ECORAD 2004 – IRSN Collection Series, Suppl. 1, vol. **39** (2004)

DOI : 10.1051/jp4 :20030178 (will be inserted by the publisher)

Arguments for revising radioecological transfer factors : how to improve and extend current syntheses

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Abstract. The Handbook of parameter values for the prediction of radionuclide transfer in temperate environments (TRS 364) was published in 1994 by the IAEA, in collaboration with the IUR. It was based on a review of available data up to the end of 1992. It is composed of values for empirical transfer parameters commonly used in radiological assessment models. Certain values are based on data which are 20 years old. The availability of literature data inevitably improves with time. The amount of data, however, will vary for each element since data have been compiled due to a variety of reasons, such as Chernobyl, waste disposal or dose reconstruction. A number of good critical reviews have been produced in recent years for some of the transfer parameter values which merit consideration. When addressing transfers through continental ecosystems, TRS 364 is one of the key sources for many models : it is widely used both in the radiation protection and radioecological communities. In particular, many radiation protection models need to predict transfer of a large number of radionuclides. This requires information on transfer of many less mobile radionuclides, which do not usually comprise an important component of discharges or dose. Such information is often sparse and difficult to collate. It is thus essential that such information is kept up-to-date and that any relevant recent literature is included, especially considering the paucity of existing data sources. Moreover, since the early nineteen nineties, there has been considerable debate in the scientific community regarding the validity of using empirical approaches for determining transfer factors. For instance, the identification of single values, when considerable variation is observed and can be explained to varying extents, seems a simplification leading to avoidable errors in predictions. This, in itself, is a strong argument for revision if the information given which is now known to be incorrect, inadequate (given new available information) and incomplete. This effort is a task of the IAEA-EMRAS programme on environmental modelling.

1. INTRODUCTION

Many safety, performance, or radiological impact assessments, concerning either actual or potential releases, need biosphere parameters such as transfer or accumulation factors to quantify various environmental processes, for which they needs relevant, reliable quantification. One international example is the IAEA SRS No. 19 (2001) [1] on the assessment of routine releases. If future assessment requirements are considered, such as those linked to overall environmental impact assessments (Hunter, 2001 [2]), most of the current biosphere models will still have a need for updated and reviwed compilations of relevant parameter and values.

In the literature, as stated by Whicker et al. (1999) [3], comprehensive syntheses of available parameter values (such as the well known Balkema series of Coughtrey, Jackson, Jones and Thorne, 1983-1985 [4]). are not very numerous. One important document is widely used by the scientific community, namely the IAEA Technical Reports Series No. 364, "Handbook of parameter values for the prediction of radionuclide transfer in temperate environments", published in collaboration with the International Union of Radioecologists (IUR) in 1994 [5]. Its contents reflected radioecological data up to 1992. TRS 364 is widely used as a major source of information, because it addresses numerous environmental transfer parameters and radionuclides. It is therefore quoted in numerous impact assessments, even if amended or supplemented by the scientific community (radiation protection, radioecology). Moreover, many radiation protection models need to predict transfer of a large number of radionuclides. This requires information on transfer of many less mobile radionuclides, which do not usually comprise an important component of discharges or dose. Such information is often sparse and difficult to collate. TRS 364 provides an important source of such information, and is one of the key cited sources for many models. It is thus essential that such information is kept up-to-date and that any relevant recent literature is included, especially considering the paucity of existing compendia of data sources.

Since 1992, new data have been produced, such as post-Chernobyl information (Shaw, 2001 [6]), and new experimental results (e.g. lysimeter studies), potentially providing significant additional information to supplement existing data and syntheses, which are now more than 10 years old. A number of high quality critical reviews have been produced in recent years for some of the transfer parameter values which merit consideration. International programmes have also been devoted to the construction and validation of radioecological models : BIOMOVS, IAEA/VAMP, IAEA/BIOMASS, European Frameworks, etc. It is now thought by many of the radioecological and radiation protection community that there is sufficient new information available to warrant reconsideration of a significant proportion of the values given in TRS 364.

As a consequence, there are arguments for updating TRS 364 and extending it, in terms of compartments, processes and radionuclides, and even for improving its current contents, because a critical analysis can highlight some weaknesses. This explains why the IAEA decided in 2003 to include the revision of TRS 364, as a topic of the EMRAS programme, "Environmental modelling for radiation safety".

2. GENERAL FEATURES OF THE REVISION OF IAEA TRS No. 364

Due to the large audience and extensive use of TRS 364, there is a need to keep such a document as relevant, accurate and consistent as possible : relevant first, because the purpose is to cover various assessment contexts such as routine releases and accidental conditions, atmospheric and liquid releases, etc. It should be accurate because mistakes should be corrected and avoided, up-to-date science should be incorporated (this could be difficult when data is scarce, as is the case for less mobile radionuclides). And last, it should be consistent : consistency between tables should be ensured, especially with regard to the radionuclides considered, particularly since integrated assessments require it.

An assessment of the quality of data obtained is of paramount importance to assure that any best estimates of the environmental parameters provided can be used in generic predictive models. In an ideal world, a critical evaluation of data would include a statistical analysis of the data. However, because of the constrained availability of resources, this was not attempted in TRS 364, and it may be difficult to carry it out in a revision.

In practice, the expected values and ranges given in TRS 364 are based on a variety of different approaches which are not always clearly specified for each value. They comprise statistical analysis, expert judgement or sometimes only a single experiment. For some of the tables (e.g. animal transfers) the approach has been specified for each value ; it is recommended to extend this approach to all other tables as far as possible.

Uncertainty should be mostly addressed by giving ranges of variation ; in some cases probability density functions could be derived through statistical analyses of databases (e.g. on some Kds and soil-to-plant transfer factors). Consideration should be given to the validity of extrapolation of statistical results to other categories (e.g. confidence intervals extrapolated from one radionuclide to another). When data is scarce or missing, a discussion should be held concerning the adequacy of the use of chemical analogues as a way of completing the tables. Variability could be reduced by revising classification systems (e.g. crop groups) and introducing co-factors (e.g. with Kds, soil-to-plant TFs).

Steady state models are routinely used for dose assessment, for screening purposes and operational releases. Information in TRS 364 is directly relevant for such models, when time dependency in transfers is not required. Many dynamic models are intended for dose assessment in emergency situations. For such models, often both empirical (using classification of systems) and semi-mechanistic approaches are used. The availability and applicability of improved dynamic models based largely on mechanistic information is limited. In general, such models have only been developed for a small number of mobile radionuclides (notably caesium and strontium), in particular types of system, such as caesium behaviour in forests or freshwaters or metabolically based models of animals. Such models have the additional advantage of being able to include the effect of countermeasures in a mechanistic way and complement other more generalised models used for radiation protection purposes. It is therefore important to consider whether the continued use of empirical transfer values is always justified and appropriate.

For an accurate mathematical description of dynamic processes a multi-compartment model, in which material fluxes are described by mechanism-based rate equations, is essential. Such dynamic models apply to cycling of radionuclides in ecosystems, retention and accumulation, migration and leaching. It is recognised that complex dynamic models, all the more because they often lack consensus and large validation, are outside of the scope of a revised TRS 364. An exception is made for dynamic one-compartment models in which processes can be modelled by a half-life concept. The dynamics of some of these processes are so important, that neglecting them would be a serious shortcoming of the new TRS. A consideration of changes with time in terrestrial and aquatic systems, expressed as ecological half-lives in different food products and reflecting processes involved in the long-term transfer from soil to vegetation, provides an important improvement for modelling.

Nomenclature should be consistent with official documents such as ICRU 65 (2001) "Quantities, Units and Terms in Radioecology" [7] which contains a list of units and quantities frequently used in the field of radioecology, and tries to harmonise units derived from varying different disciplines such as ecology, chemistry, medicine or physics.

3. ANALYSIS AND COMPLETION OF THE EXISTING TRS 364

3.1 Methodology

For each section of the new text (which will be based on ecological domains), the methodology for reviewing the current synthesis will be the following : list the processes of interest ; review the modelling aspects, in terms of compartments and processes, minimum model representation, alternative modelling, potential co-factors ; analyse the information on radionuclides currently taken into account ; consider the classification systems ; highlight weaknesses and mistakes ; and check the availability of new data.

When rewritten, the sections will contain some fundamental text devoted to explaining the main processes involved, with the usual associated parameters, co-factors influencing some processes or the determination of parameters values, and key equations used for describing and assessing the transfers. IAEA SRS 19 could be a basic reference for simple modelling. In addition, tables of transfer coefficients will be given with values depending on species (or groups of species), environmental conditions, co-factors, etc. Relevant references for processes and data will be quoted.

3.2 Foliar interception, retention and translocation

Foliar interception is a prominent process after initial release from a nuclear facility. The section is currently not clear enough and not self-sufficient for building an appropriate model structure. Foliar interception has a strong link with plant characteristics, not least because the plant stage of development is a very important factor. For contamination of surfaces, three deposition pathways are considered : direct dry or wet deposition and resuspension. With regard to the latter, only soil adhesion to vegetation is considered because it may affect the ingestion dose, inhalation is not relevant to the revision.

The interception values for dry deposition refer to experimental work mostly performed in the early nineteen seventies. Values relating to wet deposition refer mainly to two references published in 1965 and 1977. Meanwhile, much more data has become available which needs to be included, such as experimental and post Chernobyl studies. Also information on the seasonality of intercepted fractions relating to the biomass of different plant species could be greatly improved (in TRS 364 only one reference is given). Instead of providing individual experimental results, a critical compilation of values showing the dependence of intercepted fractions on precipitation intensity, time lag, season and plant species should be tabulated. It has also been deemed useful to introduce deposition velocities. Thje inclusion of irrigation processes has been identified as a need arising from the needs of waste assessment, and could be considered as a particular case of wet deposition.

Under accidental conditions, parameter values should not be averaged over the year. In such circumstances, the physico-chemical features of the source-term may be of importance (granularity, solubility). The amount of rainfall should also be specified. The European RODOS programme could be an important source of information for improving the Handbook with regard to these aspects.

In TRS 364, the processes of foliar absorption, translocation and retention need to be clearly distinguished. In TRS 364, a collation of different translocation coefficients for crops with time dependencies before harvest are given, however conclusions or recommendations on which values to use are not provided. The aim should be to derive generic parameter values from a review of a revised data compilation. In addition, the conclusions and recommendations in IAEA/ TECDOC 857 (1996) [8] should also be considered.

New data can be added on the transfer to fruit. Inclusion of time dependency in trees and understory vegetation (berries) and retention in the canopy information on interception and translocation might be useful as their characteristics are different from the other crop groups.

3.3 Soil retention and migration : Kds

Current classifications of soil systems are rather simplistic and generally limited to only four categories: sand, loam, clay and peat. This classification was also used in TRS 364, but the values were based on a relatively low number of experiments for a limited number of soils within each category. In recent years, the number of data for each category has greatly increased, and analysis of the data distribution has shown both a high variability and high degree of overlap. Revised classification systems are currently being developed which are based on mechanistic information including consideration of parameter values such as pH, soil nutrient status, % clay, exchangeable K and Ca in soil, moisture content of soil, organic matter content and the time that a radionuclide is present in a soil. Numerous multiregression analyses have been developed and give reasonable predictions on a local scale but have not been proven on a world-wide scale.

3.4 Soil-to-plant transfer factors

At the time that TRS 364 was prepared, the IUR had already introduced “crop groups” such as cereals, green vegetables, root crops, etc. It was, however, not yet then possible to use these crop groups to provide expected uptake parameters, so separate transfer values were provided for wheat, barley, rye, etc. These crop groups should now be introduced.

Currently, data given on soil-to-plant transfer factors given in TRS 364 generally uses three approaches :

(1) For Cs and Sr, separate values are given for different soil categories (sand, loam, clay and peat) with a pH constraint. In recent years, the number of data for Cs and Sr has greatly increased, and, as for Kd values, analysis of the data distribution has shown a high variability and high degree of overlap. Revised classification systems are currently being developed which are based on mechanistic information including consideration of parameter values of co-factors, such as pH, soil nutrient status, % clay, exchangeable K and Ca in soil, moisture content of soil, organic matter content and the time that a radionuclide is present in a soil, in a manner consistent with that adopted for Kds.

(2) Pu, Am and many other radionuclides, soil type is not considered. The data have largely been collated between 1980 and 1990. For some of them, e.g. Pu, Am, Co, Mn, Tc, Zn, new data are available. For others it is doubtful that sufficient values for updating tables are available.

(3) Less frequently studied radionuclides are generally based on much older literature. There are probably few data available.

Both for the second and third group it is probably worthwhile to consider the uptake data of stable elements. In particular, heavy metals have received considerable recent attention.

3.5 Animals

This section could be improved by a review of recent literature even if new data are more difficult to find than for plants. For many of the radionuclides, the tabulated data are based on a compilation originating from databases more than 20 years old. The intake rates of feedstuff by animals are based on European conditions only and should be supplemented by data for other areas of the world. Some FAO activities and results could be of use.

For ingestion doses, the application of equilibrium transfer coefficients for animal products is inappropriate for radionuclides with long biological half-lives. Therefore, in TRS 364 some transfer coefficients were modified to account for a known lack of non-equilibrium. This approach needs to be extended, possibly by providing biological half-life information, and evaluated. Furthermore, for strontium, iodine and caesium information on biological half-lives is available and can be compiled.

A statistical analysis of the data is not possible for TRS 364 because many of the values came from reviews using extensive data and individual data sets which were not available. Supplementation of recent data with these reviews was attempted, but was only possible using expert judgement. For some selected radionuclides a statistical analysis could be performed since sufficient data and data sets are available. However this would be extremely time consuming and is probably unrealistic. It is therefore recommended to use recently published reviews where statistical analysis have been attempted.

In the last decade detailed and improved information on the influence of stable analogues for caesium, strontium and iodine has become available and should be considered for inclusion in the revision of TRS 364.

3.6 Aquatic systems

This chapter should be rewritten and enhanced in terms of compartments and processes included, and environmental conditions, and needs to be consistent with IAEA SRS 19 ; in particular, bottom sediments and water particles should be distinguished, as well as trophic levels for fish.

The adsorption and desorption processes of radionuclides by suspended matter strongly influences the behaviour of radioactive substances in freshwater systems. Consequently, considerable research has been carried out on these processes which affect partition coefficient of suspended matter/water . The most common and simple approach for modelling adsorption/ desorption processes at equilibrium is based on the partition coefficient, Kd. The correlation between the partition coefficient and the chemical characteristics of the water body should be described. A variety of researchers have demonstrated that non reversible adsorption processes are of importance for some radionuclides, notably caesium. A table of non-reversible fraction rates for caesium in different freshwater ecosystems should be included.

In TRS 364, the table concerning fish is based on a large database for a wide range of elements. However, the method of selection for the expected values is not specified and appears to be inconsistent. However, data are quite comprehensive and might be supplemented by a few recent literature reviews on concentration factors for the edible portion of fish in freshwater environments.

3.7 Semi-natural systems : forests

It is important to include semi-natural ecosystems for caesium because the range of products harvested differs greatly from other ecosystems and the rate of transfer to food products is often much higher than for other ecosystems. Furthermore, the ecological half-lives of caesium in many products harvested from these ecosystems is much longer than in agricultural systems. For some populations, consumption of semi-natural products is common, for others it is confined to certain special groups. For both cases, such consumption can form a major proportion of ingestion dose in the mid-long term after deposition.

The section on semi-natural ecosystems in TRS 364 was largely based on simple aggregated transfer coefficients, since the inherent variability and complexity of such systems make predictions using other approaches difficult. Recently, some dynamic models have been developed which allow the estimation of transfer to certain forest products, but the number of products considered is limited. It is therefore recommended that the aggregated transfer coefficients are also used in the revision to be able to include as wide a range of products as possible. The extension of the range of radionuclides included should also be attempted. Fortunately, data availability has greatly increased since 1992, largely from the considerable focus in both Europe and the CIS on caesium transfer to forest products.

Some game species and humans consume large quantities of mushrooms which can take up large quantities of caesium. Mushroom consumption can therefore be a dominant pathway for the ingestion of caesium. Currently, much more data on mushroom transfer is now available. For arctic systems, a significant improvement of TRS 364 is now possible for quantifying transfer of caesium to reindeers and other arctic food products and its time dependency, based on a review carried out under the Arctic Monitoring and Assessment Programme (AMAP).

3.8 Food processing

Currently TRS 364 includes a lot of tables, which are particularly useful in a context of accidental management. It is recommended to simplify them by focusing on the most efficient processes for radioactivity reduction. For instance, focusing on the processing of dairy products and mushrooms.

4. EXTENSION OF TRS 364 IN SCOPE, COMPARTMENTS AND PROCESSES

At a methodological level, a discussion on chemical analogy should be introduced as a way to overcome the lack of data for some radionuclides, especially if consistency between the tables should be reached.

TRS 364 was limited to temperate climatic zones because, at the time, few data from other climatic zones were available. Since 1992, new data on both arctic regions and tropical environments have become available. From this new data it appears that the climate itself seems to have little direct influence on radioecological transfer parameters, but that nevertheless the impact of the climate on other important parameters is large. Climate and parent rock material determine, to a large extent, the development of soil type. In tropical areas, several soil types occur in which radionuclide uptake by crops consistently deviate from the expected values given in TRS 364.

Atmospheric resuspension and wash-off should be introduced as new processes because they are prominent secondary pathways of contamination. Concerning wash-off, in recent years, many attempts have been carried out to develop new approaches which allow modellers to predict radionuclide migration from catchments by using more simple generic models including half-lives and transfer factors. Due to the data available in the international literature following the Chernobyl accident, it is possible to produce a limited, but somewhat instructive, list of half-lives and transfer factors mainly for ^{90}Sr and ^{137}Cs. This list could be helpful for generic models applicable to both steady state and dynamic conditions.

5. CONCLUSION

The revision of the IAEA TRS No. 364, "Handbook of parameter values for the prediction of radionuclide transfer in temperate environments", published in collaboration with the IUR, is long overdue. Such a revision is an ambitious effort but essential if the recent progress in radioecology is to be incorporated and made available to many different users. Such an undertaking is only possible by an international collaboration launched through the IAEA/EMRAS programme.

Since such an activity demands expertise and resources, all institutions and all experts are welcome to contribute and participate through the existing channels : IAEA/EMRAS, IUR or personal contacts.

Acknowledgments

The authors are grateful to B. Howard, L. Monte, and M. Frissel for drafting the first version of the document which led to the launching of the current revision of the IAEA/TRS 364.

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