Transfer of Ra-226 to Chinese cabbage (Brassica pekinensis Rupr.) from Soil Contaminated with U-Mill Tailings

Marko Černe
Jožef Stefan Institute
Jamova 39
1000 Ljubljana, Slovenia
marko.cerne@ijs.si

Borut Smodiš, Radojko Jačimovič
Jožef Stefan Institute
Jamova 39
1000 Ljubljana, Slovenia
borut.smodis@ijs.si, radojko.jacimovic@ijs.si

ABSTRACT

Soil-to-plant transfer of natural radionuclides is a common phenomenon in uranium mining and milling areas. The Brassica plants are known to have the ability to accumulate radionuclides and are therefore suitable for potential phytoremediation of soils contaminated with U-mill tailings. In the present study, transfer of $^{226}$Ra to Chinese cabbage (Brassica pekinensis Rupr.) was investigated from soil contaminated with U-mill tailings (UMT). A pot experiment was carried out in order to provide the growing conditions controlled as far as possible. $^{226}$Ra concentration ratios were calculated for different levels of soil contamination under various growing conditions. Ca in soil and plants was considered due to possible impact on the $^{226}$Ra uptake process. Preliminary results of $119 \pm 6$ and $562 \pm 28$ Bq kg$^{-1}$ dry mass of $^{226}$Ra in cabbage leaves for 20% and 60% of UMT in the soil, respectively indicated increased accumulation of $^{226}$Ra in more contaminated soil. Measurement results are presented and the $^{226}$Ra transfer from soil to plants is critically evaluated.

1 INTRODUCTION

Uranium mining and milling activities may have an impact on higher levels of natural radionuclides in the environment nearby [1]. Long-lived radionuclides contained in U-ore processing materials are of major radiological concern due to inner or external exposure of ionizing radiation received by the inhabitants living close to the uranium mine areas [2]. Radionuclides emitted from the waste disposal sites can retain in the soil by adsorption on the soil colloids thus becoming immobile or more mobile depending on the soil properties, physicochemical conditions and microbial activity of soil [3]. The fraction of radionuclides that is soluble in the soil solution and exchangeable from soil colloids to the soil solution is bioavailable for the plant root uptake [4]. Radionuclides accumulated in the plants are taken up by roots from the contaminated soil [4] or/and by leaves via wet or dry deposition of the resuspended radioactive material [5]. The uptake via leaves in terrestrial plants differs quite a lot from one plant species to another and can be less effective and more limited than root uptake, but it is very important in aquatic plants [6]. It is suggested for radionuclides that can be taken-up by plants to have chemical behaviour similar to essential macro-elements which may compete with the radionuclides for the root uptake [7]. Radium and calcium analogy in plants was already reported by several authors [7- 8]. Accumulation of $^{226}$Ra in plants was
described in many studies for sites contaminated with U-mill tailings [9-10] and in laboratory-based experiments [11]. The members of Brassicaceae family tend to be highlighted particularly, as they were reported to tolerate and accumulate metals and radionuclides in higher quantities [12].

2 MATERIALS AND METHODS

2.1 Experimental design

The statistical ANOVA design was used as the experimental basis where three different soils (A, B and C; acidic-pH 4.4, alkaline-pH 7.2 and alkaline-pH 7, respectively) and four levels of soil contamination were applied in a two-factor design. Chinese cabbage was sown on contaminated and non-contaminated soil in 2L plastic pots. Plants were irradiated by fluorescent lamps with a daily light phase of 12 h. Contaminated soils were prepared by mixing a garden soil with UMT in three different ratios, representing different contamination scenarios. The non-contaminated soil was used as a control.

2.2 Sampling and sample preparation

Plants: plants of Chinese cabbage were harvested after 4 months of growth to obtain sufficient amount of the sample for the $^{226}$Ra measurements. After harvest, plants were separated on roots and leaves, carefully washed with a tap water, dried in an oven and left outside at a room temperature to reach a constant mass. Dried plant leaves were milled by a rotor speed mill and homogenized.

Soils: soil samples used in preparation of soil-tailings mixtures were taken from the local fields at the root zone (30 cm) in the vicinity of Ljubljana. Three different soils were chosen according to a different pH value, organic matter content and available K and P. The tailings samples were taken from the Boršt tailings pile of the former Žirovski vrh uranium mine [1]. Soil and tailings samples were air dried, sieved and mixed, representing 20%, 40% and 60% of UMT in the contaminated soil, respectively.

2.3 Determination of $^{226}$Ra in soil and plant samples

The direct gamma-ray spectrometry was used for the measurement of $^{226}$Ra in soil and cabbage leaves. The measurements of plant and soil samples were done from each pot separately in order to ensure appropriate statistical evaluation.

Plants: cylindrical polystyrene vials of 1.7 cm in diameter (inner) and 4.3 cm height (sample height) were filled with about 4-6 g of dried plant leaves, sealed hermetically using a glue gun and stored for 4 weeks to allow radioactive equilibrium of the $^{226}$Ra series to be established. The system was calibrated by a certified mixed-gamma source simulating vegetation containing $^{210}$Pb, $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{203}$Hg, $^{113}$Sn, $^{137}$Cs, $^{54}$Mn, $^{88}$Y, $^{65}$Zn and $^{60}$Co.

Soils: cylindrical polystyrene containers of 5.9 cm in diameter (inner) and 3.8 cm height (sample height) were filled with about 100-130 g of dry soil material, sealed hermetically using a glue gun and stored for 4 weeks to allow radioactive equilibrium of the $^{226}$Ra series to be established. The system was calibrated by a certified mixed-gamma source simulating soil containing $^{210}$Pb, $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{203}$Hg, $^{113}$Sn, $^{137}$Cs, $^{54}$Mn, $^{88}$Y, $^{65}$Zn and $^{60}$Co.
2.4 Determination of Ca in plants

A \( k_0 \)-neutron-activation analysis was used for the determination of Ca in plants [13].

2.5 Complementary determinations

Pedological parameters of soils were assayed in the Centre for Pedology of the Agronomy Department, Biotechnical faculty, University of Ljubljana.

2.6 Evaluation of \(^{226}\)Ra transfer to Chinese cabbage

In order to evaluate the transfer of radionuclides to plants comprising the uptake of radionuclides from soil and their subsequent translocation to above-ground tissues, a plant-to-soil radionuclide concentration ratio need to be determined. Concentrations ratio (CR) which describes the accumulation of radionuclides in the plants, is defined as a ratio of total activity concentration on a dry mass basis (Bq kg\(^{-1}\) d. m.) in the plant (harvestable) to that in the soil [10].

3 RESULTS AND DISCUSSION

3.1 \(^{226}\)Ra in soil

Table 1 shows \(^{226}\)Ra activity concentrations in soils contaminated with UMT in different ratios (20 %, 40 % and 60 % of UMT in the soils) and in the control soil.

<table>
<thead>
<tr>
<th>Soil</th>
<th>Control soil</th>
<th>20 % of UMT in the soil</th>
<th>40 % of UMT in the soil</th>
<th>60 % of UMT in the soil</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>A0: 51 ± 3</td>
<td>A20: 1726 ± 72</td>
<td>A40: 3441 ± 144</td>
<td>A60: 5065 ± 210</td>
</tr>
<tr>
<td>B</td>
<td>B0: 62 ± 3</td>
<td>B20: 1805 ± 74</td>
<td>B40: 3632 ± 151</td>
<td>B60: 5187 ± 211</td>
</tr>
<tr>
<td>C</td>
<td>C0: 103 ± 3</td>
<td>C20: 2091 ± 88</td>
<td>C40: 3873 ± 161</td>
<td>C60: 5346 ± 221</td>
</tr>
</tbody>
</table>

3.2 \(^{226}\)Ra in plants

3.2.1 Evaluation of the uptake

Activity concentrations of \(^{226}\)Ra in Chinese cabbage (Fig. 1) ranged from 140 ± 8 to 877 ± 85 Bq kg\(^{-1}\) dry mass for 20 % and 60 % of UMT in the soil, respectively. \(^{226}\)Ra measured in the cabbage was increasing with its total concentration in the contaminated soil. According to \(^{226}\)Ra and Ca similar chemical behaviour, an approach of \(^{226}\)Ra uptake evaluation based on Ca uptake might be applied, as reported in the literature for \(^{226}\)Ra and Ca analogue relation in the plants [7-8]. Ca exchange capacity in the soil (Table 2) and cabbages’ shoot concentrations (Fig. 2) were increasing with UMT content in the contaminated soils; which means that cation exchange capacity (CEC) may be one of the reasons for \(^{226}\)Ra transfer to Chinese cabbage.
despite of its non-essential role in the plants. Elevated $^{226}$Ra and Ca concentrations in tested plants indicated that uptake process may be similar as the uptake of other $^{226}$Ra analogues, such as Sr and Ba [14]. The highest $^{226}$Ra bioaccumulation was observed in plants from soil A which might be attributed to an acidic pH value and lower content of available P in this soil and consequently higher $^{226}$Ra activity concentrations in these plants due to lower biomass produced, compared to soil B and C, respectively. However, further investigations are required to clarify this phenomenon.

![Figure 1: $^{226}$Ra activity concentrations in Chinese cabbage for different levels of soil contamination (error bars correspond to the standard combined uncertainties, k = 2)](image)

Table 2: UMT content [%] and exchangeable Ca [mmol/100 g] for control and contaminated soils used in the pot experiments

<table>
<thead>
<tr>
<th>UMT in soil</th>
<th>Exchangeable Ca</th>
</tr>
</thead>
<tbody>
<tr>
<td>A0 (control)</td>
<td>6.46</td>
</tr>
<tr>
<td>A20 (20 % of UMT in the soil)</td>
<td>28.71</td>
</tr>
<tr>
<td>A40 (40 % of UMT in the soil)</td>
<td>46.43</td>
</tr>
<tr>
<td>A60 (60 % of UMT in the soil)</td>
<td>68.23</td>
</tr>
<tr>
<td>B0 (control)</td>
<td>21.85</td>
</tr>
<tr>
<td>B20 (20 % of UMT in the soil)</td>
<td>34.90</td>
</tr>
<tr>
<td>B40 (40 % of UMT in the soil)</td>
<td>50.77</td>
</tr>
<tr>
<td>B60 (60 % of UMT in the soil)</td>
<td>66.79</td>
</tr>
<tr>
<td>C0 (control)</td>
<td>21.70</td>
</tr>
<tr>
<td>C20 (20 % of UMT in the soil)</td>
<td>38.77</td>
</tr>
<tr>
<td>C40 (40 % of UMT in the soil)</td>
<td>52.22</td>
</tr>
<tr>
<td>C60 (60 % of UMT in the soil)</td>
<td>68.86</td>
</tr>
</tbody>
</table>
3.2.2 Evaluation of concentration ratios

$^{226}$Ra CR values ranged from 0.06 to 0.17 for 20% and 60% of UMT in the soil, respectively (Fig. 3). The values are in agreement with the results of other studies for $^{226}$Ra transfer to plants growing on a soil contaminated with U-mill tailings [11, 15]. The results indicate that a caution is needed when growing Brassica crops at the mine areas potentially contaminated by uranium due to possible transfer of $^{226}$Ra to food chain and the related dose absorbed by the nearby living inhabitants.

3.2.3 Phytoremediation potential

Extraction of radionuclides from the contaminated soil by plants, called phytoextraction, is the most often used phytoremediation technique. It depends mainly on the level of soil contamination, bioavailability of radionuclides contained in the soil, efficient root uptake and xylem translocation to the shoots. The phytoextraction potential [16] of Chinese cabbage for the extraction of $^{226}$Ra from soil was calculated as follows;
PEP = (a(plant) x M(plant) / a(soil) x M(soil)) x 100

PEP - phytoextraction potential [%]

a(plant) - $^{226}$Ra activity concentration in the harvested biomass [Bq kg$^{-1}$ dry mass]
M(plant) - the mass of the harvestable biomass produced in one harvest [kg]
a(soil) - $^{226}$Ra activity concentration in the soil volume [Bq kg$^{-1}$]
M(soil) - the mass of the soil volume in the root zone [kg]

The calculation was done on the basis of assumptions for plant $^{226}$Ra activity concentrations of 2000, 3500 and 5000 Bq kg$^{-1}$ dry mass for 20%, 40% and 60% of UMT in the soils, respectively, 4.2 tons/ha of Chinese cabbage dry biomass produced in one harvest and 5250 tons/ha of contaminated soil in the root zone. The calculations showed that 0.003%, 0.005% and 0.007 % of soil $^{226}$Ra was extracted into shoots in a single cycle for 20%, 40% and 60 % of UMT in the soil, respectively. The time needed to reach the target clean-up level of 200 Bq kg$^{-1}$ dry mass of residual $^{226}$Ra in the soil [11] would last as long as time needed for $^{226}$Ra to decay, which means the phytoextraction to be inefficient. The capability of Chinese cabbage to extract $^{226}$Ra from soil to shoots was too low. Realistic scenario should be implemented by more advanced and sophisticated approach to enhance $^{226}$Ra bioavailability from soil to plants and/or applying other Chinese cabbage cultivars or plant species with $^{226}$Ra hyper-accumulation capacity. We calculated that an ideal plant species that would be suitable for a commercial or economically viable phytoextraction of $^{226}$Ra from soils contaminated with U-mill tailings should accumulate $^{226}$Ra in the shoots at least 10 times of soil $^{226}$Ra concentration and have the characteristics of 20 tons/ha of dry biomass production. In this case, 3.8% of soil $^{226}$Ra would be extracted into shoots and the time needed to establish the target clean-up level would last 25 years. It was suggested that also plants with a lack of metal hyper-accumulation characteristics may be used for $^{226}$Ra phytoextraction if they have the preference for high Ca acquisition like calcicole plants [17]. Ca channels might be suitable for $^{226}$Ra uptake as reported for other $^{226}$Ra analogues; like the uptake of Sr and Ba by plants growing on Sr and Ba enriched solution [14].

4 CONCLUSIONS

Chinese cabbage was showed to be an appropriate plant species for testing $^{226}$Ra transfer from soil to plant. The pot experiment in laboratory-based conditions results as an effective way of growing under conditions controlled as far as possible. The uptake of Ca by plants was applied for assessing the $^{226}$Ra uptake evaluation due to their analogue behaviour. Bioaccumulation of $^{226}$Ra in tested plants was found to differ among the tested soils and was the highest in plants grown in soil A. $^{226}$Ra CR values ranged from 0.06 to 0.17 for 20 % and 60 % of UMT content in the soil, respectively, and were comparable to the literature data for sites contaminated with U-mill tailings. Phytoremediation of $^{226}$Ra from UMT-contaminated soils was ineffective due to low cabbage phytoextraction potential of 0.003 %, 0.005% and 0.007 % for 20 %, 40% and 60 % of UMT content in the soil, respectively. A more advanced and sophisticated phytoremediation techniques should be applied in order to use the Chinese
cabbage for phytoextraction of $^{226}\text{Ra}$ from soils contaminated with U-mill tailings. Alternatively, the application of other Chinese cabbage cultivars or plant species with $^{226}\text{Ra}$ hyper-accumulation capacity is recommended as a more effective approach. Despite the cabbage low phytoremediation potential, the $^{226}\text{Ra}$ bioaccumulation in the leaves indicates that caution is needed when growing Brassica crops at the uranium mine areas, to prevent transfer of $^{226}\text{Ra}$ to food chain and the related potential dose exposure to the nearby living inhabitants.

ACKNOWLEDGMENTS

This work was supported by the Rudnik Žirovski vrh company. Assistance of the Slovenian Research Agency (contract no. P2-0075) is also acknowledged.

REFERENCES


