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Sustainable agricultural use of natural water sources containing elevated radium activity

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HIGHLIGHTS

- The environmental implications of using water containing Ra for irrigation were investigated.
- Radium was found to accumulate in crops leaves following the evapotranspiration current.
- Sorption of ^{226}Ra to soil particles hinders its matrix mobility.
- Crops can be irrigated with the activity of ^{226}Ra of 0.6–1.6 Bq L^{-1}.

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ABSTRACT

Relatively elevated concentrations of naturally occurring radium isotopes (^{226}Ra, ^{228}Ra and ^{224}Ra) are found in two main aquifers in the arid southern part of Israel, in activity concentrations frequently exceeding the limits set in the drinking water quality regulations.

We aimed to explore the environmental implications of using water containing Ra for irrigation. Several crops (cucumbers, melons, radish, lettuce, alfalfa and wheat), grown in weighing lysimeters were irrigated at 3 levels of ^{226}Ra activity concentration: Low Radium Water (LRW) < 0.04 Bq L^{-1}; High Radium Water (HRW) at 1.8 Bq L^{-1} and (3) Radium Enriched Water (REW) at 50 times the concentration in HRW. The HYDRUS 1-D software package was used to simulate the long-term ^{226}Ra distribution in a soil irrigated with HRW for 15 years. Radium uptake by plants was found to be controlled by its activity in the irrigation water and in the soil solution, the physical properties of the soil and the potential evapotranspiration. The ^{226}Ra apeared to accumulate mainly in the leaves of crops following the evapotranspiration current, while its accumulation in the edible parts (fruits and roots) was minimal. The simulation of 15 years of crop irrigation by HYDERUS 1-D, showed a low Ra activity concentration in the soil solution of the root zone and a limited downward mobility. It was therefore concluded that the crops investigated in this study can be irrigated with the natural occurring activity concentration of ^{226}Ra of 0.6–1.6 Bq L^{-1}. This should be accompanied by a continuous monitoring of radium in the edible parts of the crops.

1. Introduction

Relatively elevated concentrations of natural radium isotopes are found in groundwater in the southern part of Israel in the two main aquifers of the Negev Desert and the Arava Valley: the Nubian Sandstone aquifer (Kurnub Group) and the Lower Cretaceous aquifer (Judea Group). Radium is transferred from the host rock into the aquifer by geochemical processes and is commonly found in the groundwater as four isotopes: ^{226}Ra ($T_{1/2}$ = 1600 y) from the ^{238}U decay series, ^{228}Ra ($T_{1/2}$ = 5.75 y), and ^{224}Ra ($T_{1/2}$ = 3.66 d) from the ^{232}Th decay series and to a lesser exten ^{223}Ra ($T_{1/2}$ = 11.435 d) from the ^{235}U decay series (Vengosh et al., 2007). The water in some of the wells in the Southern Arava contains radium isotopes at concentrations of 1–2 Bq L^{-1}. These activity concentrations exceed the Maximum Contaminant Level (MCL) for drinking water in Israel set at 0.5 Bq L^{-1} and 0.2 Bq L^{-1} for ^{226}Ra and ^{228}Ra, respectively (Koch and Haquin, 2008).

With regard to uptake by plants, most of the information is expressed as a transfer factor (*TF*), which is calculated as the ratio of the element concentration in plants to its concentration in soil. A comprehensive compilation of soil to plant transfer factors was

Abbreviations: Ra, Radium; BTC, breakthrough curve; ET_p, potential evapotranspiration; MCL, Maximum Contaminant Level; TF, transfer factor; REW, Radium Enriched Water; HRW, High Radium Water; LRW, Low Radium Water.

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conducted by the International Atomic Energy Agency (IAEA, 2010), in which they are reported separately for temperate environments and for tropical and subtropical environments. Ra transfer factors are reported for up to 12 crop categories, for different plant tissues in part of the crop categories, and for up to 4 soil types. The largest amount of Ra data relates to temperate environments, far fewer data are available for tropical ecosystems and none are reported for subtropical environments (to which the Arava Valley belongs). The *TF* depends on soil characteristics, plant type, tissue type, climate conditions and the physico-chemical form of the radionuclide (Vandenhove et al., 2005; Vandenhove et al., 2009; IAEA, 2010).

Field and laboratory measurements can be combined with mathematical models to yield better predictions of the long term distribution pattern of radionuclides in the soil. Particularly, in cases where environmental and health hazards can occur under conditions of regularly fertigation with radionuclides. Mechanism-base numerical models have become a frequently used tool for simulations and pre dictions related to water and ions in the vadoze zone (Van Genuchten and Šimůnek, 2004). Long-term (<200 years) simulation of ^{226}Ra fate in agricultural ecosystems, where phosphogypsum fertilizer was applied for 27 years, was studied by Coelho et al. (2013), using the HP-1 multicomponent transport model. The software combines HYDRUS 1D (Šimůnek et al., 1998), a model simulating one-dimensional variably saturated water flow and ion transport in soils, with PHREEQC (Parkhurst and Appelo, 1999), a model that calculates geochemical reactions and ions distribution in soils.

Elevated radium concentrations may be a dominant factor in the potential utilization of groundwater as drinking water, as well as for agricultural purposes. Natural occurring radionuclides, including radium, are present in foodstuffs at varying activity concentrations. In prone areas, where radium concentrations in the ground water are significantly higher than the limits in the drinking water standards, it is important to investigate the effect of using such water sources for agricultural purposes. Irrigation with water containing elevated radium concentrations may contaminate the soil and the radium may consequently find its way into the food chain.

The overall objective of the current study was to develop criteria for the sustainable use of water containing high level of radium as irrigation water. Specifically, we aimed to explore chemical reactions of the added radium to soil particles and its movement in the soil, to investigate radium uptake and transport in plants and to develop a comprehensive soil-plant model for predicting Ra uptake in plants.

2. Materials and methods

2.1. Lysimeter studies

To study the effect of drip irrigation on radium accumulation in the soil and plant uptake an experimental setup consisting of 9 weighing-drainage lysimeters was installed at Yotveta in the Arava Valley in southern Israel. The lysimeter system, illustrated in Fig. 1, allowed us to carry out controlled experiments in which water and Ra fluxes were measured with high accuracy, and hence, to compute water and ions balances. Each lysimeter consisted of a PVC container filled with soil, a bottom layer of highly conductive porous media (rockwool) in tight contact with the soil, and a drainage pipe filled with the same material extending downward from the lysimeter bottom. The height and the diameter of the growth containers were 0.85 m and 1.5 m, respectively, corresponding to a volume of 1.5 m^3. The rockwool drainage extension prevented saturation at the lower soil boundary while permitting water to flow

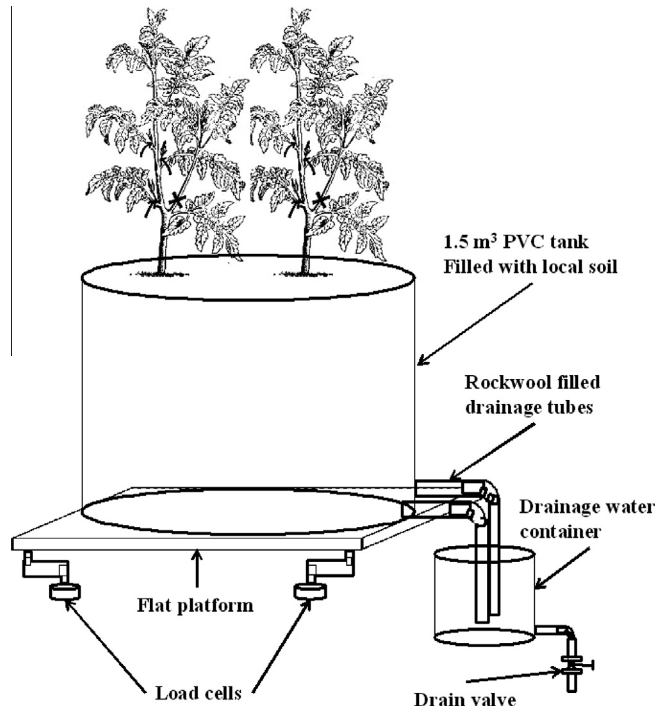

Fig. 1. A schematic representation of a single weighing-drainage lysimeter.

out of the soil and be collected. The lysimeter system included automatic water and fertilizer preparation and delivery. Ion and water boundary conditions of the lysimeters are as in Tripler et al. (2012). Individual lysimeters were positioned on square weighing platforms with load cells situated in each corner.

All lysimeters were filled with local soil, either an Arava loamy-sand soil, or an Arava sandy soil). The physical and mechanical parameters of the soils are described in Table 1. Both soils have average activity concentrations of ^{226}Ra of 21 ± 3 Bq kg^{-1} and ^{232}Th of 12 ± 2 Bq kg^{-1}.

Several controlled experiments were carried out at three radium concentration levels of the irrigation water: (1) lysimeters 1–3 with Radium Enriched Water (REW) – 50–100 Bq L^{-1}; (2) lysimeters 4–6 with High Radium Water (HRW) at 1.8 Bq L^{-1}; and (3) lysimeters 7–9 with Low Radium Water (LRW) – <0.04 Bq L^{-1}. The purpose of the REW treatment was to simulate an accelerated process of accumulation over a long period of time, i.e. to study the effect of many years of irrigation with water containing high concentration of radium. Along the course of the entire study, the daily irrigation quantity was set so that the drainage is 1/4–1/3 of the total irrigation water amount. This criterion afforded optimized steady state conditions of water content and soil solution ions concentration. Each treatment was equipped by an irrigation valve and a water meter, which gave a signal pulse, for every 5×10^{-4} m^3 of delivered water, to an attached automated controller. The daily water balance was calculated from:

$$\int_0^L \frac{\partial \theta}{\partial t} dZ = Ir(t) - Dr(\psi) - ET(\psi, ET_p) \quad (1)$$

where θ (m^3 m^{-3}) is the water content, t is the time, z is a specific depth of interest, L is the total depth of the lysimeter, Ir is the irrigation water amount, Dr is the collected drainage water at the bottom of the lysimeter, ET is the evapotranspiration, whereas ψ and ET_p represents the soil's matric head and the potential evapotranspiration, respectively. The right hand side of Eq. (1) expresses the measured difference in the weight of the lysimeter between 00:30 and 23:30 of the same day.