

Determination of cesium transfer factors by instrumental neutron activation analysis



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ABSTRACT

Food-chain models are used to predict radionuclide ingestion after fallout deposition. These models include those transfer processes (soil-to-plant transfer factor(s) [TF], plant-to-animal transfer coefficient(s) [TC] and concentration ratio [CR]) that are likely to be important for radiological assessment. The range of variability for transfer factors for the same plant groups is great, about 4–5 orders of magnitude, which limits their applicability. A better way to determine the best estimate the factors for radiocaesium and other important radionuclides is if the site-specific data are available. Soil, plant and animal samples were collected from a pasture area in Hungary during the vegetation period in 2016. Stable ¹³³Cs concentration was analysed by comparative method with neutron activation analysis (NAA). The comparator and the samples were irradiated in thermal neutron flux $2.55 \times 10^{12} \text{ ncm}^{-2}\text{s}^{-1}$ for 2 h (soil) and 6 h (vegetation, animal samples) in the TRIGA Mark II research reactor at the Nuclear Engineering Teaching Laboratory. After an appropriate decay time (12 days) the samples were measured by gamma-spectrometry and analysed. The observed stable caesium TC_{pm} (0.48–0.53) and CR_{pm} (0.41–0.45) were very close to ¹³⁷Cs factors in the IAEA 2009 Report of 0.49 and 0.54, respectively. This methodology is particularly suitable for the simultaneous study of natural caesium in ecosystem compartments. Consequently, the transfer of stable caesium in a pasture field may be regarded as a useful analogy in predicting the long-term changes of ¹³⁷Cs affected by site-specific environmental factors.

1. Introduction

One of the serious impacts for the environment and human health is the proper evaluation of radioactivity from elevated naturally occurring isotopes or those from industrial activities. Artificial radionuclides produced by nuclear explosions and nuclear facilities, if released into the environment, may reach the human body through several transfer pathways. This is considered as one of the important routes through which radionuclides can enter the human body via the food-chain pathway (Pochin, 1988). There are two important radionuclides, ⁹⁰Sr and ¹³⁷Cs, for the assessment of radiation exposure to the public because of their relatively long half-lives (~30 years), and their extraordinary transferability in the environment. ¹³⁷Cs has physical and chemical properties similar to potassium and ⁹⁰Sr behaves calcium, and as such these radionuclides elements get actively included in the food-chains and ultimately animal and human bodies.

Radionuclides can be deposited on soil either by direct deposition from the atmosphere or from the use of surface water for irrigation. The terrestrial food-chain models are designed to accept an input of

radionuclides from either the atmosphere or the hydrosphere. These models include those transfer processes that are likely to be important for the radiological assessment of routine discharges. The soil-to-plant transfer factor(s), TF(s) and plant-to-animal transfer coefficient(s), TC(s) are two of the most important parameters widely used to estimate the internal radiation dose from radionuclides through food and water ingestion. The large collections of TFs and TCs were collected by different organizations (IAEA, 2010, 2009; IUR, 1992; NRPB, 1994; UNSCEAR, 2008). However, these are not exhaustive as a wealth of TF and TC data has been produced by other scientists (Djelic et al., 2016; Frissel et al., 2002; Nisbet and Woodman, 2000; Wang et al., 1999) that has not been included in the above mentioned collection. All these results have been used as criteria in order to accept the data into the whole data base or to reject them. Transfer factors defined as the ratio between what is in the plant and what is in the soil are the most usual way to quantify the biological and geological relocation processes. However, their uses have some difficulties. The range of variability for transfer factors of the same plant groups is large, about 4–5 orders of magnitude, which limits their applicability (Nisbet and Woodman,

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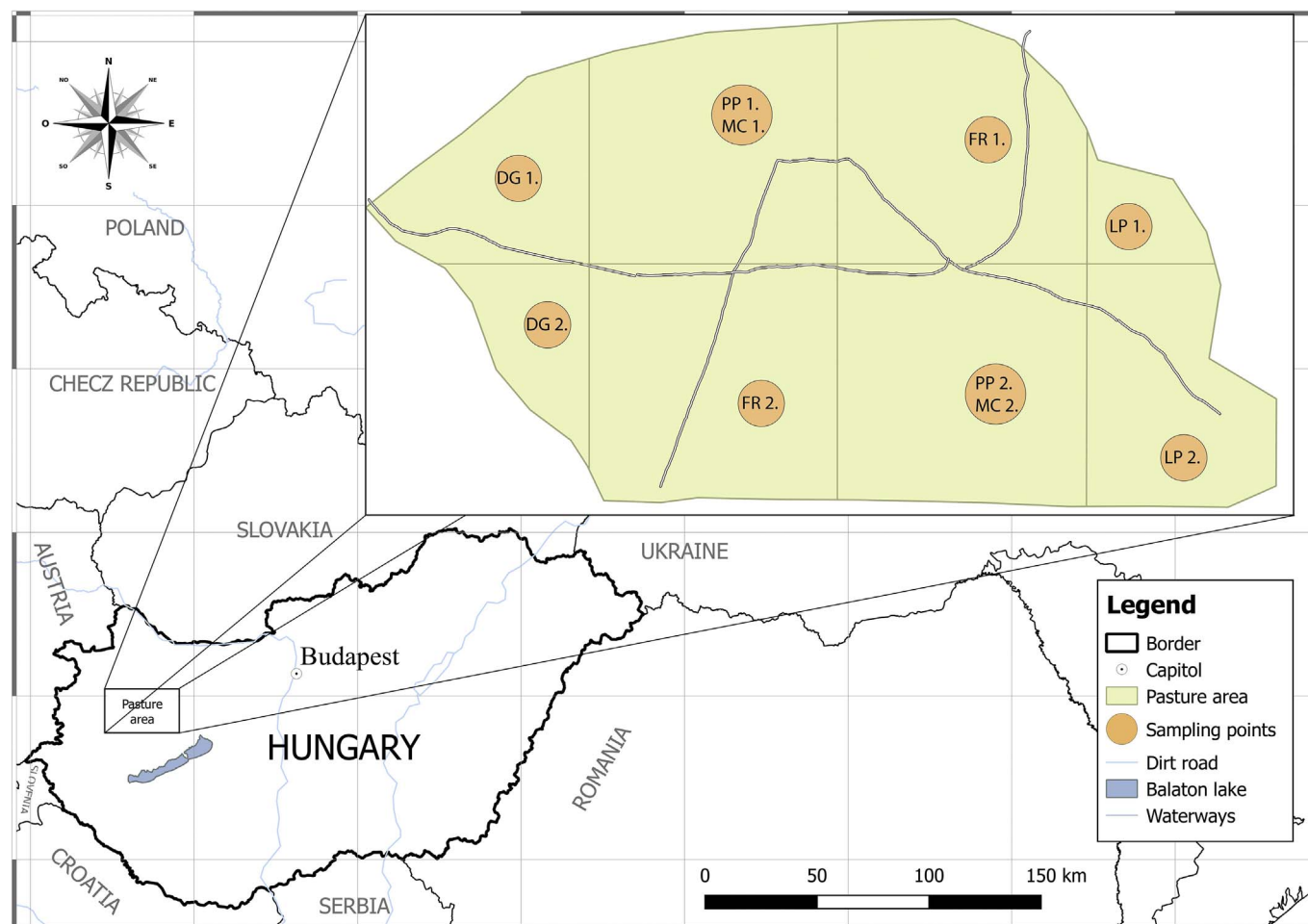


Fig. 1.

2000). The reason for the different values of the transfer factors is obvious; this macroscopic parameter integrates a number of chemical, biological, hydrological, physical soil properties, and plant and animal physiological processes. Each of these characteristics show their own variability and in addition may be influenced by external factors such as climate and human agricultural practices (Ehlken and Kirchner, 2002). A better way to determine the best estimate TFs and TCs for radio-caesium and other important radionuclides is to utilize site-specific data if available. The estimation of site-specific transfer factors and their coefficients for radionuclides in various pathways is essential for a more accurate assessment of the long-term radiological hazard to the population, especially in the surrounding agricultural region of a nuclear power plant.

The determination of TFs and TCs in environmental matrices means the concentration of radionuclides must be determined with good accuracy. The determination of radio-caesium in biological and geological materials is notoriously difficult in the uncontaminated environmental samples. If a sample with sufficient radionuclide such as ^{137}Cs is available, it becomes easy to measure with gamma ray spectrometry. However, often there is not always an appropriate amount of sample available. Therefore, a freshly added radiotracer method is another possible method, although with this manner the added radio-caesium may not be at equilibrium in the soil environment. These errors occur because TFs of radio-caesium obtained under experimental conditions were usually higher than those of ^{137}Cs under environmental condition (Choi et al., 2003). To solve this problem, the naturally occurring stable element ^{133}Cs can be quantified, which should exist in equilibrium. Several papers have shown that TFs of ^{133}Cs and ^{137}Cs have a positive

significant correlation (Tsukada et al., 2003) and similar observations were reported for rice (Uchida et al., 2009; Uchida and Tagami, 2007) and potato (Tsukada and Nakamura, 1999). Hereby, the measurement of the stable caesium could be a useful technique for obtaining more realistic factors of ^{137}Cs .

The analysis of the stable isotope of caesium in different environmental samples is performed by a number of methods that have the needed selectivity and sensitivity. However, caesium is highly electro-positive and has the lowest ionization energy of any element so that electroanalytical techniques are not applicable, and for spectrophotometry sensitivity is very poor. Inductively coupled plasma mass spectrometry (ICP-MS) and neutron activation analysis (NAA) are the best applicable and preferred technique of analysis for stable ^{133}Cs with the very good detection limits (Akhter et al., 2003; Iyengar and Woittiez, 1988; Severo et al., 2004). Advantages to ICP-MS include the favourable detection limits, high throughput and the ability to measure more than one element simultaneously. The major disadvantage is the difficulty of sample preparation which needs chemical dissolution and the potential interference from near-by interfering ions. The NAA method has several advantages: it is a non-destructive method, high degree of sensitivity, and generally does not require extensive sample preparation or processing. Major limitations of NAA include a nuclear reactor to irradiate the samples and the need for a longer decay time to achieve good detection limits. The analysis for ^{133}Cs with NAA method and determination of TF, TC and CR for caesium can be reliably done for different environmental and biological samples as described in this study.

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