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Proposal for new best estimates of the soil-to-plant transfer factor of U, Th, Ra, Pb and Po

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ABSTRACT

There is increasing interest in radiological assessment of discharges of naturally occurring radionuclides into the terrestrial environment. Such assessments require parameter values for the pathways considered in predictive models. An important pathway for human exposure is via ingestion of food crops and animal products. One of the key parameters in environmental assessment is therefore the soil-to-plant transfer factor to food and fodder crops. The objective of this study was to compile data, based on an extensive literature survey, concerning soil-to-plant transfer factors for uranium, thorium, radium, lead, and polonium. Transfer factor estimates were presented for major crop groups (Cereals, Leafy vegetables, Non-leafy vegetables, Root crops, Tubers, Fruits, Herbs, Pastures/grasses, Fodder), and also for some compartments within crop groups. Transfer factors were also calculated per soil group, as defined by their texture and organic matter content (Sand, Loam, Clay and Organic), and evaluation of transfer factors' dependency on specific soil characteristics was performed following regression analysis. The derived estimates were compared with estimates currently in use.

1. Introduction

The transfer of artificial radionuclides along terrestrial food chains has been studied extensively during the last 40 years. The natural radionuclides U and Th have received less research attention than many artificial radionuclides, despite being naturally ubiquitous in the environment. There has been increasing interest in radiological assessments of the discharges of naturally occurring radionuclides in the terrestrial environment, both in terms of current releases from industrial sites as from the presence of historical contaminations.

Apart from the obvious presence of naturally occurring radionuclides (NORs) in uranium deposits, a wide range of uranium- and thorium-bearing minerals (and their daughters) are being mined and processed commercially. In most minerals, natural levels of radionuclides are very low. In others, e.g. zircon and rare earths the concentration of ^{238}U and ^{232}Th may be considerably elevated, with activities up to 40 and 70 kBq kg^{-1} for thorium and uranium, respectively. Enhanced levels of NORs may be associated with abandoned waste dumps, installations and surroundings from certain industries, which have involved extraction or processing of raw materials containing NORs. This can result in considerable exposure to the public. The most prominent examples in Europe, apart from the residues of uranium mining and milling, are the waste generated by the phosphate processing industry, the scales from the oil and gas extraction industry, the ashes from coal-based power production, and the slag produced by the metal mining and smelting industry (Vandenhove et al., 2000; IAEA, 2003).

The processes by which radionuclides can be incorporated into vegetation can either be (1) through activity interception by external plant surfaces (either directly from the atmosphere or from resuspended material), or (2) through uptake of radionuclides via the root system. For this compilation, we intended to assemble data restricted to soil-to-plant transfer. The soil-to-plant transfer factor (TF) is defined as the ratio of the concentrations of radionuclides in plant (Bq kg^{-1} dry mass) to that in soil (Bq kg^{-1} dry mass).

The primary objectives of this review were (1) to compile published information on the soil-to-plant transfer factor of U, Th, ^{226}Ra, ^{210}Po, ^{210}Pb; (2) to carry out a critical review of the data in

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terms of their quality and usefulness; (3) to propose new estimates for transfer factors (TF) for use in terrestrial food chain models; (4) to compare the values derived with previous estimates; and finally (5) to evaluate the dependency of the TF on specific soil and plant characteristics.

This study is carried out in the framework of the activities of a Working Group addressing the current revision of the *Handbook of Parameters Values for the Prediction of Radionuclide Transfer in Temperate Environments – TRS 364* (IAEA, 1994).

2. Data collection and treatment

2.1. Data collection and data acceptance

Literature collection consisted of peer-reviewed publications as well as reports. A total of about 200 references were consulted, and of these 104 were retained. Each reference was critically reviewed and data were retained/excluded based on the following criteria:

- Only individual data from matching crop–soil combinations were retained, with summary data from reviews being excluded;
- Experimental results had to be clear and concise. Clear information on sample collection and preparation was required and if there was any doubt whether concentration in crops (or TF) was expressed relative to fresh or dry mass, data were not considered;
- Information on TF was only included if there was clear indication of which plant compartment was sampled and analyzed;
- The minimal soil information required for associated TF data to be entered was the concentration of radionuclides in the soil and the type of contamination. If soil concentrations were expressed on wet weight basis (and no information available on soil water content to calculated concentrations per dry mass) or if contaminant level was expressed as available fraction, TF data were not included;
- Data from areas with high natural radioactivity were included, except when it concerned specific substrates (non-soils) (e.g. uranium tailings, red mud, phosphogypsum);
- Data from pot experiments were included, independent of pot size.

In addition to TF data, information was collected on, e.g. climate and experimental conditions, contamination history and contaminant concentration, soil type, soil characteristics (pH, sand and clay content, organic matter content, cation exchange capacity, exchangeable Ca and Mg and soil solution concentrations of Ca^{2+} and $Mg^{2+}$and amorphous iron content) and plant characteristics (Ca, Mg, K content, yield) in order to evaluate the possible dependence of the TF on these properties. In cases where the TF or plant concentrations were expressed relative to fresh weight, the fresh weight/dry weight conversion coefficient was applied (IAEA, in preparation). Soil adhering to roots or leaves has the potential to carry NORM to the food chain. Most articles did not include information if the food products were washed or peeled, which may be an additional source of variability.

2.2. Data treatment and statistical analysis

The TF values were grouped according to two criteria (1) crop groups and (2), if sufficient data were available, plant group/soil texture. Plants were grouped in several crop groups as agreed by the EMRAS TRS-364 working group. Crop plant groups included the following: Cereals were considered in one category – All cereals – and as two separate groups, Cereals (e.g. wheat, barley, rye) and Maize because maize grain is more than the other cereal grains used as animal feed. TF data for rice were not included in the Cereal data. The other groups consist of: Leafy vegetables (e.g. lettuce, spinach, Chinese cabbage, Brussels sprouts), Non-leafy vegetables (e.g. tomato, cucumber, egg plant), Legumes (e.g. peas, beans), Root crops (e.g. carrot, radish, turnip), Tubers (e.g. potatoes), Fruit (e.g. apple, pear, berries), Herbs (e.g. mustard, parsley), Other crops (e.g. sunflower seeds, tea leaves), Grasses (single species), Natural pastures, Leguminous fodder (e.g. alfalfa, clover). Additionally, TF to non-edible plant parts (e.g. straw, shoots of root crops, shoots of legumes) were recorded. TF was also calculated for more global crop groups: “Fodder” comprising leguminous pasture species (like clover), straw from Cereals and Maize and shoots of Non-leafy vegetables, Root crops, Tubers; “Pastures/grasses” comprising TF data to Natural pastures and grasses.

Where sufficiency of data allowed, TFs were also calculated based on the plant group/soil texture criterion. Soils were grouped according to the percentages of sand, clay contents and organic matter. A soil was included in the Organic group if the organic matter content was $\geq$20%. For mineral soils, three groups were created according to the following criteria: Sand group: sand fraction $\geq$ 65%; clay fraction $<$ 18%; Clay group: clay fraction $\geq$ 35%; Loam group: mineral soils not fitting above criteria.

It could be argued that in the analysis of the soil-to-plant TF for the long-lived natural radionuclides U, Ra and Th the dependency of the TF on the soil concentration should be considered. Sheppard and Sheppard (1985) and Sheppard and Evenden (1988a) reported a log-normal dependency of TF-U on soil concentration. On the other hand, Blanco et al. (2002) did not find a relation between TF-U and soil concentration. For this compilation, covering a broad range of soil concentrations (7–250,000 Bq kg^{-1} for U, 4–60,000 Bq kg^{-1} for Ra and 4–89,000 Bq kg^{-1} for Th), different soil groups and crop groups and experimental conditions, no relation between TF or log TF and the soil concentration was found ($R^2 < 0.01$). However, it should be born in mind that non-linearity can contribute to the uncertainty in the TF.

For all crop groups considered, the following dataset descriptors were calculated: geometric mean (GM); geometric standard deviation (GSD); arithmetical mean (AM), standard deviation (SD), minimum (min) and maximum (max) values. GM and GSD are preferred to AM and SD since TF values are generally log-normally distributed. GM and GSD were only calculated when the number of observations was $\geq$3 for each crop group. The number of observations is also given. The above descriptors were also calculated per soil group (Clay, Loam, Sand, Organic) if at least 10 entries were available for that crop group. Discussion of data always refers to GM (GSD), unless specified differently. GM (GSD) and AM (SD) were clearly different. Overall, the value derived for AM is 3 to 5-fold higher than the value derived for GM.

Statistical analysis of data was performed with the statistical software packages *Statistica for Windows* (Statsoft, 2004). Outlier analysis was performed using box–whisker plots. A value was defined an outlier if it was over 1.5-times the interquartile range (for the log-transformed data). Normality of the dataset was evaluated with the Shapiro Wilk’s test and the Kolmogorov–Smirnov test for normality. Only log-transformed datasets were normally distributed. Ratio data, as the TF, tend to be log-normal based on the central Limit Theorem. Normal distribution could not be verified if the number of data within a crop group was too small (e.g. 3). Log TF data were also normally distributed within a broad soil category [e.g. log TF data obtained for all crop groups for a specific soil category (e.g. Sand)] or crop category (e.g. log TF data obtained irrespective of soil type for a specific crop group e.g. All Cereals). As such the conditions were fullfilled to calculate GM and GSD and perform comparisons between groups with ANOVA. Mean values were ranked by Tukey’s multiple range tests when more than two