Comparison and Cultural Review of Biosphere Modeling Using Foliar Uptake of Technetium-99 in Radishes

by

Kayla L. Pierson

A THESIS

submitted to

Oregon State University

University Honors College

in partial fulfillment of the requirements for
the degree of

Honors Baccalaureate of Science in Chemical Engineering (Honors Associate)

Honors Baccalaureate of Arts in International Studies in Chemical Engineering

Presented May 13, 2011
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AN ABSTRACT OF THE THESIS OF

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Abstract approved:

_____

Kathryn Higley

Abstract

Biosphere models predict the transport of radionuclides in the environment with an end goal of assessing the possible risk to humans. Little data exists to support these mathematical biosphere models. The objectives of this work were to verify foliar uptake in biosphere models using experimental data, compare existing biosphere models based on cultural differences, and contributing new values to the literature. Radishes were grown and their leaves were contaminated by a single deposition of technetium-99 to assess the amount of radionuclides transferred to the roots. Liquid scintillation counting was used to determine the radioactivity of the samples at harvest. The radish roots had concentrations between 0 and 500 Bq/g which was on the same order of magnitude as the ANDRA and EPRI model predictions. The Studsvik model prediction was three orders of magnitude higher than the experimental values and was not confirmed. The EPRI, ANDRA, JGC and Studsvik models were compared and contrasted based on location, diet, and construction which led to differences in dose predictions of 0-70 Sv/y.

Key Words: biosphere modeling, technetium, radish, radiation

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I understand that my thesis will become part of the collection of Oregon State University. My signature below authorizes release of my thesis to any reader upon request. I also affirm that the work represented in this thesis is my own work.

Kayla L. Pierson, Author

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INTRODUCTION

Understanding how radioactive contaminants affect the environment is important for the responsible use of nuclear power and the long term storage of nuclear waste. Significant research and analysis has been, and continues to be, completed to verify the safety of radionuclides in the nuclear fuel cycle and to understand the transport of radionuclides upon an unexpected release from any source in the nuclear fuel cycle (1). Storage of nuclear waste is essential for use of nuclear fuel, and a main storage method being looked into by many counties is high level waste repositories (HLWRs) (2). HLWRs are deep geologic underground storage facilities that protect radioactive waste with stable geology and engineered barriers (3).

Performance assessment models exist for HLWRs to determine their effectiveness and verify the safety of the repository (4). The performance assessment includes many stages and models to predict radionuclide transport (5). Infiltration, percolation and seepage predict water movement into and out of the repository barriers. Near-field transport predicts the breakdown of the engineered barriers and the release of radionuclides. Geosphere models predict transport in geologic media, separated into the unsaturated and saturated (groundwater) zones. (6) The saturated zone is where radionuclides enter groundwater and surface water, becoming a potential dose to humans. The biosphere modeling predicts radionuclide transport from this point on (7).

Biosphere modeling predicts the human dose caused by the transport of radioactive contaminants in the environment due to a release of radionuclides (1). Biosphere modeling encompasses all modes of radionuclide travel that could lead to human uptake, such as travel in groundwater and surface water, irrigation and crop uptake, animal uptake, air transport, and storage in soils (6). One of the main goals of biosphere modeling is to understand how humans would be affected by a release from an HLWR (8). One component of biosphere modeling assesses the dose to humans due to ingestion of contaminated crops. Crops can be contaminated from soil, soil uptake, or from the leaves, foliar uptake.

Foliar uptake is the fraction of radioactive material that is deposited on plant leaves, absorbed into the leaves and transported to the edible portion of the plant. The radionuclides are assumed to be from contaminated irrigation sources, such as surface water or groundwater. The radionuclides are absorbed into the plants based on a number of factors, including the leaf size, the type of plant (grain, fruit, root vegetable, or leafy green) and the radionuclide. Portions of the radioactive material are then transported from the leaves to the edible part of the plant. The edible portion varies based on plant type as can be expected. For example, fruits and leafy greens where the edible part of the plant is exposed to irrigation would have the edible portion aboveground; whereas, a root vegetable has the edible portion underground, completely separate from the leaves. A basic example of a foliar uptake model is given in Equation 1:

$$C_{plant} = \frac{C_w V_{irr} F_i F_t}{Y} \qquad \text{Eqn. 1}$$

where:

C_{plant} is the radionuclide concentration in the edible portion of the plant, Bq/kg fresh weight,

C_w is the radionuclide concentration in the irrigation water, Bq/L,

V_{irr} is the volume of irrigation water, L/m^2/y,

F_i is the fraction interception, which outlines what portion of the irrigation water is deposited on the leaves of the plant,

F_t is the fraction translocation, which outlines what fraction of the radionuclide that is absorbed into the plant tissue will travel to the edible portion of the plant, and

Y is the crop yield, kg fresh weight/m^2/y.

Additional terms can be added to Equation 1 to account for other processes. A food processing term can be included in the numerator similar to F_i and F_t which accounts for washing and cooking of food. A weathering, or wash-off, term can be included in the denominator to account for rain, wind and other natural phenomena that remove absorbed radionuclides from the leaf surface over time.

Much time and effort has been put into modeling contaminant transport, but insufficient time has been put into validating the modeling (9). This is understandable as modeling can be done for all different plant types and elements; whereas, confirming the model means taking the time to grow all different types of plants, contaminating them with numerous different radioisotopes, and verifying the numerous factors and coefficients

that make up the models. This paper focuses specifically on a small portion of model confirmation.

Radish leaves were contaminated at one point during their lifetime to determine the amount of foliar uptake and the transport of the contaminant to the edible portion of the radish. The results were compared to three different concentration ratio biosphere models: Electric Power Research Institute's Yucca Mountain Model (EPRI), the Swedish Nuclear Fuel and Waste Management Company's Studsvik Model, and France's ANDRA model. All three models are concentration ratio models, which assume only the concentration of the radioactive element in the water is important in determining the concentration in the plant. Specific activity models, which look at the amount of the radioactive element in the soil, use the ratio of the stable isotopes in the soil and the plant to determine the partitioning of the radioactive isotope in the plant (10), were not assessed in this paper because no radioactive elements were added to the soil.

EPRI, Studsvik, ANDRA, and an additional fourth model, Japan's JGC model, are compared side by side for cultural and structural differences. With these choices, a broad range of ecosystems, agricultural production, and cultural values are presented. The diet of each model was used in all of the models to determine and compare the yearly dosage to a human due to contamination from foliar uptake. This means that the JGC, and all other diets, were put into the JGC, ANDRA, EPRI and Studsvik models to compare how each model works with each different diet. A number of cultural practices and model structures explain the differences in the models and their results.

The four foliar uptake terms of the biosphere models are presented below. The ANDRA model is described in Equation 2 (9):

$$C_{plant} = C_{irr} T_{irr} \left[\frac{F_1 F_2}{\lambda_w Y}\right] \quad \text{Eqn. 2}$$

where:

C_{plant} is the concentration of radionuclide in the plant, Bq/kg fresh weight,

C_{irr} is the concentration of radionuclide in the irrigation in the water, Bq/L,

T_{irr} is the amount of irrigation water applied to the plant, L/m^2/y,

F_1 is the fraction interception,

F_2 is the fraction translocation,

Y is the yield, kg fresh weight/m^2, and

λ_w is the wash-off, 1/y.

The EPRI model is described in Equation 3 (6):

$$C_{plant} = I_{crop} V_{irr} C_w \left[\frac{F_{abs} F_{p2} F_{trans}}{Y}\right] \quad \text{Eqn. 3}$$

where:

C_{plant} is the radionuclide concentration in the edible part of the plant, mol/kg fresh weight

I_{crop} is the fraction interception,

V_{irr} is the rate of irrigation water, m^3/m^2/y,

C_w is the radionuclide concentration in the irrigation water, mol/m^3,

F_{abs} is the fraction of intercepted water that is absorbed from the external surface into the plant tissue,

F_{p2} is the fraction of internal contamination that remains after food processing,

F_{trans} is the translocation fraction, and

Y is the yield, kg fresh weight/m^2/y.

The Studsvik model is described in Equation 4 (11):

$$C_p = N_{irr} T_L \frac{LAI * StoCap * K_{ret} * C_w}{Y_P} \quad \text{Eqn. 4}$$

where:

C_p is the radionuclide concentration in the plant, Bq/kg,

N_{irr} is the number of irrigation events,

T_L is the translocation factor,

LAI is the leaf area index, m^2/m^2,

$StoCap$ is the water storage capacity of the leaves, m^3/m^2,

K_{ret} is the element dependent retention factor,

C_w is the radionuclide concentration in the irrigation water, Bq/m^3, and

Y_p is the yield, kg/m^2.

The JGC model is shown in Equation 5 (5):

$$C_{crop\_irr} = \mu_{crop}\left(d_{crop} C_{rw}\right)\left[\frac{(1-F_{crop})+F_{trans}}{Y_{crop} W_{crop}}\right] \quad \text{Eqn. 5}$$

where:

C_{crop_irr} is the radionuclide concentration in the crop, Bq/kg,

μ_{crop} is the interception fraction for the irrigation water on the crop,

d_{crop} is the depth of irrigation water applied to the crop, m/y,

C_{rw} is the radionuclide concentration in the river water, Bq/m^3,

F_{crop} is the fraction of external contamination on the crop lost due to food processing,

F_{trans} is the translocation fraction,

Y_{crop} is the yield, kg fresh weight/m^2, and

W_{crop} is weathering rate, 1/y.

The values from each foliar uptake model are put into a dose equation to determine the human dose. A standard dose equation for ingestion due to crops is shown in Equation 6 (5):

$$D_{crop} = DC_{ing} ING_{crop} C_{crop} \quad \text{Eqn. 6}$$

where:

D_{crop} is the individual dose by ingestion of a crop, Sv/y,

DC_{ing} is the dose coefficient for each radionuclide, Sv/Bq,

ING_{crop} is the ingestion rate for the crop, kg/y, and

C_{crop}, is the radionuclide concentration in the plant, Bq/kg fresh weight.

Multiple dose equations from a variety of sources, including external exposure, inhalation, animal product uptake, and water, are combined to determine the total dose to a critical population (5).

The critical population is the group of people most at risk – the group who would receive the highest dose. This group is determined by each model, and is a country, and sometimes age, specific group. Most likely this group lives near the high level waste repository storing nuclear waste. The diet of this group was analyzed for each model.

Experiments were performed using technetium-99 (Tc-99) as the contaminant. Technetium-99 is a major fission product of nuclear waste, and its environmental transport is little understood. It is beta emitter, as seen in Equation 7, with a half-life on the order of 200,000 years. Although Tc-99 is a beta emitter, its long half-life implies that it persists in the environment for an extensive time. In addition, it does not bind well to soils, and it is very mobile in the environment. Technetium-99 is good for lab work and health concerns because it is a low energy (0.10 MeV) emitter. (12)

$$^{99}Tc \rightarrow {}^{99}Ru + \beta \qquad \text{Eqn. 7}$$

METHODS

The experimental data was collected by contaminating radish leaves with water containing Tc-99. Treatments per plant of 5,000 and 10,000 Bq were chosen because they had high enough concentrations that they would be detected easily by the liquid scintillation counter with an expected 1-10% translocation, but they were low enough to not be extremely harmful to the workers. It was determined that 0.2 mL was a manageable amount of water to put on the leaves without having water fall off. Water concentrations had to be 25,000 and 50,000 Bq/mL to account for the volume and treatment required. Linear concentrations were chosen to determine whether the translocation occurred linearly or not.

The radishes were contaminated only once in their lifetime in order to look at one specific event. At each contamination event, there were five replicates of each of the two Tc-99 concentrations. With an additional 5 control radishes receiving no contamination but being grown under the same conditions, a total of 45 radishes were grown.

Radishes were given one of two treatments 5,000 Bq/plant or 10,000 Bq/plant. To deliver this treatment, contaminated water was placed onto the radish's secondary leaves with a pipette. The total water amount was 0.2 mL with 0.1 mL being placed on two separate leaves. The solutions placed on the leaves were 25,000 Bq/mL, stock D, for the 5,000 Bq/plant treatment and 50,000 Bq/mL, stock C, for the 10,000 Bq/plant treatment. These solutions were prepared by diluting a small sample of Tc-99 contaminated water. The

original solution, called stock A, was at a concentration of $1.1x10^6$ Bq/mL. It was then diluted down, called stock B, to obtain a more accurate reading of its radioactivity, at which point it was $2.00x10^5$ Bq/mL. This solution was then used to make the $2.5x10^4$ and $5.0x10^4$ Bq/mL solutions used on the radishes.

The radishes were planted on September 23, 2010 in 4" square pots. Two radish seeds were planted in each pot, and at day 5 the less healthy looking seed was removed. Radishes were grown in a greenhouse approved for use with radioactive materials. The first treatment of radiation was given on day 16, and subsequent treatments were given every 4-6 days. All treatments were performed on the secondary leaves for consistence; therefore, the secondary leaves had to develop before dosing could begin. A pipette was used to remove 0.1 mL of stock solution and the solution was carefully placed on each leaf, as seen in Figure 1. The small beads of water rested on the leaves; however, occasionally some water fell off the leaves. This error was noted in a note book and those plants were removed from the data set. The removed plants would have low radioactivity in the leaves and high radioactivity in the roots due to soil uptake. Soil uptake would also change the uptake pathway from all other plant samples and would give erroneous results. Table 1 shows the days on which contamination occurred.

Lights in the greenhouse were used for a portion of the growing period due to the fall season and lack of full summer sunshine. Lights were turned on in the evening from 7-9pm after day 19.

Figure 1: This figure shows the drops of contaminated water on the surfaces of the leaves. This figure also shows the primary and secondary leaves.

Once all contamination events were completed and the radishes had matured, they were harvested. The harvest took place on October 24, 2010, day 32 as seen in Table 1. Upon harvesting, the radishes were removed from their pots, gently rubbed to remove excess dirt, and cut at the base of the leaves. Small root tailings were left in the pot. Figure 2a shows the radishes on harvest day. After cutting the leaves and roots were treated as separate entities, although they were recognized as coming from the same plant. The leaves and roots were weighed and then put in a dryer at 65°C for at least 48 hours, as shown in Figure 2b. Not all the radishes were crushed on the same day, so radishes waiting to be crushed were left in the dryer. When the plants were dry they were weighed again.

The radishes were crushed and put into an acid digest to prepare the samples for liquid scintillation counting (LSC). Dried radishes roots were dipped in liquid nitrogen. Then the radishes leaves and roots were separately crushed and hammered in a pestle and

mortar until they were fine particles (<5 mm long and <1mm wide). About a 100 mg sample of each radish part was measured out in a vial to prepare for acid digestion.

Table 1: This table shows the days on which planting, watering, dosing, and harvesting of the radishes occurred.

Day	1	2	3	4	5	6	7	8	9	10	11
Event	Plant	Water			Water		Water		Water		
Day	12	13	14	15	16	17	18	19	20	21	22
Event	Water		Water		Dose/ Water			Water/ Lights	Dose	Water	
Day	23	24	25	26	27	28	29	30	31	32	
Event	Water			Dose/ Water			Water		Dose/ Water	Harvest	

(a) ***(b)***

Figure 2: (a) This picture shows a radish on harvest day. ***(b)*** This picture shows the root and leaves separated and ready for drying.

The acid digestion procedures are listed in Thompson, 1998 (13). The acid digestion consisted of a mixture of 0.3 mL nitric acid and 0.3 mL perchloric acid. The vials were placed in a water bath at 56-59°C for 2 hours after the acid was added to the vials. The vials were then allowed to cool for 24 hours and then left to sit for 5 more days. After the 6th day, 15 mL of Hionic-Flour, the LSC fluid, was added to the vials. The vials were run

in the LSC a week later. Three weeks after the first LSC reading the samples were run on the LSC again; however, this time the samples were shaken before being put in the LSC.

Liquid scintillation counting is a technique that detects beta particles using fluorescence (14). Aqueous samples are mixed with an aqueous LSC fluid that contains an organic solvent and fluors (14). When the sample decays and emits a beta particle, the energy interacts with the solvent, causing the molecules to become excited (14). The excitation is transferred from the solvent to the fluors, and the electrons in the fluors move to higher energy levels (14). When the electrons move back to their ground state the fluors fluoresce, while two photomultiplier tubes count the light given off by the fluorescence (14). The magnitude of the light is proportional to the radioactivity of the sample (14).

Two main interferences cause error in LSC; chemical quenching and color quenching (14). Chemical quenching occurs when the energy of the beta particle is transferred to the sample instead of to the solvent; this energy is lost and cannot be counted (14). Color quenching occurs when the wavelength of the fluoresced light is the same wavelength as the sample (14). The fluoresced light is absorbed by the sample and is not counted by the photomultiplier tubes (14).

The radish samples were analyzed in the LSC for 10 minutes, and an average of the activity for one minute was given. This value was converted to seconds to give the activity of the sample in Bq. A blank sample of 0.1 mL DI water and a vial containing

10μL of stock A were also run. These additional samples also contained 15 mL of Hionic-Fluor.

Four matrix spikes were run with the samples to determine the efficiency of the LSC in reading the samples. A compilation of all of the control plants was used to make two LSC vials of root and leaf blanks each. A known amount of radioactivity (5,000 Bq) was put into the LSC vial with the plant material, and the vials were put into the dryer. When all water had evaporated from the vials, the vials were removed from the dryer and treated as all other samples, including the acid digestion and adding the Hionic-Fluor.

RESULTS

The radioactivity in the plants of the matrix spike samples did not get treated in exactly the same way as all other samples. Radionuclides in the experimental samples were contained within the plant tissue, having traveled from the surface through the plant; whereas, the radionuclides in the matrix spike samples were applied to the exterior the crushed radishes. It would be expected that radioactivity remained on the surface of the spike samples and was more easily vaporized during the acid digestion.

Only the first set of LSC readings were used because all vials had the same amount of settling. The second set of readings were shaken, but due to the number of samples and the time for each LSC reading, the first vials read would have less settling of particles than the last vials and there could have been some bias in the measurements. The LSC readings were corrected with an efficiency term calculated by the matrix spikes. The efficiency was a simple calculation as the ratio of the known concentration of the spike, C_{known}, divided by the LSC reading on the spike, C_{LSC}, as seen in Equation 8. There was no significant difference between the root and leaf efficiencies, so an average of all four samples was used. The efficiency calculated was 1.75, which makes sense as lower energy beta emitters, like Tc-99, are subject to high quenching (14). Every reading taken by the LSC was multiplied by the efficiency to account for errors in the LSC readings. One major source of error was failure to extract all the radionuclides from the radishes during the acid digestion as described in the above. Chemical quenching also occurred as

there were large radish particles not fully dissolved in the acid digest sitting at the bottom of the vials. Color quenching added a small error as the sample solutions were brownish.

$$Efficiency = \frac{C_{known}}{C_{LSC}} \quad \text{Eqn. 8}$$

Results are given in groups of the dosage day, dosage amount and plant type. The list of the groups is shown in Table 2. The first letter represents the dosage day: *A* is for the first day, *B* is for the second, *C* is for the third, and *D* is for the fourth. The group number represents the dosage amount, *5* for 5,000 Bq/plant, and *10* for 10,000 Bq/plant. The last letter represents the plant type, *r* for root and *l* for leaf. If the group name does not have a second letter that means that it represents the whole plant, the root and leaves combined. Two outliers were found in the data using Dixon's Q-test with 90% confidence. However, these samples were included in the analysis due to the complex and varied nature of biological systems. Many more factors are at play in biological systems than in simple measurements and there is important variability in the biological world. However, as mentioned previously, the plants which had problems in the dosing were removed from the groups. The number of plants in each group is also listed in Table 2.

A mass balance on each plant was performed in order to determine the total amount of radioactivity that could be accounted for. This percent of radioactivity recovered was determined using Equation 9. The percent recovered is *PR*, C_{root} is the total of radioactivity in the root of the plant, C_{leaves} is the total amount of radioactivity in the leaves of the plant, and C_{dose} is the total dose given to the plant. The percent recovered

values were grouped by dosage day and amount. Figure 3 displays the percent recovered for each group.

Table 2: This table shows the groups for the data analysis.

Group Name	Dosage Group	Dosage Day	Dosage Amount (Bq/plant)	Plant Type	Plants in Group
A-10-l	First	16	10,000	Leaf	5
A-10-r	First	16	10,000	Root	5
B-10-l	Second	20	10,000	Leaf	4
B-10-r	Second	20	10,000	Root	4
C-10-l	Third	26	10,000	Leaf	4
C-10-r	Third	26	10,000	Root	4
D-10-l	Fourth	31	10,000	Leaf	5
D-10-r	Fourth	31	10,000	Root	5
A-5-l	First	16	5,000	Leaf	4
A-5-r	First	16	5,000	Root	4
B-5-l	Second	20	5,000	Leaf	4
B-5-r	Second	20	5,000	Root	4
C-5-l	Third	26	5,000	Leaf	4
C-5-r	Third	26	5,000	Root	4
D-5-l	Fourth	31	5,000	Leaf	4
D-5-r	Fourth	31	5,000	Root	4

$$PR = \frac{C_{root}+C_{leaves}}{C_{dose}} * 100 \qquad \text{Eqn. 9}$$

The percent recovery was around 60-80% with 70% being the average. It is possible that the radishes were not crushed small enough and that some radioactivity is still contained within the plant tissue. This would primarily be a problem with the radish root, as it was much more challenging to crush than the leaves which fell apart quite easily. Fairly large pieces of radish root remained in the LSC vials after acid digestion, and it is possible they still contain some level of radioactivity that is not accounted for in the measurements. In addition, these radish pieces could have affected the LSC's ability to take measurements.

This should be accounted for with the matrix spikes, as they were subjected to the same procedure as all other plants, with the exceptions mentioned earlier. Another source of error could be the small radish roots that were left in the soil and not recovered. This error is expected to be small compared to other sources.

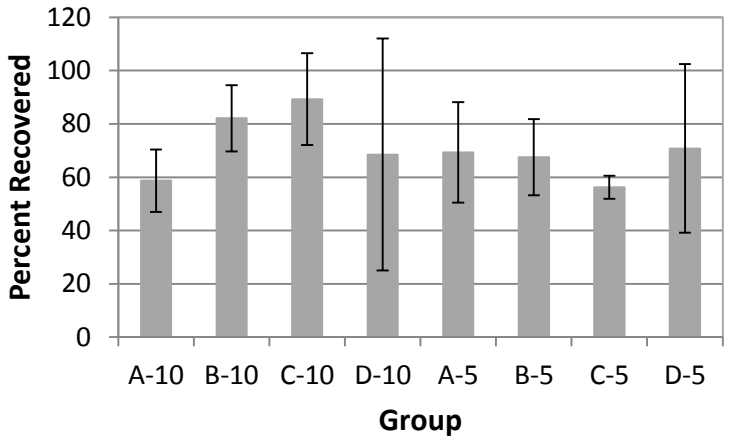


Figure 3: Percent Recovery. The average percent recovery of each group is shown. The error bars represent one standard deviation from the mean.

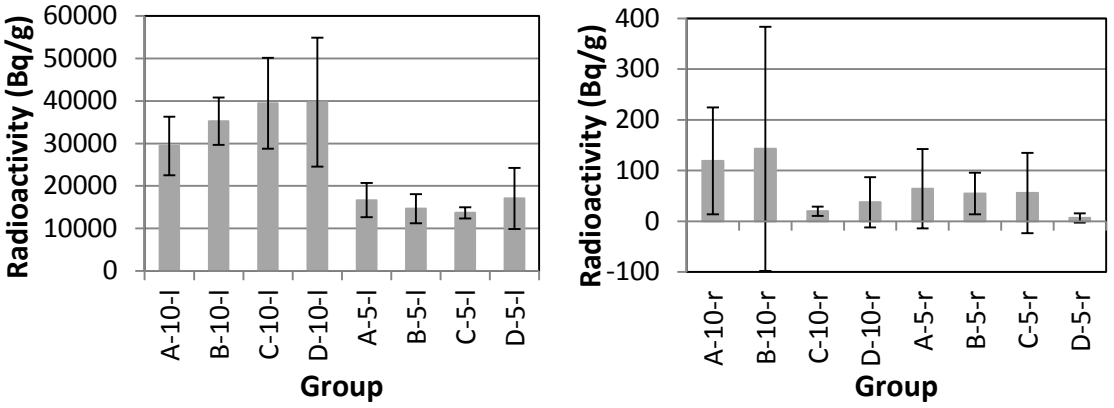
It is interesting to note that the plants with a 10,000 Bq dose had a higher percent recovery than the plants with a 5,000 Bq dose. One possible explanation is weathering, the removal of surface contamination by wind or water. This would make sense in a natural environment, but not in a greenhouse. However, on a couple of days condensation formed on the root of the greenhouse and fell on the radishes. This condensation was more of a problem for the 5,000 Bq plants due to their location and the location of the condensation. The last dosage, group D, was not fully absorbed at the time of harvest, and some of that dosage could have weathered off. Weathering in the form of condensation could have occurred on other days leading to reduced percent recovery.

Other experimental error could come from making the stock solutions and pipetting out the stock solutions onto the radish leaves. The small root tailings left in the pots and acid vapor formed during the acid digestion could also contain small amounts of radioactivity that were not accounted for.

The results of the experiment are shown in Figure 4 with the values normalized to Bq/g. The leaf averages are shown in *(a)* while the root averages are shown in *(b)*. The error bars on each group represent one standard deviation from the average value. Radioactivity in the leaf samples increased as the dosage day progressed farther into the plant's lifetime, while the reverse happened for the roots. This makes sense as the longer the radioactivity is on the plant, the more likely it is to travel from the leaves to the root as the plant uses water from the leaves. One interesting point to note is that the concentrations in the 10,000 Bq/plant were not necessarily linearly proportional to the concentrations in the 5,000 Bq/plant measurements.

Predicted values from the three models (ANDRA, EPRI and Studsvik) were calculated in order to compare the results obtained from the experiment. First, values were calculated using the parameters outlined in the models and changing the concentration to 25,000 and 50,000 Bq/mL, as seen in Figure 5. Next, values were calculated to simulate the experiment as shown in Figure 6. The models assume spray irrigation with all water contaminated by radionuclides. The experiment had only one dose of irrigation water on the leaves containing radionuclides. To account for the differences in irrigation the fraction interception was changed to 1 to represent that all the irrigation water remained

on the leaves, as seen in Figure 1. The irrigation water volume was determined by using 0.2 mL of contaminated water and dividing by the pot area. Values for the model simulations can be found in Appendix A.



(a) ***(b)***

Figure 4: These figures show the average values of the radioactivity per gram in each plant group with the leaf averages shown in ***(a)*** and the root averages shown in ***(b)***. The error bars represent one standard deviation from the mean.

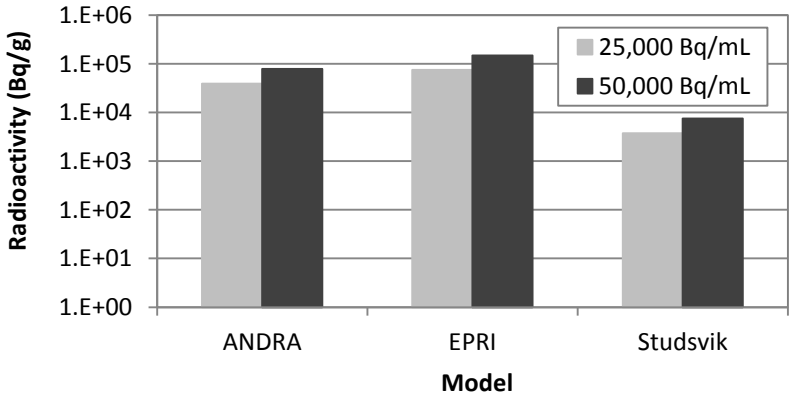


Figure 5: This figure shows the predicted concentrations of radionuclides in plants based on the three different models using the standard model values. The 25,000 Bq/mL are light grey and the 50,000 Bq/mL are dark grey. Note that the graph is in log scale.

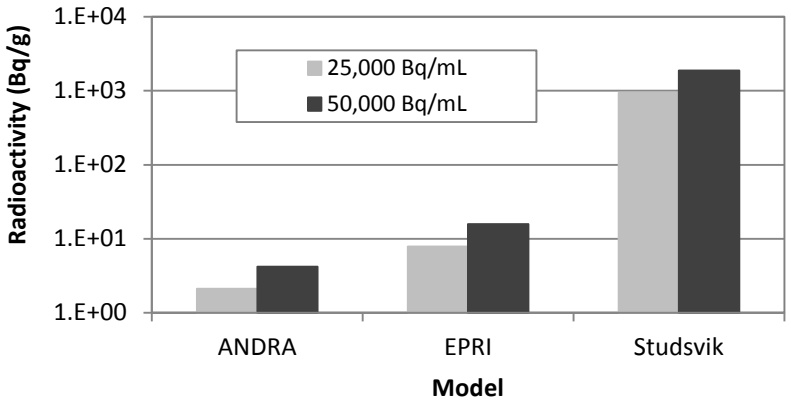


Figure 6: This figure shows the predicted concentrations of radionuclides in plants based on the three different models using the experimental model values. The 25,000 Bq/mL are light grey and the 50,000 Bq/mL are dark grey. Note that the graph is in log scale

The translocation factor is an important term in the models. Average values of the percent translocation by group are shown in Figure 7, where the error bars represent one standard deviation. The percent translocation is 100 times the translocation factor, F_t. The translocation factor for root vegetables in the EPRI, ANDRA and Studsvik models was 0.1, or a 10% translocation. Figure 7 shows that the average percent translocation is around 1%, one tenth of the value used in the models. The highest percent translocation of any radish was 2.0%, which is still lower than the model predictions.

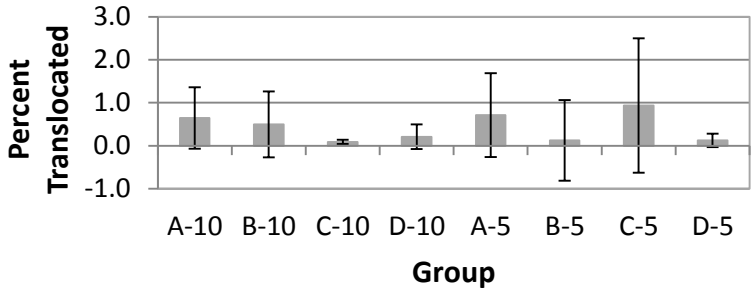


Figure 7: The percent translocation, 100 times the translocation factor, is shown. Values of 10% translocation are used in the models, but experimental values are lower than that.

DISCUSSION

Some plant types have different development stages, such as germination, developing into a fruiting body, and then the growth of the edible plant part. These stages could influence the effect of translocation in the plant due to different growth and nutrient requirements by the plant. Radishes were determined not to have any large development stages beyond the first set of leaves and the second set of leaves, so dosage days were not determined by plant growth. Days were chosen based on convenience. However, it is still interesting to comment on the effect of the dosage day. The first dosage had very noticeable changes in the plant. There were white spots on the leaves in the specific areas where water had been placed. The plants looked stunted in their growth compared to the other plants, as seen in Figure 8. The photo in Figure 8 was taken on day 22 between the second and third dosing. The plants in the back row were contaminated in group A while the plants in the next row were contaminated in group B. The plants in the front row were not contaminated. The stunted growth is most apparent in group A and is also visible in group B when compared with the uncontaminated groups.

In response to the stunted growth, a calculation of the average radish mass by group was performed, as seen in Figure 9. The control group is shown and error bars represent one standard deviation. The control group has masses that are similar to groups A-10 and B-5, but masses that are smaller than every other group. This implies that the contamination of Tc-99 did not stunt the radish growth compared to the control group. However, there is

still a positive correlation between mass and dosage day. This correlation may indicate an increased radish mass due to Tc-99 contamination.

Figure 8: This picture shows the white spots on the leaves of the first dosage group and the stunted growth compared to the other plants.

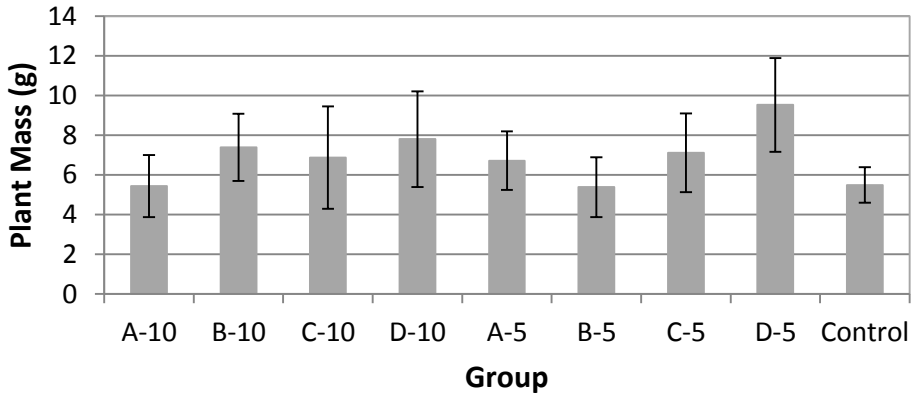


Figure 9: The average radish mass per group is shown with error bars representing one standard deviation. Positive correlation can be seen between dose day and plant mass, indicating that the early dose days had an adverse effect on plant mass. However, the control group masses were comparable to groups A-10 and B-5, but were smaller than all groups, possibly indicating an augmented growth with later dosages.

The translocation factor used in the models for root vegetables is 0.1 (6), (9), (11), but the highest translocation factor determined from experimental data was 0.02. The models are being conservative in their estimate of translocation compared to radishes. However, due to the grouping of many plant species this hedging makes sense. It is possible that other root vegetables have higher translocation factors. For radishes a recommended translocation factor would be 0.05 based on data collected in this study. This value is 1.5 times the highest translocation factor given from the data, allowing for a healthy safety margin.

The weathering, or wash-off, term can be described by the percent recovery, as seen in Figure 3. Experimental results show percent recovery around 70%, which corresponds to a weathering factor of 0.7. The ANDRA model has a weathering term, λ_w, 15 in the denominator (0.067 in the numerator) for all plant types (9). EPRI does not have a weathering term, but does include the fraction absorption, F_{abs}, which is 0.5 for root vegetables (6). Studsvik does not have a weathering or fraction absorption term. The ANDRA weathering term is much lower than the experimental data; whereas, the EPRI absorption term is a little smaller than the experimental value. These lower values in the models are for plants in outdoor conditions where they are subjected to more weathering conditions than in a greenhouse where the experimental plants were grown. The more conservative values are a safe estimate.

The data in Figure 5 agrees with data presented in literature (9). Note that the Studsvik model has predicted concentrations that are one order of magnitude lower than the

ANDRA or EPRI models in Figure 5. This trend is reversed in Figure 6 where the Studsvik model is two orders of magnitude higher than ANDRA or EPRI models. This implies that one irrigation event is much more potent in the Studsvik model than in the ANDRA or EPRI models. In the "normal" model values the Studsvik plants are watered only 4 times in comparison with the ANDRA and EPRI models which are watered during the plants lifespan, which in this experiment was 13 times. Since the models are linear it would seem that the number irrigation events should be linearly proportional to the radioactivity in the plant.

The Studsvik model should predict a normal concentration that is about 1/3, or 33%, of the value of the EPRI or ANDRA models if the number of irrigation event is linearly proportional to radioactivity in the plant, but it does not. In the normal model the Studsvik value is 10% of the ANDRA value and 5% of the EPRI value. The Studsvik model would predict a value close to EPRI and ANDRA for the experimental data if one irrigation event had the same importance in all three models, but it did not. The volume of irrigation water calculated for this experiment is six orders of magnitude smaller than the normal value, and the fraction interception is one order of magnitude larger, accounting for an overall decrease of five orders of magnitude for EPRI and ANDRA. The change in Studsvik was only a factor of four, decreasing from four irrigation events to one. The difference in factors led to the Studsvik model over predicting the concentration of radioactivity in the sample by two orders of magnitude when using only one irrigation event. However, more experiments could be done to verify if the model is a better predictor of actual data for more than one irrigation event.

The results from ANDRA and EPRI are in the correct order of magnitude with the experimental results shown in Figure 6. The radioactivity in the plants from the experiment ranges from 0-500 Bq/g, which encompasses the values of the ANDRA and EPRI models. The biosphere models take into account a number of assumptions and broad generalizations, including the translocation fraction, the volume of irrigation water, and the interception fraction. In addition, the models predict that every plant behaves the same, but this cannot be true in a complex biological system. The models are intended as an estimate which gives the proper order of magnitude. They are not intended to predict exact values. Therefore, the experiment agrees with and confirms the EPRI and ANDRA models when using one irrigation event.

All three models predict that the irrigation water with double the concentration had double the concentration of radioactivity in the plant. However, that trend was not clearly defined in the experimental data. Figure 4b shows that the average concentration in plant tissues for the 50,000 Bq/mL plants can be one half to double the value of the 25,000 Bq/mL plants. This large variation draws into question the assumption that the uptake increases linearly with concentration. Further studies should be under taken to more fully explore this assumption.

Now that the models have been confirmed (or not) with experimental values, a more in depth analysis of the models will take place. The *LAI*StoCap*Kret* term in the Studsvik model can be interpreted as a fraction interception term, as ANDRA's fraction interception term is modeled in a similar manner (9). With that modification, all four

models have C_w (concentration in the irrigation water), F_i (fraction interception), Y (yield), and F_t (fraction translocation) terms. All the models have some form of measuring the amount of irrigation water used, although the method is varied. The ANDRA has an additional term for weathering; whereas, the EPRI model has an additional term for contamination removed during food processing. The JGC has additional terms for both weathering and food processing.

The weathering term is approached differently in almost all of the models. JGC and ANDRA have a weathering term inversely proportional to the concentration; whereas, EPRI has a fraction absorption term that accounts for weathering. The Studsvik model has no weathering term whatsoever. The weathering term makes sense as there is most likely to be wind or rain that can wash the water off the leaves. An example of weathering occurred in the experiment with condensation from the greenhouse roof falling on the radishes and washing some of the contaminated water off the leaves and onto the soil. Especially with the lack of irrigation required in Sweden, it would seem that rain could modify the foliar uptake by displacing the irrigation water from the leaves onto the soil, and adding a weathering term might help the Studsvik model better predict experimental data.

The food processing term is included in the EPRI and JGC model but not the ANDRA or Studsvik model. Food processing can include a number of steps: washing food before eating, peeling, and cooking. Processing is unlikely to matter for radishes because they are normally eaten raw and the leaves, where most of the contamination remains, are not

eaten. On the other hand, potatoes are also a root vegetable and they are often peeled, and almost always cooked before eating. The processing term becomes very important in the case of potatoes, especially if cooking and peeling removes a large fraction of the contaminant load. The food processing term can also play an important role for plant types other than root vegetables. Leafy greens and fruit have a foliar uptake that is directly on the edible portion of the plant, and not washing the food would have a higher concentration than if the food was washed, assuming that washing does remove some contaminant load. EPRI and JGC assume that their constituents wash and/or process their food before eating it, but ANDRA and Studsvik do not. Cultural habits may account for this but even within cultures people vary.

Two important points come out of analyzing the importance of the processing term. The first point is that the models are broad generalizations that group crops into plant types that may not be representative. Classifying root vegetables separate from leafy greens is important but there is still significant variability in the root vegetable category. Having separate data and assumptions for every specific plant and its conditions would be ideal, but it is impractical. The amount of time and effort required to categorize each plant individually is enormous and unfeasible in the best estimate that a biosphere model provides. Values must be estimated for each plant type, and assumptions must be made, which brings up the second point.

The models are intended to give a best estimate of the dose a human might achieve. The levels the models provide are more like order of magnitude predictions rather than exact

value equations. Using conservative estimates is important for safety, but it also comes down to an economic factor. Too conservative estimates can give unreasonable data and cause problems in the economic viability of a project. Therefore, a safe but reasonable assumption would be to include a food processing term, but have a small removal of contaminant load based on the plant type.

Studsvik's model is a product of their geographic location. Sweden receives plenty of rain in the summer, so irrigation is done only a discrete number of times (11). For grain crops irrigation is occasionally performed during the germination stage and could affect the uptake from soil, but would not be involved in foliar uptake (11). Therefore, the Studsvik model uses the number of irrigation events to predict the plant concentration as opposed to the total irrigation water that is used in the other models.

Location is a key factor in the overall model design and diet. The four models that were analyzed were chosen for specific reasons. EPRI is from the United States where the experiment was conducted and is used as the base case for the analysis. ANDRA is from France where the nuclear industry is very strong. ANDRA is very similar to the EPRI model and can be used as a comparison. All of ANDRA's original work is written in French, so it is not cited here. Instead, papers in English where different models are compared are used as references. Studsvik was chosen because of its unique approach to the volume of irrigation water. It better reflects the experiment design. JGC was chosen from Japan to look at a different cultural view point and diet.

One factor in the diet of each model is autarky, or, self-sufficiency. Autarky is a measure of how much locally produced food is eaten. None of the four models have a term for autarky so the diet is representative in how much food is eaten from local sources for the critical group. This term would depend on the location and habits of the population, whether they live in the city or country and whether they buy mostly from a grocery store where food is produced far away or they have their own garden to grow food or they buy locally at Farmers Markets. The worst case scenario would be a population living close to a release of radioactive materials that is highly autarkic. This view is taken on by ANDRA, JGC and Studsvik and EPRI 1996. EPRI 2009 assumes little autarky, due to the limited farming done in southern Nevada where the model is developed for (6).

Location not only impacts the autarky of a region, but also the irrigation habits. As previously mentioned, Sweden doesn't irrigate much, but irrigation is common in desert regions. Irrigation is also dependent on coastal versus inland sites. This depth of analysis is not included here, but has been explored by others (10). Irrigation is a site specific value, it depends on the location. It also depends on the plant type. Some terms in the models are on site, plant type or radionuclide specific, while others are independent, as shown in Table 3. Each model uses different dependencies in their model, so a letter is used to represent the model in Table 3: S for Studsvik, A for ANDRA, E for EPRI and J for JGC.

The diet for each model was taken from the original resource except for ANDRA due to the language barrier. The ANDRA diet was taken from Limer, 2009 (10). Two different

diets were used in the Studsvik model, one that was produced in the original document, here named Studsvik 1, (8), and one that was modified to better describe the people surrounding the high level waste repository, here named Studsvik 2 (15). The EPRI model had multiple diets associated with it, and two of those were chosen: the diet in 1996 paper (16) and the diet in the 2009 paper (6). Each new EPRI paper redefines what the diet of the critical population is. The 1996 paper began with an assumed diet based on written communication with a researcher (16). In 2002, the diet had been modified based on a scientific study, which reduced the amount of autarky in the region (17). The 2009 diet further reduced the amount of autarky (6). The difference in the 1996 and 2009 diet show the extreme of autarky, eating all local or eating just a little bit local. This variable is highly dependent on the location of the individual, their cultural habits, and their personal habits. All diets used in this study are shown in Table 4.

Table 3: This shows the general model terms and their dependencies on site, plant type and radionuclide. Each letter represents the model: S for Studsvik, A for ANDRA, E for EPRI and J for JGC.

General Term	Site Dependent	Plant Dependent	Radionuclide Dependent	Independent
Concentration of Irrigation Water				J, S, E, A
Volume of Irrigation Water	S	J		E, A
Yield	A	J, S, E, A		
Fraction Interception		J, S	S, E, A	
Fraction Translocation		J, S, E, A	S, E, A	
Processing		J, E	J	
Weathering		J, E	J	A

The JGC diet has a rice specific component that is not present in any other diet (5). For all models besides the JGC model, the rice component was added to the grain component.

The Studsvik diets do not have a specific fruit component, and it is unclear which plant group fruit fits in. The EPRI 1996 diet has the highest consumption rates of all the diets; whereas, the EPRI 2009 diet is the smallest due to low rates of autarky.

Table 4: This table shows the different diets used for modeling.

Foodstuff	Units	JGC	Studsvik 1	Studsvik 2	EPRI 1996	EPRI 2009	ANDRA
Rice	kg-fw/yr	59.8					
Grain	kg-fw/yr	28.5			60	0.23	77.3
Root Vegetables	kg-fw/yr	47.2	70	70	80	4.73	31.4
Green Vegetables	kg-fw/yr	26.7	40	60	40	3.78	4.6
Fruits	kg-fw/yr	28.5			46	12.68	59.6
Total	kg-fw/yr	190.7	110	130	226	21.42	172.9

A value for C_{crop} was calculated for each plant type according to each model using the values shown in Appendix A. This C_{crop} value was used in Equation 6 for each plant type and all the doses were added to come up with a total dosage from foliar uptake. Water concentrations of 25,000 Bq/mL were used for C_w to represent the experimental values; however this concentration is very high for likely exposure pathways. All six diets were run on all four models with the results shown in Figure 10 where the diets are listed on the x-axis and the models are shown as columns. The JCG model is in black with white dots, Studsvik in black, EPRI in white with stripes and ANDRA in white with black dots. The Studsvik model has the lowest dosage of all models as expected, from the limited number of irrigation events. The EPRI has the highest dosage because the C_{crop} value in fruit is very high. The Studsvik diets do not have a specific fruit component, and are not affected by the EPRI fruit dose. Grains are not irrigated in Sweden, so there is no grain

component in either Studsvik diet, further reducing the EPRI model predictions, which also had elevated C_{crop} values for grain.

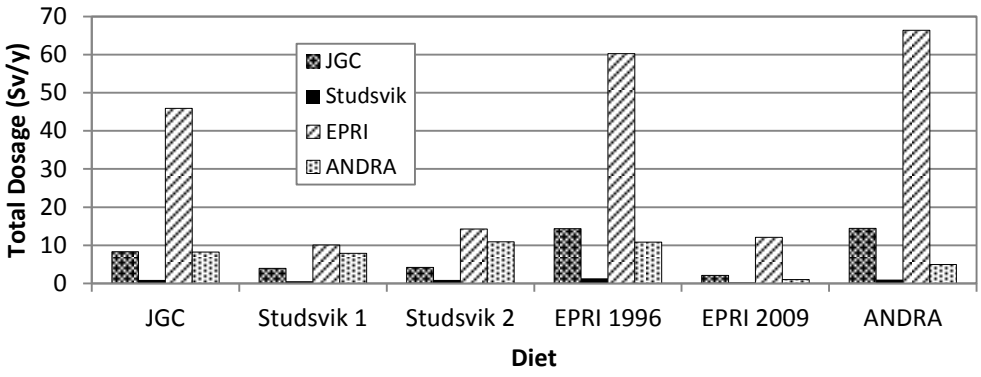


Figure 10: This shows all six different diets run on all four different models. The JCG model is in black with white dots, Studsvik in solid black, EPRI in white with black diagonal stripes and ANDRA in white with black dots.

The values in Figure 10 only represent a small fraction of the yearly dosage. The values are for foliar uptake of Tc-99. They do not represent dosage from other parts of crop uptake or other sources of radioactivity. The values do not include all radionuclides either. Separate calculations would need to be performed for each radionuclide and each source of exposure. This task has been completed by some models, and for JGC, foliar uptake was a dominant exposure pathway for Tc-99 (5).

CONCLUSIONS

In conclusion, the ANDRA and EPRI models are consistent with experimental data because they were on the same order of magnitude as the radish roots (0-500 Bq/g). An order of magnitude is the best estimate given by a biosphere model due to the complex nature of the system and the multiple assumptions that are included in the biosphere model. The Studsvik model was not consistent with experimental data because it was two orders of magnitude larger than the experimental values. The dose from a single irrigation event, as calculated from the Studsvik model, is significantly larger than that same irrigation event calculated in the EPRI or ANDRA models. The experimental results did not exhibit linear behavior with concentration. It is recommended that this relationship be further explored with more concentrations and more replicates. Timing of the contamination is related to percent uptake. The earlier in the life of the plant that the contamination occurs, the more translocation takes place.

The diet that is used in the model contributes minimally to the JGC, ANDRA or Studsvik models, which all had an annual dosage of 0-15 Sv/y for foliar uptake of Tc-99. However, the EPRI model varies from 10-70 Sv/y depending on the diet's components. The fruit and grain concentrations for EPRI are larger by an order of magnitude and contribute to the high doses received using that model. The Studsvik diets did not have fruits or grains and were much lower in yearly dose than models that included fruits and grains, even those in small amounts like the EPRI 2009 diet.

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APPENDIX

APPENDIX A

Values used in modeling.

For biosphere modeling values used in Figures 5 and 6, all normal values are taken directly from the literature except for water concentration which was the value used experimentally (25,000 Bq/L). Experimental values were changed to 1 for the fraction interception and 0.019 $L/m^2/y$ for the volume of irrigation water.

Table A-1: These are the values used in the ANDRA model for the normal and experimental simulations in Figures 5 and 6.

Parameter	Units	Normal Value	Experimental Value
C_{irr}	Bq/L	2.50E+07	2.50E+07
T_{irr}	L/m^2*y	1800	0.019
F_1	-	0.198	1.0
F_2	-	0.1	0.1
Y	kg fw/m^2	1.5	1.5
λ_w	1/y	15	15

Table A-2: These are the values used in the EPRI model for the normal and experimental simulations in Figures 5 and 6.

Parameter	Units	Normal Value	Experimental Value
C_w	mol/m^3	0.411	0.411
I_{crop}	-	0.1	1.0
V_{irr}	m^3/m^2*y	1.8	1.9E-05
F_{abs}	-	0.5	0.5
Y	kg/m^2*y	3	3
F_{p2}	-	1	1
F_{trans}	-	0.1	0.1

Table A-3: These are the values used in the Studsvik model for the normal and experimental simulations in Figures 5 and 6.

Parameter	Units	Normal Value	Experimental Value
n_{irr}	-	4	1
LAI	-	5	5
StoCap	m^3/m^2	3.00E-04	3.00E-04
K_{ret}	-	0.5	0.5
C_w	Bq/m^3	2.50E+10	2.50E+10
Y_c	kg fw/m^2	2	2
T_L	kg/kg	0.1	0.1

For human dose values used in Figure 10, all values are taken from literature.

Table A-4: These are the values used in the JGC model for the normal and experimental simulations in Figure 10.

Parameter	Units	Rice	Grain	Root Vegetables	Green Vegetables	Fruit
μ_{crop}	-	0.05	0.05	0.3	0.3	0.5
d_{crop}	m/y	1.9	0.7	0.7	0.7	0.7
C_{rw}	Bq/m^3	2.50E+10	2.50E+10	2.50E+10	2.50E+10	2.50E+10
F_{crop}	-	0.5	0.5	0	0.9	0
F_{trans}	-	0.12	0.12	0.11	0.28	0.12
Y_{crop}	Kg fw/m^2	0.5	0.4	2.4	3.1	1.4
W_{crop}	1/y	8.4	8.4	18	18	18
C_{crop}	Bq/kg	3.51E+08	1.61E+08	1.35E+08	3.58E+07	3.89E+08

Table A-5: These are the values used in the Studsvik model for the normal and experimental simulations in Figure 10.

Parameter	Units	Grain	Root Vegetables	Green Vegetables	Fruit
n_{irr}	-	4	4	4	4
LAI	-	5	4	5	5
StoCap	m^3/m^2	3.00E-04	3.00E-04	3.00E-04	3.00E-04
K_{ret}	-	0.5	0.5	0.5	0.5
C_w	Bq/m^3	2.50E+10	2.50E+10	2.50E+10	2.50E+10
Y_c	kg fw/m^2	2	2	2	2
T_L	kg/kg	0.1	0.1	1	1
C_{crop}	Bq/kg	3.75E+06	3.00E+06	3.75E+07	3.75E+07

Table A-6: These are the values used in the EPRI model for the normal and experimental simulations in Figure 10.

Parameter	Units	Grain	Root Vegetables	Green Vegetables	Fruit
C_w	Bq/mL	25000	25000	25000	25000
C_w	mol/m^3	0.411	0.411	0.411	0.411
I_{crop}	-	0.1	0.1	0.1	0.1
V_{irr}	m^3/m^2*y	0.8	0.8	0.8	0.8
F_{abs}	-	0.5	0.5	0.5	0.48
Y	kg/m^2*y	0.4	3	3	0.7
F_{p2}	-	1	1	1	1
F_{trans}	-	0.1	0.1	1	1
C_{crop}	mol/kg	0.00411455	0.000549	0.0054861	0.022571
C_{crop}	Bq/kg	2.50E+08	3.33E+07	3.33E+08	1.37E+09

Table A-7: These are the values used in the ANDRA model for the normal and experimental simulations in Figure 10.

Parameter	Units	Grain	Root Vegetables	Green Vegetables	Fruit
C_{irr}	Bq/L	25000000	25000000	25000000	25000000
T_{irr}	L/m^2*y	1800	1800	1800	1800
F_1	-	0.198	0.198	0.198	0.198
F_2	-	0.1	0.1	0.6	0.1
Y	kg fw/m^2	1.5	1.5	1.5	1.5
λ_w	1/y	15	15	15	15
C_{crop}	Bq/kg	3.96E+07	3.96E+07	2.38E+08	3.96E+07