Phytoremediation Dynamic Models of Radionuclides $^{134}$Cs and $^{60}$Co in Sunflowers Plants (Helianthus annuus. L) Using Matlab

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Abstract. In the development of phytoremediation this method can be simulated quite concisely and precisely. Simulations are carried out to predict plant behavior towards several different treatments, for example plant species, also types and concentrations of contaminants. In this study a dynamic model of phytoremediation has been made using sunflower plants and $^{134}$Cs and $^{60}$Co radionuclides. This study was developed by mimicking the interaction of soil and plants to be simulated into Phytoremediation Dynamic Model (PDM). Diverse mathematical algorithms implemented to characterize phytoremediation, systems such as differential equation, statistical correlation, and dynamic system approach. The error value obtained is different for each contaminant for each variation in concentration, which ranges from 0.0006-0.6349 for $^{134}$Cs contaminants and 0.0089-0.4157 for $^{60}$Co contaminants. The error value is quite small, and the overall simulation data has approached the experimental data. Factors that influence the results of calculated data include saturation point values, as well as the absorption rate of each part of the plant obtained from calculations and estimates. This model has proven to be able to mimic plant responses to contaminants.

Keywords: phytoremediation, sunflower, radionuclides, phytoremediation dynamic model

1. Introduction

Operation and utilization of nuclear facilities normally are expected not to cause adverse impacts on the environment or the communities around these nuclear facilities. Because in a nuclear facility accident it is possible to result in the release of a number of radionuclide materials produced by fission or activation into the environment (Tjahaja & Sukmabuana, 2008).

One of the fission results that may be released into the environment in nuclear accidents is $^{134}$Cs, $^{137}$Cs and $^{60}$CO (Kinoshita et al., 2011; Niimura et al., 2015; Ohkura et al., 2012; Tjahaja & Sukmabuana, 2008; Yasunari et al., 2011; Yoshida & Takahashi, 2012). The radionuclides are potentially harmful to human health and the environment, due to emitted gamma radiation and long half-life (2.05 years for $^{134}$Cs, 30 years for $^{137}$Cs and 5.2 years for $^{60}$Co) (Center & ASANO, 2013; EPA, n.d.), so that the radionuclides can be in the environment for a long time. Radionuclides released into the soil can be absorbed by plants and end up in humans through the respiration, and the food chain. Radionuclides that enter the human body or other living things can be sources of internal radiation that damage cells and tissues (Chussetijowati et al., 2009).

One of the efforts made to reduce radionuclide contamination in the environment is phytoremediation. In its development phytoremediation is a method that can be simulated quite concisely and precisely. Simulations are carried out to predict plant behavior towards several different treatments, for example plant species, also types and concentrations of contaminants, so that the preparation of environmental management strategies and decision making can be made objectively without directly affecting environmental conditions. In this study a model of absorption and transport of radionuclide $^{134}$Cs, and $^{60}$CO will be made on sunflower plants (Helianthus annuus. L). Sunflower plants were chosen because it is one of the plants that has been successfully used in the nuclear accident phytoremediation process. For example, in the case of the Chernobyl (1986) and Fukushima (2011) nuclear accidents.
2. Method

2.1. Interaction Between Research Variables

This phytoremediation dynamic model begins by determining the four basic components of the dynamic system, namely stocks (level variables), flows (rates), converters (auxiliary variables), and connectors.

First, stocks (level variables) consist of two main components, biotic and abiotic components. The biotic component is represented by roots, stems, and leaves which will later be used as stocks. While for the abiotic component it is influenced by the soil and atmosphere.

Second, the flows (rates) component in the dynamic model is the previous stocks radionuclide concentration. This variable is certainly related to radionuclide uptake by roots, radionuclide transfer from roots to stems, and stem to leaves, then phytovolatilization also the decay of the radionuclide contaminant.

The third component is converters (auxiliary variables). This model is influenced by the plant diffusivity value of contaminants and the threshold value that can be accumulated by each of the stocks components.

Based on the above variables, it can be arranged a causal loop diagram as shown in figure 1. Each arrow illustrates the causal relationship of each variable quantitatively, where positive arrows indicate if the value of the causal factor rises, then the value of the factor due to going up (growth, strengthening), if the causative factor falls, then the factor is also will go down, whereas arrows marked negative have meaning if the value of the causative factor rises, then the resultant factor will go down and vice versa.

Figure 1. Causal Loop Diagram of Phytoremediation

2.2. Sampling Methods and Preparation (Hydroponic Solutions and Plants)

The sampling of plants and hydroponic solutions was carried out at intervals shortly after hydroponic solution, 1, 2, 4, 6, 8, 24 hours, 5, 6, 10 d, 15, 20 and 30 days. Each sample is randomly selected 8 (3 control and 5 treatment medium plant) plants which have the same relatively physical characteristics. The selection of samples need to be prioritized the withered plants and dying. The hydroponic solution sampling done by entering 100 ml of sample plants hydroponically previously taken into bottles.

Plant samples were washed to remove dirt and then cut to separate the three main components. The wet weight of each part of the plant is measured before it is dried. The drying process is carried out using an oven at 100°C until the sample mass is stable. This step is carried out in order to obtain sample dry weight and the absorption of radionuclides by plants only.

Plants from experiments were evaluated using a High Purity Germanium (HPGe) detector equipped with Multi Channel Analyzer (MCA) (Tjahaja & Sukmabuana, 2007) for 600 seconds for each sample.
2.3. Model Completion

Completion of models from simultaneous differential equations using the equation in table 1. The equation is the rate of change in concentration of radionuclides $^{134}\text{Cs}$ and $^{60}\text{Co}$ which accumulated in plant body parts. The equation consists of ten variables that are resolved using the ode45 formula in the M-file MATLAB program.

| Section           | Mathematical Equation                                                                 |
|-------------------|----------------------------------------------------------------------------------------|
| Soil              | $\frac{dS_{\text{soil}}}{dt} = -F_{\text{Ext}} - \Delta S_{\text{decay soil}}$         |
|                   | $\Delta S_{\text{decay soil}} = S_{\text{soil}} - S_{\text{soil}}e^{-\lambda t}$    |
|                   | $F_{\text{Ext}} = \left( S_{\text{soil}} \times \left( \frac{S_{\text{soil}}}{\text{Init}_S_{\text{soil}}} \right)^{\text{Fraction}} \right) \times R_{\text{ext}}$ |
| Root              | $\frac{dS_{\text{root}}}{dt} = F_{\text{Ext}} - F_{\text{Tran}} - \Delta S_{\text{decay root}}$ |
|                   | $\Delta S_{\text{decay root}} = S_{\text{root}} - S_{\text{root}}e^{-\lambda t}$     |
|                   | $F_{\text{Tran}} = (S_{\text{root}} - \text{ThC}_{\text{root}}) \times R_{\text{Tran}}$ |
| Shoot             | $\frac{dS_{\text{shoot}}}{dt} = F_{\text{Tran}} - F_{\text{inc}} - \Delta S_{\text{decay shoot}}$ |
|                   | $\Delta S_{\text{decay shoot}} = S_{\text{shoot}} - S_{\text{shoot}}e^{-\lambda t}$   |
|                   | $F_{\text{inc}} = (S_{\text{shoot}} - \text{ThC}_{\text{shoot}}) \times R_{\text{inc}}$ |
| Leaf              | $\frac{dS_{\text{leaf}}}{dt} = F_{\text{inc}} - F_{\text{vol}} - \Delta S_{\text{decay leaf}}$ |
|                   | $\Delta S_{\text{decay leaf}} = S_{\text{leaf}} - S_{\text{leaf}}e^{-\lambda t}$    |
|                   | $F_{\text{vol}} = (S_{\text{leaf}} - \text{ThC}_{\text{leaf}}) \times R_{\text{vol}}$ |
| atmosphere        | $\frac{dS_{\text{atm}}}{dt} = F_{\text{vol}}$                                          |
|                   | $C_t = C_0 e^{-Rt}$                                                                   |

Where $S_\text{ }$ is Stock and $\text{ThC}_\text{ }$ is threshold concentration. Each function has a sub-index soil, root, shoot, leaf and atmosphere. $R_\text{ }$ represents the transfer rate of contaminant concentrations in each part. $F_\text{ }$ is the concentration gradient of each adjacent stock. $F_\text{ }$ has several sub-indexes such as ext for extraction, tran for translocation, inc for incorporation, vol for volatilization. Init$_S_{\text{soil}}$ is the initial concentration value in the soil which will be simulated later. Fraction is the bioavailability of contaminants in the soil. $\Delta S_\text{ }$ is the difference between the value of concentration before and after decay.

The parameters used in modeling are obtained from literature studies, numerical calculations, and estimated. The parameters used in making this dynamic model can be seen in table 2.

Experimental data is simulated along with phytoremediation models as a reference and to determine the trend of responses from changes in the value of variables in the system. The simulation results are depicted in a scatter graph between the experimental data and the simulated data.

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Table 2. Parameter values used in the simulation

| Name (units)                      | Category     | Value  |
|----------------------------------|--------------|--------|
| $^{134}$Cs                       |              |        |
| decay constant ($/s$)            | Reference    | 0.000038 |
| Fraction                         | Reference    | 0.018  |
| Extraction rate (Bq/grs)         | Estimated    | 0.0024 |
| Translocation rate (Bq/grs)      | Estimated    | 0.0053 |
| Incorporation rate (Bq/grs)      | Estimated    | 0.004  |
| Volatilization rate (Bq/grs)     | Estimated    | 0.00595 |
| Root threshold (Bq/gr)           | Estimated    |        |
| 0.85 Bq/gr                       |             | 0.35   |
| 1.31 Bq/gr                       |             | 0.35   |
| 1.74 Bq/gr                       |             | 0.23   |
| 2.24 Bq/gr                       |             | 0.25   |
| 2.67 Bq/gr                       |             | 0.07   |
| Shoot threshold (Bq/gr)          | Estimated    |        |
| 0.85 Bq/gr                       |             | 1.05   |
| 1.31 Bq/gr                       |             | 1.235  |
| 1.74 Bq/gr                       |             | 1.535  |
| 2.24 Bq/gr                       |             | 2.65   |
| 2.67 Bq/gr                       |             | 0.9    |
| Leaf threshold (Bq/gr)           | Estimated    |        |
| 0.85 Bq/gr                       |             | 0.85   |
| 1.31 Bq/gr                       |             | 0.9    |
| 1.74 Bq/gr                       |             | 1.74   |
| 2.24 Bq/gr                       |             | 1.78   |
| 2.67 Bq/gr                       |             | 1.32   |
| $^{60}$Co                        |              |        |
| decay constant ($/s$)            | Reference    | 0.000015 |
| Fraction                         | Reference    | 0.05   |
| Extraction rate (Bq/grs)         | Estimated    | 0.0001 |
| Translocation rate (Bq/grs)      | Estimated    | 0.0014 |
| Incorporation rate (Bq/grs)      | Estimated    | 0.0033 |
| Volatilization rate (Bq/grs)     | Estimated    | 0.004  |
| Root threshold (Bq/gr)           | Estimated    |        |
| 4.21 Bq/gr                       |             | 0.083  |
| 8.54 Bq/gr                       |             | 0.3    |
| 12.19 Bq/gr                      |             | 0.5    |
| Shoot threshold (Bq/gr)          | Estimated    |        |
| 4.21 Bq/gr                       |             | 0.02   |
| 8.54 Bq/gr                       |             | 0.4    |
| 12.19 Bq/gr                      |             | 0.01   |
| Leaf threshold (Bq/gr)           | Estimated    |        |
| 4.21 Bq/gr                       |             | 0.08   |

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3. Result and Discussion

3.1. Phytoremediation Dynamic Model $^{134}\text{Cs}$ at a concentration of 0.85 Bq/gr

The first simulation was carried out on $^{134}\text{Cs}$ contaminants with an initial concentration of 0.85 Bq/gr. The uptake of $^{134}\text{Cs}$ by plants is shown by comparing between two data, calculated and experiment data. The results of the model in Figure 2 are not very visible because there are not many experimental data obtained with an initial concentration of 0.85 Bq/gr as a comparison, but the changes in concentration values accumulated by plants with relatively similar trends with stem and leaf. In this simulation the estimated saturation point of root absorption was 0.35 Bq/gr, stem 1.05 Bq/gr, and leaves 0.85 Bq/gr. The saturation estimation is done manually with an approach using estimated value to obtain a graph of the simulation results that are close to the experimental data.

Furthermore, the difference in values in the calculated and experimental data is used to evaluate the accuracy of the model by determining the error value. The error value is calculated using the RMSE (Root Mean Square Error) method which is the average value of the number of squared errors. The smaller error value obtained mean a better accuracy of the data simulation results. From the graph obtained an error value of 0.0006 for roots, 0.0056 for stems and 0.0029 for leaves. The error value obtained is very small so that it can be said that the simulation results approached the experimental data.

![Figure 2](image.png)

Figure 2. Uptake and transport of 0.85 Bq/gr $^{134}\text{Cs}$ in sunflower

3.2. Phytoremediation Dynamic Model $^{134}\text{Cs}$ at a concentration of 1.31 Bq/gr

The second simulation was carried out with an initial concentration of 1.31 Bq/gr. Figure 3 shows changes in concentration values accumulated by plants with a relatively similar trend pattern. In the comparison graph of calculated data and experimental data below, it can be seen that the model cannot fully follow changes in plant behavior. The results of the experimental data show changes in concentration values that form peak and valley. In this simulation the estimated saturation point of root absorption is 0.35 Bq/gr, stem 1.235 Bq/gr, and leaves 0.9 Bq/gr. From the graph obtained an error value of 0.3426 for roots, 0.3961 for stem and 0.6349 for leaves. The error value obtained is quite small so that it can be said that the simulation results approach the experimental data.
3.3. Phytoremediation Dynamic Model $^{134}$Cs at a concentration of 1.74 Bq/gr

The third simulation was carried out with an initial concentration of 1.74 Bq/gr. The results of the comparison between the calculated and experimental data showed the changes in concentration values accumulated by plants with a relatively similar trend pattern. In this simulation, estimated saturation of root absorbance was 0.23 Bq/gr, stem 1.535 Bq/gr, and leaves 1.74 Bq/gr. In the graph above it can be said that the model can approach plant behavior because the simulation results have approached the distribution of experimental data values. The value of the errors obtained is 0.2642 for roots, 0.2523 for stems and 0.1242 for leaves. A small error value proved that the simulation results are close to the experimental data.

3.4. Phytoremediation Dynamic Model $^{134}$Cs at a concentration of 2.24 Bq/gr

The fourth simulation was carried out with an initial concentration of 2.24 Bq/gr. The results of the comparison between the calculated data and experimental data showed changes in concentration values accumulated by plants with a relatively similar trend pattern. In this simulation the estimated saturation point of root absorption is 0.25 Bq/gr, stem 2.65 Bq/gr, and leaves 1.78 Bq/gr. In the graph it can be said that the model can approach crop uptake behavior because the simulation approached the distribution of experimental data values. The error values obtained were 0.2335 for roots,
0.1104 for stems and 0.0226 for leaves. The error value obtained is quite small so that it can be said that the simulation results approach the experimental data.

3.5. Phytoremediation Dynamic Model $^{134}$Cs at a concentration of 2.67 Bq/gr

The fifth simulation was carried out with an initial concentration of 2.67 Bq/gr. In this simulation the estimated saturation point of root absorption is 0.07 Bq/gr, stem 0.9 Bq/gr, and leaves 1.32 Bq/gr. The error value obtained were 0.2719 for roots, 0.1633 for stems and 0.0201 for leaves. The error value obtained is quite small so that it can be said that the simulation results approach the experimental data.

3.6. Phytoremediation Dynamic Model $^{60}$Co at a concentration of 4.21 Bq/gr

The sixth simulation was carried out on 60Co contaminants with an initial concentration of 4.21 Bq/gr. Figure 7 shows the changes in concentration values accumulated by plants with a similar pattern as the absorption of 134Cs in each part. In this simulation estimated saturation points of root absorption are 0.083 Bq/gr, stems 0.02 Bq/gr, and leaves
0.08 Bq/gr. The error value is 0.2041 for roots, 0.1368 for stems and 0.2175 for leaves. The error value obtained is quite small, so it can be said that the simulation results approach the experimental data.

3.7. Phytoremediation Dynamic Model $^{60}$Co at a concentration of 8.54 Bq/gr

The seventh simulation was carried out with an initial concentration of 8.54 Bq/gr. From figure 8 shows changes in concentration values accumulated by plants with a relatively similar trend pattern. In this simulation, the estimated saturation point of root absorption is 0.3 Bq/gr, stem 0.4 Bq/gr, and leaves 0.75 Bq/gr. The error value is 0.2546 for roots, 0.2073 for stems and 0.4157 for leaves. The error value obtained is quite small so that it can be said that the simulation results approach the experimental data.

3.7. Phytoremediation Dynamic Model $^{60}$Co at a concentration of 12.19 Bq/gr

The eighth simulation was carried out with an initial concentration of 12.19 Bq/gr. From figure 4.9 shows changes in concentration values accumulated by plants with a relatively similar trend pattern. In this simulation the estimated saturation point of root absorption is 0.5 Bq/gr, stem 0.01 Bq/gr, and leaves 0.12 Bq/gr. The error value is 0.1908 for roots, 0.0089 for stems and 0.0524 for leaves. The error value obtained is quite small so that it can be said that the simulation results approach the experimental data.
3.7. Comparative Analysis of Calculated Data and Experimental Data

In this study a model was used to mimic the uptake and transport behavior of radionuclide \(^{134}\)Cs and \(^{60}\)Co in sunflower plants. Based on the comparison graph of the calculated and the experimental data above, it can be seen that sunflowers have the similar absorption and transport behavior both of \(^{134}\)Cs and \(^{60}\)Co in variated concentration. In addition, the higher the concentration of \(^{134}\)Cs and \(^{60}\)Co dissolved in the planting media used as the initial concentration value in the model, the higher the concentration of \(^{134}\)Cs accumulated in the sunflower body parts. This proven that sunflowers can absorb and accumulate \(^{134}\)Cs well. But different from \(^{60}\)Co uptake. The greater the initial concentration, the less absorbed.

The ability of sunflowers to absorb \(^{134}\)Cs with several concentration variations is evidenced by changes in concentration of contaminants observed from the sampling and simulation results. Figure 2 to 6 showed that absorption \(^{134}\)Cs has occurred since the beginning of the simulation time. Absorption of Cs by plants occurs such as absorption of K elements. According to G. Zhu (Zhu & Shaw, 2000), elements Cs can enter into the plant through two mechanisms in the root cell membrane just like the K element. Sunflower plants absorb \(^{134}\)Cs through the roots, then distributed throughout the plant parts. From the results above, the concentration of \(^{134}\)Cs accumulates the most in the leaves, then in the stem and the lowest in the roots. The reason the leaves have the highest accumulated value of \(^{134}\)Cs is the presence of the process of forming carbohydrates in the process of photosynthesis. The accumulation of \(^{134}\)Cs in leaves is related to the role of K elements in the photosynthesis process, because K and Cs are in the same group. Potassium has function in the process of opening and closing the stomata and is an activator of many enzymes that are important in the process of photosynthesis and respiration.

The ability of sunflowers to absorb \(^{60}\)Co with several concentration variations is evidenced by changes in concentration of contaminants observed from the results of sampling and simulation results. Figure 7 to 9 shows that \(^{60}\)Co absorption has occurred since the beginning of the simulation time. The existence of \(^{60}\)Co in plants occurs because cobalt (Co) is a trace element whose existence is very much needed by plants but in very small amounts. In the cobalt plant it functions as a catalyst in the process of nitrogen fixation (Maria & Co, 2004)(Maria and Co 2004) and as an ethylene forming which can inhibit aging, fruit ripening and leaf abortion.

The results of the comparison between the calculated and experimental data are performed using the RMSE method (Root Mean Square Error). The error values obtained vary for each concentration ranging from 0.0006-0.6349 for \(^{134}\)Cs and 0.0089-0.4157 for \(^{60}\)Co. The error value is quite small, and the overall simulation data has approached the experimental data. Based on these data it can be said that the model has been able to adjust to changes in the value of the variables entered into the model. Some of the factors that influence the results of the calculated data include saturation point values, as well as the absorption rate of each part of the plant obtained from calculations and estimates, while a decrease in the ability of plants to absorb contaminants caused by plant organ damage due to exposure to radiation from external and internal radiation of contaminants cannot be estimated and included in this model. From the data above, it can be said that this model has been able to display an approach to plant behavior and is able to adjust to changes in the conditions of concentration of contaminants.
4. Conclusion

Based on the results of the research, the following conclusions can be drawn: The phytoremediation dynamic model used in this study has been able to display an approach to plant behavior and is able to adjust to changes in type and concentration of contaminants. The error value obtained is different for each contaminant for each variation in concentration, which ranges from 0.0006-0.6349 for $^{134}$Cs contaminants and 0.0089-0.4157 for $^{60}$Co contaminants. The error value is quite small, and the overall simulation data has approached the experimental data. Factors that influence the results of calculated data include saturation point values, as well as the absorption rate of each part of the plant obtained from calculations and estimates, while the decrease factor in the ability of plants to absorb contaminants caused by plant organ damage due to exposure to radiation both external and internal radiation originating from contaminants cannot be estimated and included in this model.

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