (24). Schüttelkopf + Kiefer (61) have measured Ra CR for vegetation in the Black forest in Germany around two uranium prospecting mines and an uranium waste deposit that is several centuries old, left over from mining Co and Ag. They recorded CR in the range  $1 \times 10^{-3}$  -  $5 \times 10^{-2}$  (fresh plant) for vegetables, and  $2.8 \times 10^{-3}$  -  $2.7 \times 10^{-2}$  (fresh plant) for pasture. These values agree reasonably well with those determined for vegetables grown in the vicinity of Morro do Ferro in Brazil. This is a highly weathered remains of an igneous intrusion occurring ca. 70 million years ago and results in increased Ra as well as Th, U and rare earth element concentrations. Here CR for vegetables ranged from  $4 \times 10^{-4}$  -  $1.9 \times 10^{-2}$  (fresh plant) with a mean value of  $8 \times 10^{-3}$ .

Eriksson + Fredriksson (24) investigated a total of 37 different sites throughout Sweden examining Ra, U and Th concentrations in soils and vegetation. From 40 crop samples an average Ra-226 CR of  $1.6 \times 10^{-2}$  (dry plant) was calculated. For cereal the CR ranged from  $1.09 \times 10^{-2}$  -  $2.02 \times 10^{-1}$  (dry plant) giving a mean value of  $1.5 \times 10^{-2}$ . The effect of soil type on the Ra-226 content and availability was also examined, only a small proportion of Ra-226 present in soil was shown to be available for plant uptake. Ra-226 appears to become associated with the organic matter in the soil and also with clay minerals but to a lesser extent. In addition pH affects Ra-226 availability to plants, with increasing availability as the pH decreases.

Adriano et al, (2) grew rice plants under greenhouse conditions, in a soil contaminated with Ra-226 and the surface of which was flooded (ca. 3cm standing water). CR of 1.40 (dry plant) and 0.99 (dry plant) were measured for the foliage and grain, respectively. These values are higher than those reported by other workers, however this may reflect the high level of soil contamination and/or the anaerobic conditions. Enhanced radionuclide uptake under flooded conditions has been widely observed (67), (1).

The mean CR determined by Eriksson and Fredriksson (24), corrected for the appropriate water content of each crop category, was taken for BIOPATH calculations. The CR values for pasture, cereals, leaf and root vegetables used are,  $4.0 \times 10^{-3}$ ,  $1.4 \times 10^{-2}$ ,  $1.6 \times 10^{-3}$  and  $3.0 \times 10^{-3}$ , respectively. It was not considered necessary to adopt the values of Adriano et al, (2) because of their unrealistic nature with respect to typical agricultural conditions in Switzerland.

### 3.1.15 Rubidium

Studies of Rb behaviour in soil and its uptake by plants are extremely limited. At one time it was assumed Rb uptake could be represented on the basis of K uptake from soil, however this has been demonstrated to be false, K uptake occurring very much more rapidly. A few experiments have been conducted determining the behaviour of Rb-86 in soils (54), (29). Here it is bound on exchange sites of clay minerals (c.f. Cs) and shows relatively low diffusion through the soil. As a consequence, the extent and rate of Rb uptake is not limited by the ability of the plant roots to accumulate Rb but by its low diffusion rate in the soil. Coughtrey et al (15) concluded a CR of 0.3 (dry plant) was representative for Rb, from the limited data they had compiled. In the absence of any more relevant data this value was adopted for BIOPATH, correcting for the water content of the four crop categories.

### 3.1.16 Selenium

A wealth of data exists referring to the behaviour of stable Se in plant and soil systems. This interest results from toxicity symptoms found in animals grazing seleniferous soils, whereas deficiency symptoms were recorded on selenium deficient soils. Subsequently Se was considered to be an essential element to animals.

The chemistry of Se in soils is complex, due to the various oxidation states in which Se can exist in soil. However, Selenite and selenate are the two more common forms in which Se occurs in soils (15). In acid soils Se is often present as insoluble ferric selenite, whereas in alkaline soils selenite is oxidized to soluble selenate, a process which is enhanced in the presence of organic matter (15). The availability of an addition of radioactive Se in soil will largely depend on its chemical form, in conjunction with the soil pH, organic matter content, iron content, native Se etc. Se added in the selenite form is likely to be less available than the selenate because ca. 90% becomes fixed by iron complexes and clays. Microbial activity has also been shown to alter the chemical form of Se (15), however there are insufficient data to predict the extent of these changes or the organic forms that will occur, however it is not unlikely that volatile Se compounds may be produced (e.g. dimethyl selenides).

Soil-to-plant CR, covering four orders of magnitude have been calculated for stable Se. This large range results from the diverse experimental conditions for which they have been calculated. For example, measurements under normal conditions (neither deficiency or toxicity) yield CR in the range  $5 \times 10^{-3}$  -  $2.5 \times 10^{-2}$ , but for seleniferous soils ( $\geq 3 \mu\text{g g}^{-1}$  dry soil)  $\text{CR} > 1 \times 10^{-1}$  appear more common. Very high CR have been determined for Se amendments to soils, especially for Se deficient soils (1-10). One of the major problems is assessing how appropriate these values are to describe the behaviour of radioactive Se added to a normal soil. Switzerland has not been shown to have Se deficient soils and Gissel-Nielsen (28) showed the vegetation contained adequate Se concentrations. In view of this, it was not considered appropriate to adopt the very high CR that have been measured for Se amendments to Se deficient soils. The values for BIOPATH were taken from Coughtrey et al (15) for crops grown on normal soils. These values are,  $3.0 \times 10^{-1}$ ,  $4.3 \times 10^{-2}$ ,  $4.5 \times 10^{-2}$  (dry plant) for pasture, cereals, leaf and root vegetables, respectively. The CR for cereal is a mean value taken from Lindberg + Bingefors (40) who analysed cereals and soils throughout Sweden (range  $5 \times 10^{-3}$  -  $1 \times 10^{-1}$ , mean  $4.3 \times 10^{-2}$ ).

### 3.1.17 Strontium

The literature relating to Sr behaviour in soils and its uptake by plants is extensive and largely results from its importance as a fallout radionuclide. Coughtrey et al, (15) recently reviewed the Sr literature for plant CR

and noted a considerable variation both between species and within the same group of vegetation types on different kinds of soil or at different ages. The CR for radioactive Sr, although exhibiting a similar range of variation, were consistently larger than those for the stable element. In addition to this, pot experiments produced greater CR than those reported for field studies, this is considered to be due to depletion of calcium reserves in pot experiments. One reason for the difference in CR between stable and radioactive Sr is their differing behaviour in soil. In general, only a small fraction (2-10%) of stable Sr in soils is available for plant uptake, by contrast when soluble  $^{90}\text{Sr}$  compounds are added to soil, a large proportion (80-90%) remains available for plant uptake. Only a small proportion becomes strongly absorbed and unavailable, even over considerable periods of time (15). The extent to which these processes occur depends upon the soil pH and soil organic matter content, since Sr is held mostly in exchange sites. This factor also limits the vertical migration of Sr in soils.

The Sr content of vegetation is closely related to the Sr and Ca content of the soil (15). The relationship between these two elements has been noted by many workers, so that at one time plant concentrations were predicted by reference to observed ratios (OR). However these relationships are complex depending on the soluble, exchangeable, and total fractions of each element and the chemical form of the radioactive Sr as well as soil characteristics.

The CR values for BIOPATH are taken from Ng's (49) compilation of experimental data. However, all greenhouse experiments were excluded, since they had been carried out in small pots where Ca depletion was likely to have occurred. In addition to this results from the very early work were also excluded because not all the transfer pathways to the crops were clearly distinguished. The mean CR for each crop type was taken for the BIOPATH calculations. The values for pasture ranged from  $6 \times 10^{-2}$  - 1.4 with a mean CR of  $5.8 \times 10^{-1}$ , for cereals the CR ranged from  $9 \times 10^{-3}$  -  $2.8 \times 10^{-1}$  with a mean CR of  $1.2 \times 10^{-1}$ . The CR for leaf and root vegetables ranged from  $1.6 \times 10^{-3}$  - 1.05, and  $8.4 \times 10^{-3}$  -  $4.6 \times 10^{-1}$ , respectively with mean values of  $1.5 \times 10^{-1}$  and  $1.4 \times 10^{-1}$ .

### 3.1.18 Technetium

In recent years a considerable number of plant uptake experiments have been made with Tc-99. This results from the extremely high CR reported by various groups (21), (27), (35) in the literature. When Tc-99 is applied to the soil as the pertechnetate ion it is rapidly taken up by

crops, resulting in very large CR, e.g. 1.4 - 153 (dry plant) (21), (35). Pot and culture solution experiments yield even greater CR e.g. 95-1890 (dry plant) (27). The uptake of Tc-99 by plants is only limited by its availability and concentration in the soil. Many of the above CR result from short term experiments in which unrealistically high concentrations of Tc-99 were applied as the pertechnetate ion. The pertechnetate anion ( $\text{TcO}_4^-$ ) is highly soluble in water and poorly sorbed to soil thus making it extremely mobile. This is considered the most stable chemical species under aerobic conditions in aqueous solution and is the form in which it appears following treatment of wastes (45).

Eriksson (21) conducted a five year lysimeter experiment in an open field on the transfer of Tc-99 from eight different soils to crops of red clover and spring wheat. The Tc-99 was mixed into the top soil in the pertechnetate form. The initial plant uptake was considerable, with the average CR being 132 (dry plant) for red clover in the first year. The CR then decreased rapidly with time so that after 5 years the crop uptake was reduced by a factor of 2000. The plant availability of Tc-99 was very high at the commence of the experiment but decreased with time, due to plant uptake, chemical reduction of the pertechnetate to less soluble forms and movement to deeper horizons. The main feature coming out of this work is the importance of the chemical form of the released radioactivity and the time-scale involved. For accidental releases where a single deposition occurs, transfer of Tc to the vegetation is considerable. Under continuous releases of small amounts of Tc-99 e.g. as a result of operating a nuclear installation, Eriksson considered that transfer to food would be of the same order of magnitude as for Cs.

In the context of a high level waste repository it is unlikely that a sudden very large release of Tc-99 will occur, to the biosphere, but will be more closely described as a chronic release, due to the long time-scales and distance involved, during which dispersion, dilution and retardation affects will occur in the geosphere. As a result CR equivalent to 10 (dry plant) were used for the crop categories of pasture, leaf and root vegetables and a value of 5 (dry plant) for cereals. These values are still very much higher than those suggested by Eriksson and therefore conservative.

It should be noted that when the lower CR value is being used a higher Kd value should be used for sorption on the soil. For situations where the Kd is very low (effectively no sorption) a high CR is required and depletion of the soil by plant uptake should not be neglected.

A slightly lower value was given to cereals because significantly lower CR have been recorded for the grain than the straw (31), (21) as Tc-99 does not appear to be re-translocated once it has been taken up in the xylem. Cross-contamination during harvesting and processing from the straw is improbable as the Tc-99 is not associated with the soil and surficial contamination.

### 3.1.19 Thorium

There are very few experimental determinations for the uptake of Th by crops reported in the literature. Somewhat more data exist for Th behaviour in soils. Eriksson + Fredriksson (24) showed that the Th in soil is highly correlated with the soil clay content and they could only determine a weak relationship between the radionuclide and the organic matter content. In contrast, Wahlgren + Orlandini (76) found that Th is closely related to the organic fraction. Despite this confusion it is agreed that the fraction of the Th in the soil which is available to the crops is small. Its availability can be ranked accordingly;  $Ra > U > Th$ , Ra being the more readily taken up. A mean CR of  $3.8 \times 10^{-3}$  (dry plant) was determined by Erkiesson + Fredriksson (24) for 37 different crops, sampled throughout Sweden. This value has been used for BIOPATH in the absence of any more relevant data. It was also noted that the uptake into grain was significantly less, so the mean of these values was taken for the cereal group of foods (range  $7.4 \times 10^{-5}$  -  $4.4 \times 10^{-2}$ , mean CR  $8.0 \times 10^{-4}$ , all expressed in dry weight).

### 3.1.20 Tin

Sn is strongly absorbed onto the humus (organic) fraction of soil (8) so that only a small proportion will remain available for uptake by plants. Microbial activity can result in chemical changes of Sn, however, the importance and extent to which this occurs has not yet been elucidated.

Tin is known to be toxic to plants at relatively low concentrations and it is in this area that most work has been carried out. Despite this a number of laboratory experiments on plant/soil relationships have been made. Romney et al, (55) found the concentration of Sn in plants was effectively independent of the soil contamination level. Myttenaere et al, (46) obtained CR in the range  $3.11 \times 10^{-1}$  -  $6.38 \times 10^{-1}$  (dry plant) for Sn-113 uptake

by rice, under laboratory conditions. When there is excess Sn in the soil substantial amounts may be absorbed, but remain localised in the roots showing limited translocation (15). In the absence of any more relevant data for CR a value equivalent to 0.4 (dry plant) was adopted for all crop categories. This is slightly larger than the 0.25 (dry plant) anticipated by Coughtrey et al, (15) for stable Sn.

### 3.1.21 Transuranium Elements

The uptake of the transuranics by plants has been studied under both field and laboratory conditions to a limited extent. These studies have focused upon Pu, Am, Np and Cm, since they have been identified as the most important environmental contaminants in the transuranic series to date and result from fallout and discharges from nuclear facilities. Of these radionuclides, plutonium has received the most attention as it has been released in the greatest quantities. Although the behaviour of these four radionuclides in the environment is similar, experimental work has shown that all transuranium elements do not behave identically to plutonium and are not equally absorbed by plants, as was once assumed. However, it is established that root uptake by plants is generally low (less than  $5 \times 10^{-3}$ ) although Np is something of an exception showing greater mobility in these systems. One important consequence of this is that in situations where the transuranics are released as an aerosol or occur in a layer on the soil surface, contamination of plants on their exposed surfaces (via deposition or resuspension) is higher by upto three orders of magnitude than the contamination resulting from root uptake.

Watters et al, (78) noted that the physiological availability of a transuranic can be increased or decreased by at least an order of magnitude by soil amendments and indigenous soil factors. For example, chelating agents generally increase Pu and Am availability to plants, whereas liming treatment decreases the availability of Am to plants. At the present time there are insufficient data to accurately predict their behaviour in terms of availability to plants. This is, in part, the reason for the large range of CR reported in the literature which makes it impossible to rigidly define a CR for a given crop type.



### Americium

Concentration ratios for Am reported in the literature range from  $10^{-7}$  to  $10^{-1}$ . The highest measured CR values ( $10^{-3}$ - $10^{-1}$ ) result from field studies of native plant species growing in areas with high ground surface contamination or in areas with active deposition. For example Romney et al (56) studied the Am content of wild plants growing in the Nevada Test Site. In such cases a significant proportion of the Am has been shown to be present on the plant surface as a result of resuspension or deposition. In laboratory experiments where these two processes are precluded and Am reaches the plant via root uptake alone, CR's in the range  $10^{-7}$  -  $10^{-4}$  are recorded. The specific CR's vary with crop type and it has been observed that grain and seeds consistently contain 10-100 times less Am than the remainder of the plant e.g. leaves, straw, (22), (53), (59). As there are insufficient data to award different CR values for the main crop groups a value corresponding to  $2 \times 10^{-3}$  dry plant/dry soil was selected for use in BIOPATH. This lies at the upper end of the laboratory determined values and although it possibly overestimates the importance of root uptake it allows the impact of resuspension (occurring to a limited extent) to be included in this value. The value for cereals is lower ( $2.2 \times 10^{-5}$ ) since uptake into the grain is less than into the vegetative parts.

### Curium

There are only a few data available in the literature for Cm-244. Schreckhise and Cline (59) conducted a laboratory experiment on the comparative uptake of Pu, Am, Cm and Np, in which no significant difference in uptake between Am-241 and Cm-244 was found. This trend is also confirmed by the preliminary results of Pimpl and Schmidt's (53) uptake experiments conducted under greenhouse conditions. For the purposes of the BIOPATH code a value identical to that adopted for Am is recommended and although it is likely that the CR for grain is less than this, in the absence of conclusive data a value equivalent to  $2 \times 10^{-3}$  (dry plant) is recommended for pasture, leaf and root vegetables. Few data exist for cereal which range from  $1.8 \times 10^{-4}$  -  $2.0 \times 10^{-3}$  and the mean value  $1.1 \times 10^{-3}$  was used for BIOPATH.

### Neptunium

The uptake of Np by plants occurs to a considerably greater extent than the other transuranics studied (59) and the literature values range between  $5 \times 10^{-5}$  and  $1.3 \times 10^{-1}$  (49), (22). Eriksson (22) has measured CR for

spring wheat (grain) and clover shown in outdoor lysimeters. The values ranged from  $4.8 \times 10^{-5}$  -  $3.0 \times 10^{-2}$  and  $1.1 \times 10^{-3}$  -  $3.8 \times 10^{-2}$ , respectively. Schreckhise and Cline's comparative uptake study (59) resulted in a CR of 0.4 (dry plant) for Np. In the absence of any relevant data for root vegetables this value was adopted for root vegetables, whilst the arithmetic mean of the literature values for each of the other groups was selected. The CR for pasture and cereals ranged from  $1 \times 10^{-3}$  -  $2.7 \times 10^{-2}$ , mean CR  $9.4 \times 10^{-3}$ , and  $5 \times 10^{-5}$  -  $1.3 \times 10^{-1}$ , mean  $1.7 \times 10^{-2}$ , respectively. The range of values for leaf vegetables was from  $1.3 \times 10^{-2}$  -  $4 \times 10^{-2}$  with a mean CR  $2.7 \times 10^{-2}$ .

### Plutonium

A considerable wealth of data exists for Pu CR with reported values ranging from less than  $10^{-7}$  up to  $10^{-1}$ . Pu CR's based on greenhouse studies are generally several orders of magnitude lower than those derived from field data. The higher CR's based on field data are due to the additional contamination resulting from resuspension or deposition as discussed previously. Cawse (12) has measured CR for grass (hay) and two cereal crops (oats, barley) at eleven sites throughout England and the values ranged from  $3 \times 10^{-3}$  to  $2 \times 10^{-2}$ , and  $1 \times 10^{-2}$  to  $2 \times 10^{-1}$ , respectively. These values comprise the upper end of reported CR for Pu and reflect the contribution of direct deposition to the total contamination of the crop. Similarly CR ranging between  $10^{-3}$  and  $10^{-1}$  were determined for wild plants growing at the Nevada Test Site (56), in this case resuspension significantly contributes to the Pu level of the plants.

Eriksson (22) and Pimpl + Schmidt (53) have separately determined CR in the range  $10^{-7}$  -  $10^{-5}$  for cereal grain, whilst Wallace (77) indicates a value of not more than  $10^{-7}$ . For clover (pasture) Eriksson determined CR in the range  $10^{-5}$  -  $10^{-3}$ , with most values being closer to  $10^{-5}$ . Schreckhise + Cline (59) estimated a CR of  $1 \times 10^{-5}$  for Pu in their comparative uptake experiment. A general value of  $10^{-4}$  for plant uptake of Pu by vegetables and grass has been suggested by several reviewers (5), (18), (34).

CR values for BIOPATH were selected from the upper end of the range of laboratory, greenhouse and field plot experiments noted. In these cases any significant contribution via deposition or resuspension was not apparent. However, by taking the upper end of the reported values the importance of root uptake is probably overestimated resulting in a reasonably conservative value. The following values were used in the calculations,  $3.8 \times 10^{-4}$  for pasture,  $1.8 \times 10^{-3}$  for cereals,  $1.4 \times 10^{-4}$  for leaf vegetables and  $3 \times 10^{-4}$  for root vegetables.

### 3.1.22 Uranium

U is another element which has not received too much attention with respect to uptake by plants. The only relevant experimental data found, were those of Eriksson and Fredriksson (24). Some CR have also been determined by Sheppard + Thibault (63) for species indigenous to the precambrian shield of Canada. However, these species are not typical crops comprising a large number of lichens and mosses, which behave quite differently in their mode of radionuclide uptake compared with higher plants (e.g. food crops). The results are also expressed on an ashed plant/ashed soil basis, which are difficult to transform into fresh plant/dry soil values for comparison. Consequently these were not considered for use in BIOPATH.

Eriksson + Fredriksson (24) showed the U in soil is associated with the clay minerals and organic matter and the transport of U down the soil profile is limited. The plant uptake of U appeared to be related to the soil pH and calcium content and the results suggested U uptake by plants occurs from a fraction bound as carbonate or silicate complexes. The experimentally determined CR for 37 different crops sampled throughout Sweden had a mean value of  $3.8 \times 10^{-3}$  (dry plant) with a slightly lower value for cereal grain,  $1.4 \times 10^{-3}$  (dry plant). The mean values were taken for input into the BIOPATH model (corrected for crop water content).

### 3.1.23 Zirconium

Zr is strongly fixed by soils (41) leaving only a small fraction (ca. 5-15%) available for plant uptake. Schultz (60) considered that Zr is strongly adsorbed to clay particles or is precipitated as insoluble oxides or hydroxides. According to Coughtrey et al (15) its availability can be expected to increase in soils with high microbial activity, since the organic acids that microorganisms may produce increase its solubility.

The experimental data used to select CR for Biopath are taken from the review by Ng et al (49). The CR range from  $1.1 \times 10^{-2}$  -  $2.9 \times 10^{-2}$  with a mean CR  $2.0 \times 10^{-2}$  for pasture,  $3.4 \times 10^{-6}$  -  $1.8 \times 10^{-2}$  with a mean CR  $3.4 \times 10^{-3}$  for leaf vegetables, and range from  $9.9 \times 10^{-4}$  -  $4.3 \times 10^{-3}$  with a mean of  $2.1 \times 10^{-3}$  for root vegetables. No data were found for grain so the value  $2.7 \times 10^{-2}$  was selected by direct extrapolation from the other results.

4. SELECTED SOIL-PLANT CONCENTRATION RATIOS

From the data compiled in section 3, a selection was made for use in biosphere modelling with the computer code BIOPATH. The data were selected by choosing in general the higher side of the reported range, taking into account the climate, soil and crop conditions for Switzerland. The selected data are reported as fresh plant weight to dry soil weight ratios. For conversion from dry plant to fresh plant, the moisture content given in table 1 was used.

Table 3: Data base of CR for use in BIOPATH

<u>Element</u>	<u>Pasture</u>	<u>Cereals</u>	<u>Leaf Vegetables</u>	<u>Root Vegetables</u>
Ca	5.0x10 <sup>-1</sup>	1.2x10 <sup>-1</sup>	1.5x10 <sup>-1</sup>	1.4x10 <sup>-1</sup>
Ni	5.0x10 <sup>-2</sup>	4.2x10 <sup>-2</sup>	1.7x10 <sup>-2</sup>	1.6x10 <sup>-2</sup>
Se	2.5x10 <sup>-1</sup>	3.6x10 <sup>-2</sup>	3.5x10 <sup>-2</sup>	3.8x10 <sup>-2</sup>
Rb	7.5x10 <sup>-2</sup>	2.7x10 <sup>-1</sup>	3.0x10 <sup>-2</sup>	4.5x10 <sup>-2</sup>
Sr	5.8x10 <sup>-1</sup>	1.2x10 <sup>-1</sup>	1.5x10 <sup>-1</sup>	1.4x10 <sup>-1</sup>
Zr	2.0x10 <sup>-2</sup>	2.7x10 <sup>-2</sup>	3.4x10 <sup>-3</sup>	2.1x10 <sup>-3</sup>
Mo	1.1x10 <sup>0</sup>	4.0x10 <sup>0</sup>	5.0x10 <sup>-1</sup>	7.0x10 <sup>-1</sup>
Tc	2.5x10 <sup>0</sup>	4.5x10 <sup>0</sup>	1.0x10 <sup>0</sup>	1.5x10 <sup>0</sup>
Pd	5.0x10 <sup>-2</sup>	4.2x10 <sup>-2</sup>	1.7x10 <sup>-2</sup>	1.6x10 <sup>-2</sup>
Sn	1.0x10 <sup>-1</sup>	3.6x10 <sup>-1</sup>	4.0x10 <sup>-2</sup>	6.0x10 <sup>-2</sup>
I	1.0x10 <sup>-1</sup>	3.6x10 <sup>-1</sup>	1.9x10 <sup>-2</sup>	5.6x10 <sup>-3</sup>
Cs	2.0x10 <sup>-2</sup>	1.3x10 <sup>-2</sup>	1.3x10 <sup>-2</sup>	8.0x10 <sup>-3</sup>
Pb	4.5x10 <sup>-3</sup>	1.7x10 <sup>-2</sup>	1.8x10 <sup>-3</sup>	2.7x10 <sup>-3</sup>
Ra	4.0x10 <sup>-3</sup>	1.4x10 <sup>-2</sup>	1.6x10 <sup>-3</sup>	3.0x10 <sup>-3</sup>
Ac	5.0x10 <sup>-4</sup>	1.8x10 <sup>-4</sup>	2.0x10 <sup>-4</sup>	3.0x10 <sup>-4</sup>
Th	9.5x10 <sup>-4</sup>	7.1x10 <sup>-4</sup>	3.8x10 <sup>-4</sup>	5.7x10 <sup>-4</sup>
Pa	9.4x10 <sup>-3</sup>	1.7x10 <sup>-2</sup>	2.7x10 <sup>-2</sup>	6.0x10 <sup>-2</sup>
U	9.5x10 <sup>-4</sup>	1.3x10 <sup>-3</sup>	3.8x10 <sup>-4</sup>	5.7x10 <sup>-4</sup>
Np	9.4x10 <sup>-3</sup>	1.7x10 <sup>-2</sup>	2.7x10 <sup>-2</sup>	6.0x10 <sup>-2</sup>
Pu	3.8x10 <sup>-4</sup>	1.8x10 <sup>-3</sup>	1.4x10 <sup>-4</sup>	3.0x10 <sup>-4</sup>
Am	5.0x10 <sup>-4</sup>	2.2x10 <sup>-5</sup>	2.0x10 <sup>-4</sup>	3.0x10 <sup>-4</sup>
Cm	5.0x10 <sup>-4</sup>	1.1x10 <sup>-3</sup>	2.0x10 <sup>-4</sup>	3.0x10 <sup>-4</sup>

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