Journal of Environmental Radioactivity 164 (2016) 19–25

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Ageing impact on the transfer factor of ^{137}Cs and ^{90}Sr to lettuce and winter wheat

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ARTICLE INFO

Article history:
Received 13 February 2016
Received in revised form
20 June 2016
Accepted 23 June 2016

Keywords:
Transfer factor
Aging
Caesium
Strontium
Winter wheat
Lettuce

ABSTRACT

The study focuses on long-term (extending from 1 to 10 years) lysimeter experiments of the transfer factor of ^{137}Cs and ^{90}Sr to lettuce and winter wheat crops. Transfer factors (F_vs) were the ratio of the activity concentrations of the radionuclides in crops to those in soil, both as dry weight (Bq kg^{-1}). F_vs of ^{137}Cs to lettuce decreased significantly with ageing; geometric means for the 1st, 2nd and 10th year contaminated soil were 0.114, 0.030 and 0.013, respectively. However, a significant decline of F_vs for ^{137}Cs was only seen between the 1st and 2nd year for both wheat compartments (straw and grains) which disappeared thereafter. The dynamic of ^{137}Cs F_vs may be explained according to the distribution coefficient experiment (K_d) which had a value of 3600 L kg^{-1} showing a high affinity of the clay minerals for caesium. Desorption data revealed that Cs fixation enhanced with ageing. The mechanism involved may be an initial sorption of caesium species to the surface soil particles followed by progressive irreversible fixation to the interlayer of the porous clay minerals. F_vs of ^{90}Sr were high and showed trivial variation for both crops for the time course studied. Sorption of Sr^{2+} species to the clay mineral may be the governing process, which was supported by high desorption percentage (ranged 77%) with low K_d, i.e. 10 L kg^{-1}. In general, higher F_vs of ^{137}Cs and ^{90}Sr for lettuce was observed in comparison to winter wheat. The diversity of plant species and root systems would play essential roles for such behaviours.

1. Introduction

Following nuclear weapons testing and nuclear power plant accidents, for instant Chernobyl in 1986 and Fukushima in 2011 (Steinhauser et al., 2014; Thielen, 2012; IAEA, 2010), radionuclides are released to the environment, especially the fission products ^{137}Cs and ^{90}Sr. These long-lived contaminants form potential pathway of radiation to human health through food chain and contribute substantially to overall radiological dose. Transfer factor is a vital parameter to evaluate the radiological impact assessment and environmental safety studies. The soil-to-plant transfer factor accounts for the uptake of radionuclides *via* plant roots and represents the activity concentration ratio of the radionuclide per unit dry mass in the plant (Bq kg^{-1}) to that in the soil, designated as F_v (IAEA, 2009, 2010). The dynamic of F_v differ by provenance, hence, there has been an interest to determine F_v for various foodstuff worldwide. Empirical F_vs for most important agricultural products in much of Europe and the USA are already documented; however, data for certain climate and soil types are not fully covered (Frissel et al., 2002; IAEA, 2006).

The Atomic Energy Agency (IAEA) made extensive reviews on the radionuclide aggregated-transfer factors for a variety of foodstuff (IAEA, 2009, 2010). The data included were useful for routine release of radionuclides to the environment from nuclear facilities, in which equilibrium has been established between the movements of radionuclides into and out of the environment compartments. For cases dealing with acute contamination, such as nuclear accidents, equilibrium cannot be assumed, and the rate of transfer between compartments must be assumed to vary with time (IAEA, 2006, 2010). The migration and mobility characteristics of radionuclides in soil differ depending on the soil properties (soil pH, texture, concentrations of exchangeable calcium and potassium, organic matter content, etc.), climate conditions, plant species, land use and management practice (Fernandez et al., 2006; IAEA, 2006; Krouglov et al., 1997; Yamaguchi et al., 2007). On a long-term, there will be a continuous change of radionuclide bioavailability in the soil due to the radionuclide mobility (related to its soluble forms) and redistribution of the radionuclide in the root zone. Several investigations have been carried out to evaluate the impact of

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http://dx.doi.org/10.1016/j.jenvrad.2016.06.019
0265-931X/

ageing on the transfer factor of ^{137}Cs and ^{90}Sr (analogues of K^+ and Ca^{2+}, respectively) from contaminated soil to various plants (Choi et al., 2011; Djingova et al., 2005; Frissel et al., 2002; Gerstman and Schimmack, 2006; Gomez and Brown, 2015; Nisbet and Shaw, 1994a; Noordijk et al., 1992; Schimmack et al., 2004, 2007; Twining et al., 2004).

There is little information on the transfer of anthropogenic radionuclides to plants in semi–arid and arid regions in comparison to humid and temperate regions, with the fact that the climate in Syria is semi-arid. The work performed by Al-Oudat and Al-Asfary (2006) depicted the transfer factor and ageing impact of ^{137}Cs and ^{90}Sr in local conditions to several groups of crops including cereals and leafy vegetables as well as orchard trees, i.e. olive and apricot trees and grape vines. Yassine et al. (2003) investigated the effect of ageing and soil characteristics on the transfer factor of ^{137}Cs and ^{90}Sr to the edible part of cereals, leafy, non-leafy and leguminous vegetables. Recently, transfer factor of ^{90}Sr and ^{137}Cs to lettuce and winter wheat at different growth stage applications using lysimeters in an open field experiment has been released (Al Attar et al., 2015).

The present study focuses on the effect of ageing, i.e. 1–10 years, on the transfer factor of ^{137}Cs and ^{90}Sr to lettuce (*Lactuca sativa* L.) and winter wheat (*Triticum aestivum* L.). The chosen crops have different physiological functioning and are representative of leafy vegetables and cereals, respectively, according to the IAEA classifications of plants (2009). In addition, wheat is one of the main contributors to the Syrian economy with an annual production of 3.1 million tonnes in 2010 (FAO STAT, 2015) and it forms a foremost portion of the daily-diet of many countries. Lettuce is one of the major leafy vegetables in the Mediterranean region and basically consumed throughout the year. The data acquired form a baseline for future radiological assessment studies for the influence of long-term contamination by these fission radionuclides.

2. Materials and methods

2.1. Experimental design and climate conditions

The experiment was conducted using lysimeters in an open field at the agricultural station of the Atomic Energy Commission of Syria, *viz. Soujeh*, 28 km northwest of Damascus (33° 47′ latitude, 36° 07′ longitude, *ca.* 1100 m above the sea-level). The climate of the site is humid with annual rainfall of 311 mm and mean temperature of 14 °C (January 6.2 °C, July 25.3 °C) with wind speed ranging 0.0–3.28 m s^{-1} and air humidity 26.8–68.3%. To comply with radiological protection legislations, the site was lined with high-density polyethylene sheet. To avoid site disturbance by birds or animal incursion during the growing seasons, it was necessary to build an enclosed barbed wire chain-link fence, which was reinforced at the base.

Each cylindrical lysimeter (40 cm diameter × 50 cm height) was filled-up with *ca.* 40 kg of dry Incepisol soil. They were sown in December with seeds of cultivars of lettuce (*Lactuca sativa* L. *var.* local) and winter wheat (*Triticum aestivum* L. *var.* ACSAD), with a particular concern that this cultivation was the solo practice for the oldest soil during the 10 years. Sowing density was *ca.* 20 g m^{-2}. The experiment was performed without the addition of fertilizer in order to meet the local conditions at the area of study. Five replicates per crop and per year were carried out, giving a total of 10 lysimeters per year/treatment. They were distributed in randomised block design. Sowing and harvest were implemented according to the standard local agricultural scheme for one cycle of cultivation with manual remove of weeds. The total growth-period of the crops at the year of the experiment was 153 and 160 days for lettuce and wheat, respectively. Watering was performed on a regular basis throughout the growth-period of the crops with an average rate of 1.5 L per week for each lysimeter that met the field capacity so that drought stress and water-leakage were avoided.

2.2. Soil characterisation

The soil used was Inceptisol, which is *Calcisols* according to FAO WRB (2014). The soil is brown reddish and is the main agricultural soil in the country. The relative abundance of the clay minerals in the soil was determined using powder X-ray diffraction, XRD, (Stoe Transmission diffractometer, Model STADI-P). Soil pH was measured in soil/$CaCl_2$ (0.01 M) solution 1:5. Soil texture was determined in accordance to particle size distribution (Black et al., 1984). The organic matter and cation exchange capacity (CEC) were analysed as described by Page et al. (1982). Ca^{2+}, Mg^{2+}, Na^+ and K^+ were determined by Atomic Absorption Spectroscopy using Analytik Jena-AGVario 6.

Using XRD, the most abundant fraction of clays size fraction is montmorillonite. The physical and chemical properties of soil are summarised in Table 1. According to IAEA classification of soil-type (IAEA, 2010), when clay content exceeds 35% the soil is clay-group. Since silt content was over 50%, it is a silty-clay soil. The neutral pH and low percentage of organic matter were the main characteristic features of the soil studied.

2.3. Deposition of radionuclides

Each lysimeter of the 1st year was contaminated, before sowing, with 1 L of a mixed carrier free radioactive solution with activity of 148 ± 7 of ^{137}Cs and 127 ± 5 kBq of ^{90}Sr. The chosen activities of the radionuclides were similar to those used for the contamination of 2nd and 10th year lysimeters. Uncertainty of the activity concentrations of the stock solutions was estimated (according to Uncertainty Estimation No. PM-09/1, Atomic Energy Commission of Syria, AECS) at 95% confidence level and found to be ±6% for ^{137}Cs and ±1.9% for ^{90}Sr. Contamination was performed by pipetting the radioactive solution carefully and uniformly on the surface of each lysimeter in about 40 points. To prevent the radionuclides falling into, and concentrating within, soil cracks, 1 L water was applied to each lysimeter two days before the addition of the radioactive solution in order to stack the soil. This in turn ensured that the radioactive solution was spread evenly over the soil surface and less likely to resuspend by wind action. Homogenisation of the top 20 cm was performed manually.

Table 1
Mechanical and chemical properties of soil prior sowing (soils of the years studied).

Analyst	Result
pH (in H_2O)	8.1 ± 0.3
pH (in $CaCl_2$)	7.5 ± 0.1
Soil size distribution	
Clay%	38.7 ± 8.3
Silt%	51.0 ± 6.4
Sand%	10.3 ± 1.9
Exchangeable cations ($cmol_c\ kg^{-1}$)	
K^+	1.3 ± 0.1
Na^+	0.9 ± 0.1
Ca^{2+}	20.0 ± 0.5
Mg^{2+}	33.6 ± 0.7
Cation exchange capacity, CEC ($cmol_c\ kg^{-1}$)	55.8
Soluble cations ($cmol_c\ kg^{-1}$)	
K^+	0.30 ± 0.02
Na^+	11.2 ± 0.9
Ca^{2+}	18.1 ± 1.4
Mg^{2+}	9.0 ± 0.7
Organic matter %	0.8 ± 0.2