3.9 THE TRANSFER OF RADIONUCLIDES IN THE TERRESTRIAL ENVIRONMENTS - RECENT RESEARCH RESULTS IN MONSOON TROPICAL CONDITION OF VIETNAM.

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ABSTRACT

The data on Radionuclide transfer parameters in the environments, which are used in radioecological models, are very necessary for setting release limits of radioactive effluent and assessing the radiation dose to Man related to the releases of radionuclides from nuclear facilities. They strongly depend on climatic, geographic, environmental and pedological conditions. For temperate environments, they are abundant and have been established fairly well [1]. Meanwhile the literature data are still scare and dispersal for Tropical and Sub-tropical zones [2, 3]. Besides, the improvement of Environmental Transfer Models and Parameters is an important problem so that they may be adapted for Southeast Asian countries including Japan as environmental conditions and foodstuffs in this Region are significantly different from those in Europe and North America.

The paper presents measurements results of the dry deposition velocities of atmospheric aerosols carrying ^{7}Be, ^{137}Cs radionuclides and measurements results of soil to plant transfer factors (TF) for ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs resulted from the out door radiotracer experiments with large pots.

The selected soil types (Podzolic, Ferralitic, Ferralic Acrisols, Eutric Fluvisols and Orthi-thionic Fluvisols soil) and the plants (rice, black bean, cabbage, lettuce, tomato, cucumber, carrot, white radish, potato) used for the research are the most common in Vietnam.

The measured Vg values (cm/s) are in the range of 0.01 - 1.84 for ^{7}Be and 1.95 - 49.77 for ^{137}Cs. An analysis of the associated meteorological parameters showed some correlations between ^{7}Be Vg with humidity and ^{137}Cs Vg with wind velocity.

More than 400 TF (edible part) values were determined and their dependences on some soil parameters have been shown.

KEYWORDS: ^{7}Be, ^{137}Cs dry deposition velocities; ^{60}Co, ^{65}Zn, ^{85}Sr, ^{134}Cs TF; factor & multiple regression analysis.

1. INTRODUCTION

The models predicting deposition of materials from the atmosphere require knowledge of the dry deposition velocity (Vg) of the aerosols carrying the substances into the concerned collector. The value of Vg (cm/s) is defined as the ratio of the deposition rate D (1/cm^2.s) to the atmospheric concentrations A (1/cm^3). If Vg values are known and A can be calculated from source data or measured, the deposition is given as $D = \Sigma A_i \times Vg$. The selected experimental sites spread over a climatic-geographic region from 11°57'N; 108°26'E to 10°02'N; 105°47'E. Dry deposition velocity depends on some meteorological parameters. The parameters in this study were obtained in the nearest meteorological station.

The soil to plant transfer factor (TF) is one of the most important parameters used in radioecological models for predicting the concentration of radionuclides from soil to agricultural crops and estimating radiation dose to Man. TF is defined as the ratio of specific radioactivity of dry crops (Bq/kg) to specific radioactivity of dry soil (Bq/kg) in the rooting zone.

The radiotracer technique was applied to determine the soil to plant TF. In this study, ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs were used as tracers for determining their TF from some types of soil to some plants such as rice, black bean, leaf vegetables (cabbage, lettuce), fruit vegetables (tomato, cucumber), root vegetables (carrot, white radish, potato). ^{60}Co, ^{65}Zn. ^{137}Cs and ^{90}Sr are very important nuclides because ^{60}Co, ^{65}Zn are produced in nuclear facilities as activation products and ^{137}Cs, ^{90}Sr are fission products. The experience has also shown that there is no difference of behaviour of ^{137}Cs & ^{134}Cs or ^{90}Sr & ^{85}Sr.

2. EXPERIMENTS

2.1. Measurements of the Dry Deposition Velocities of Atmospheric Aerosols Carrying ^{7}Be and ^{137}Cs Radionuclides

Two air sampling stations were set up in the region where aerosol and fallout samples are collected monthly. The location of the air sampling sites is suitable for such measurements in view of the existence of several months without any rainfall.

Particulate radioactivity was collected from about 100000m^3 of air on 0.48m^2 chlorinated vinyl polychloride Petrianow filter FPP-15-1.7 using an air sampler 12-UC-34 with flow rate of 760m^3/h. The intake was 1.5m above the ground. The filters were compressed into pellets of 36mm diameter and 10 mm thickness for gamma spectrum measurement.

Dry fallout was collected throughout the month in three stainless-steel trays. Each tray is cross-sections 0.4m^2 with 10cm high sides. The resuspension of dry fallout particles was prevented by adding distilled water to a depth of about 1cm. The sample was passed through a filter to separate suspended and dissolved fractions. The filtrate was subsequently evaporated in vacuum. The residue was made into suitable samples for gamma spectrometry.

Gamma spectra were measured by using a low background system with high purity Ge detector of 15% efficiency and 1.9 keV FWHM for 1332.5 keV ^{60}Co line. The integral background (100-2000 keV) was 1.7cps. The measuring time was often no less than 20h, yielding detection limit for both the ^{7}Be and ^{137}Cs in the air and fallout not exceeding 0.02μBq/m^3 and 0.01 Bq/m^2, respectively.

2.2. Measurements of ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs TF Resulted from the out Door Radiotracer Experiments with Large Pots

The measurements have been carried out under the conditions of: (1) Radionuclides: ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs, the chloride form with carrier; (2) Levels of soil labelling: about 15000, 30000, 45000, 20000 Bq/kg soil for ^{60}Co, ^{65}Zn, ^{85}Sr, ^{134}Cs, respectively; (3) Plants: rice, black bean, cabbage, lettuce, tomato, cucumber, carrot, white radish, potato; (4) Type of experiments: out door experiments with plastic large pots of 71 l volume (ϕ 600mm x h 250mm), containing about 50 kg of soil/pot; (5) Method of mixing in a concrete mixer (Paribat S.A. AS130, French) used to label the soils. Out door experiments with plastic large pots have been implemented by supervising of local agricultural experts.

The methods and procedures of our experiments followed the protocol for Transfer Parameter Measurement [4, 5].

Specific activity concentrations of ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs in soil and plant samples were also quantitatively determined through their gamma lines by using the above-mentioned gamma spectrometer based on the standard methods [6, 7].

By using the standard chemical analytical methods, soil parameters such as pH, exchangeable K, exchangeable Ca, CEC and organic matter (OM) contents were obtained and included in the IAEA-IUR standard data sheets. The experimental conditions such as radionuclide concentrations, farming regimens, types of soil, time of contamination could also be found in the sheets [2, 3].

3. RESULTS AND DISCUSSION

3.1. The Dry Deposition Velocities of ^{7}Be and ^{137}Cs Radionuclides

The data we used for calculating Vg are monthly measurements of ^{7}Be and ^{137}Cs in both atmosphere and dry deposit during the dry periods of 1986 - 2001. The measured Vg values (cm/s) are presented in Table 1.

Table 1. The averaged ^{7}Be & ^{137}Cs dry deposition velocities over months of dry periods from 11/1986 to 11/2001 at Dalat (11°57'N, 108°26'E).

Nuclide	Vg (cm/s)			Number of observations
	Range	Mean	Error (95%)	
^{7}Be	0.01 - 1.84	0.49	0.17	34
^{137}Cs	1.95 - 49.77	16.31	7.70	14
$^{134}Cs^{*}$		49.46	2.10	1

** ^{134}Cs dry deposition velocity was obtained in 12/1986, after Chernobyl accident. At the same time, the measured Vg of ^{137}Cs was 49.77 ± 2.11 (cm/s).*

Table 1 shows that Vg for ^{137}Cs is higher by more than an order of magnitude compared to ^{7}Be. There is some evidence that ^{7}Be is on smaller size aerosols than fission products [8].

The measured Vg values of ^{7}Be and ^{137}Cs were about 20 times and 15 times, respectively higher than those obtained by other authors [9, 10, 11, 12]. This demonstrates one of the most relevant features of cold air masses during the winter monsoon periods - behind the cold front, vertical air motion is descending [13].

Factor analysis

Factor analysis was used to condense the information contained in a set of variables into a smaller set of new composite variables called factors, which are derived from the correlation structure of the initial variables. In this study, the principal component analysis included in Statgraphics plus for windows version 3.3 software, was performed to generate the factors, which were then subjected to orthogonal rotation with the varimax method.

Application of this approach for the 8 observed variables of ^{7}Be & ^{137}Cs dry deposition velocities and meteorological parameters (maximum wind velocity, mean wind velocity, downwind velocity, prevailing wind frequency, mean humidity and minimum humidity) shows that 71.2% of the overall variation in the data can be explained by two latent variables. The After Varimax Rotation resultant factor loadings are given in Table 2 and they corresponded to the following groups of variables: ^{137}Cs dry deposition velocity, maximum wind velocity, mean wind velocity, downwind velocity, prevailing wind frequency (factor 1); ^{7}Be dry deposition velocity, mean humidity, minimum humidity (factor 2).

Table 2. The After Varimax Rotation resultant factor loadings (only loadings >0.25 are reported)

	Factor 1	Factor 2
^{7}Be dry deposition velocity		0.815
^{137}Cs dry deposition velocity	0.776	
Maximum wind velocity	0.924	
Mean wind velocity	0.895	
Downwind velocity	0.703	
Prevailing wind frequency	0.656	
Mean humidity		0.937
Minimum humidity	0.489	0.693

The following groups of variables can be distinguished:

- ^{137}Cs dry deposition velocity, maximum wind velocity, mean wind velocity, downwind velocity and prevailing wind frequency: Each variable in this group shows a strong and consistent linear relationship with another.
- ^{7}Be dry deposition velocity, mean humidity, minimum humidity: The strongest linear relationships occurred for the pairs ^{7}Be dry deposition velocity/mean humidity and ^{7}Be dry deposition velocity/minimum humidity.

Regression analysis

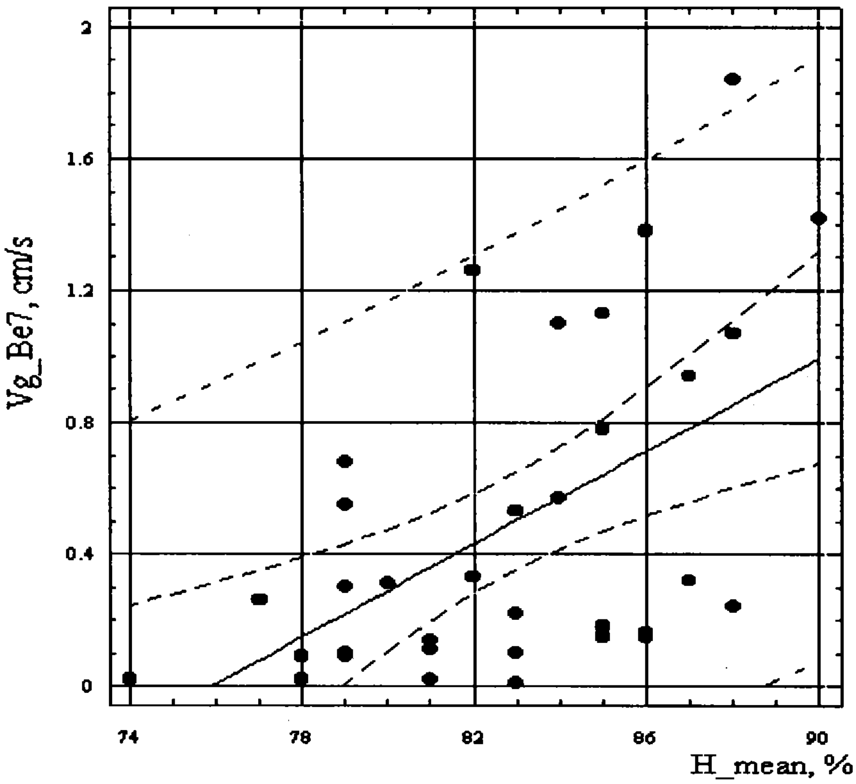
In a multiple regression model, variance (R^2) and its significance (P value) are used to estimate the relationship between obtained variables.

- ^{7}Be dry deposition velocity:

For ^{7}Be dry deposition velocity (Vg_^{7}Be) and mean humidity (H_mean), the significant relationship between the variables was found. The total variance of variables is obtained about 29.0%. The result of fitting in linear model was described by the relationship between Vg_^{7}Be and H_mean as follow:

Vg_^{7}Be = -5.355 + 0.1×H_mean

Since the P-value in the ANOVA table is less than 0.001, there is a statistically significant relationship between Vg_^{7}Be and H_mean at the 99.9% confidence level. The correlation coefficient is fitted approximatively 0.539, indicating a moderately strong relationship between 2 variables. The standard error of the estimate shows the standard deviation of the residuals to be 0.423. These values can be used to construct prediction limits for new observations of Vg_^{7}Be and this fitted model was showed in Fig. 1.



---- 95% confidence intervals
------ 95% prediction intervals

Fig. 1. The predicted values for Vg_^{7}Be using the fitted model.

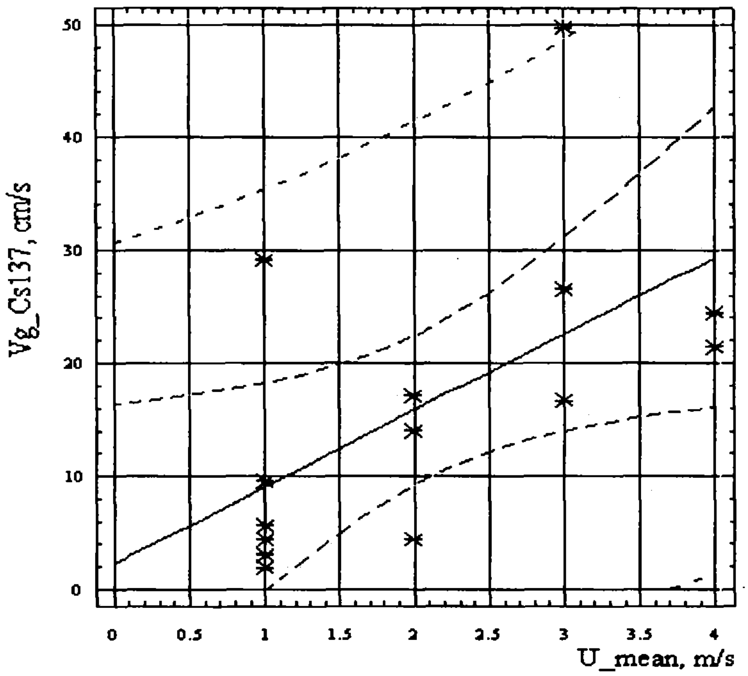
- ^{137}Cs dry deposition velocity:

The output shows the result of fitting a linear model to describe the relationship between ^{137}Cs dry deposition velocity (Vg_^{137}Cs) and mean wind velocity (U_mean). The equation of the fitted model is

Vg_^{137}Cs = 2.278 + 6.8×U_mean

Since the P-value in the ANOVA table is less than 0.030, there is a statistically significant relationship between Vg_^{137}Cs and U_mean at the 97.0% confidence level.

The total variance of variables is obtained about 33.6%. The correlation coefficient equals 0.580, indicating a moderately strong relationship between the variables. The standard error of the estimate shows the standard deviation of the residuals to be 11.311. The fitted model was showed in Fig.2.



---- 95% confidence intervals

------ 95% prediction intervals

Fig. 2. The predicted values for Vg_^{137}Cs using the fitted model.

3.2. Soil To Plant Transfer Factors Derived from Pot Experiments.

456 soil to plant transfer factors (edible part) were determined. ^{60}Co TF values ranged from 0.009 to 0.011 for rice, 0.195 to 3.632 for black beans, 0.032 to 0.230 for cabbage, 0.033 to 0.277 for lettuce, 0.139 to 0.468 for tomato, 0.409 to 0.686 for cucumber, 0.064 to 0.081 for carrot, 0.107 to 0.213 for white radish, 0.356 to 0.386 for potato. ^{65}Zn TF values ranged from 0.789 to 0.957 for rice, 0.931 to 2.453 for black bean, 0.732 to 3.158 for cabbage, 0.909 to 4.776 for lettuce, 0.579 to 2.641 for tomato, 2.204 to 3.420 for cucumber, 0.560 to 0.862 for carrot, 1.590 to 2.237 for white radish, 0.924 to 1.487 for potato. ^{85}Sr TF values ranged from 0.051 to 0.330 for rice, 0.579 to 5.892 for black bean, 1.436 to 38.500 for cabbage, 2.938 to 11.965 for lettuce, 0.393 to 4.234 for tomato, 2.477 to 2.797 for cucumber,1.076 to 1.318 for carrot, 2.654 to 2.970 for white radish, 0.571 to 0.743 for potato. ^{134}Cs TF values ranged from 0.0004 to 1.400 for rice, 0.104 to 0.769 for black bean, 0.004 to 7.500 for cabbage, 0.364 to 2.895 for lettuce, 0.361 to 1.191 for tomato, 1.577 to 2.301 for cucumber, 0.590 to 0.811 for carrot, 0.446 to 0.652 for white radish, 0.563 to 0.997 for potato.

The water content of the experimental fresh products, from which their dry matter content could be calculated, is not different from those obtained in local normal agricultural practice. This means that our out door radiotracer experiments with large pots had no effect on the growth and yield of the crops.

Average TF derived from pot experiments are presented in Tables 3, 4, 5, 6 and Fig. 3.

Table 3. Average ^{60}Co TF ($Bq.kg^{-1}$ dry weight/$Bq.kg^{-1}$ dry soil) derived from pot experiments

Plant	Type of soil	TF		TF range	Number of observations
		Mean	Error (95%)		
Rice	Ferralitic	0.010	0.002	0.009-0.011	3
Black bean	Podzolic	0.939	0.775	0.195-3.632	18
Cabbage	Podzolic	0.114	0.033	0.032-0.230	19
Lettuce	Podzolic	0.111	0.038	0.033-0.277	20
Tomato	Podzolic	0.260	0.054	0.139-0.468	18
Cucumber	Podzolic	0.550	0.077	0.409-0.686	8
Carrot	Podzolic	0.072	0.032	0.064-0.081	3
White radish	Podzolic	0.173	0.144	0.107-0.213	3
Potato	Podzolic	0.367	0.040	0.356-0.386	3

Table 4. Average ^{65}Zn TF derived from pot experiments

Plant	Type of soil	TF		TF range	Number of observations
		Mean	Error (95%)		
Rice	Ferralitic	0.892	0.224	0.789-0.957	3
Black bean	Podzolic	1.559	0.286	0.931-2.453	18
Cabbage	Podzolic	1.727	0.265	0.732-3.158	19
Lettuce	Podzolic	1.877	0.373	0.909-4.776	20
Tomato	Podzolic	1.415	0.249	0.579-2.641	18
Cucumber	Podzolic	2.759	0.325	2.204-3.420	8
Carrot	Podzolic	0.711	0.529	0.560-0.862	3
White radish	Podzolic	1.928	0.807	1.590-2.237	3
Potato	Podzolic	1.136	0.760	0.924-1.478	3

Table 5. Average ^{85}Sr TF derived from pot experiments.

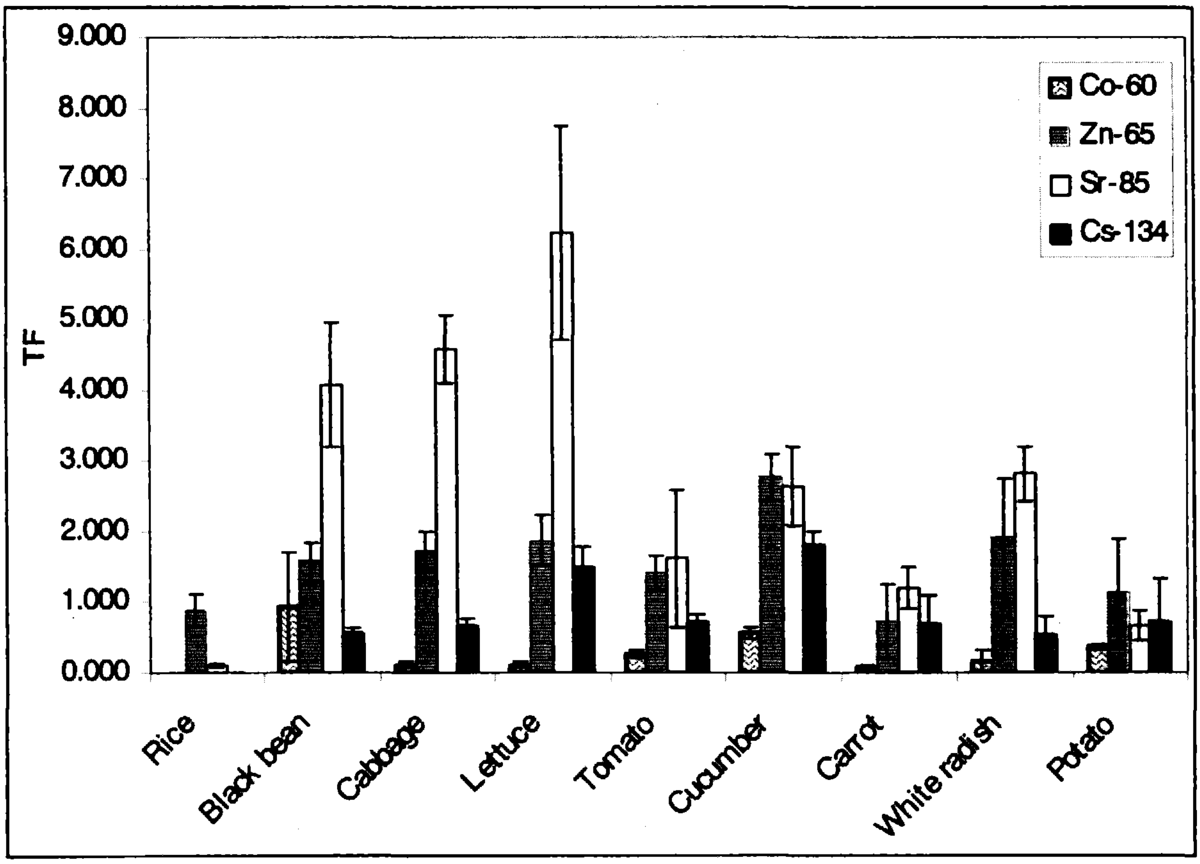
Plant	Type of soil	TF		TF range	Number of observations
		Mean	Error (95%)		
Rice	Ferralitic	0.093	0.027	0.081-0.103	3
	Ferralic Acrisols	0.199	0.075	0.128-0.330	6
	Eutric Fluvisols	0.112	0.049	0.064-0.170	6
	Orthi-thionic Fluvisols	0.080	0.075	0.051-0.110	3
Black bean	Podzolic	4.078	0.884	0.579-5.892	11

Plant	Type of soil	TF Mean	TF Error (95%)	TF range	Number of observations
Cabbage	Podzolic	2.527	0.626	1.436-4.524	16
	Ferralic Acrisols	23.999	11.017	16.248-38.500	5
	Eutric Fluvisols	10.932	1.828	9.444-14.000	6
	Orthi-thionic Fluvisols	6.995	0.871	5.700-7.716	6
Lettuce	Podzolic	6.233	1.514	2.938-11.965	16
Tomato	Podzolic	1.622	0.974	0.393-4.234	11
Cucumber	Podzolic	2.637	0.564	2.477-2.797	3
Carrot	Podzolic	1.197	0.301	1.076-1.318	3
White radish	Podzolic	2.812	0.392	2.654-2.970	3
Potato	Podzolic	0.657	0.214	0.571-0.743	3

Table 6. Average ^{134}Cs TF derived from pot experiments

Plant	Type of soil	TF Mean	TF Error (95%)	TF range	Number of observations
Rice	Ferralitic	0.094	0.022	0.083-0.100	3
	Ferralic Acrisols	0.614	0.425	0.089-1.400	8
	Eutric Fluvisols	0.094	0.119	0.0004-0.350	8
	Orthi-thionic Fluvisols	0.0012	0.004	0.0009-0.0014	2
Black bean	Podzolic	0.568	0.077	0.104-0.769	18
Cabbage	Podzolic	1.348	0.165	0.668-1.963	19
	Ferralic Acrisols	3.459	1.107	0.582-7.500	17
	Eutric Fluvisols	0.160	0.141	0.004-0.900	18
	Orthi-thionic Fluvisols	0.463	0.435	0.020-2.560	17
Lettuce	Podzolic	1.503	0.283	0.364-2.895	20
Tomato	Podzolic	0.716	0.100	0.361-1.191	18
Cucumber	Podzolic	1.812	0.191	1.577-2.301	8
Carrot	Podzolic	0.701	0.388	0.590-0.811	3
White radish	Podzolic	0.544	0.256	0.446-0.652	3
Potato	Podzolic	0.712	0.614	0.563-0.997	3

The decrease of the availability of radionuclides in soil with existing time plays an important role with radionuclide TF. By using the method described in our previous paper [14], the variations of mobile form of ^{60}Co, ^{65}Zn, ^{85}Sr and ^{134}Cs have been investigated [15]. Our results showed that equilibrium time took about 50 days for ^{85}Sr and 3 months for the rest radionuclides. TF values given here are "equilibrium data".



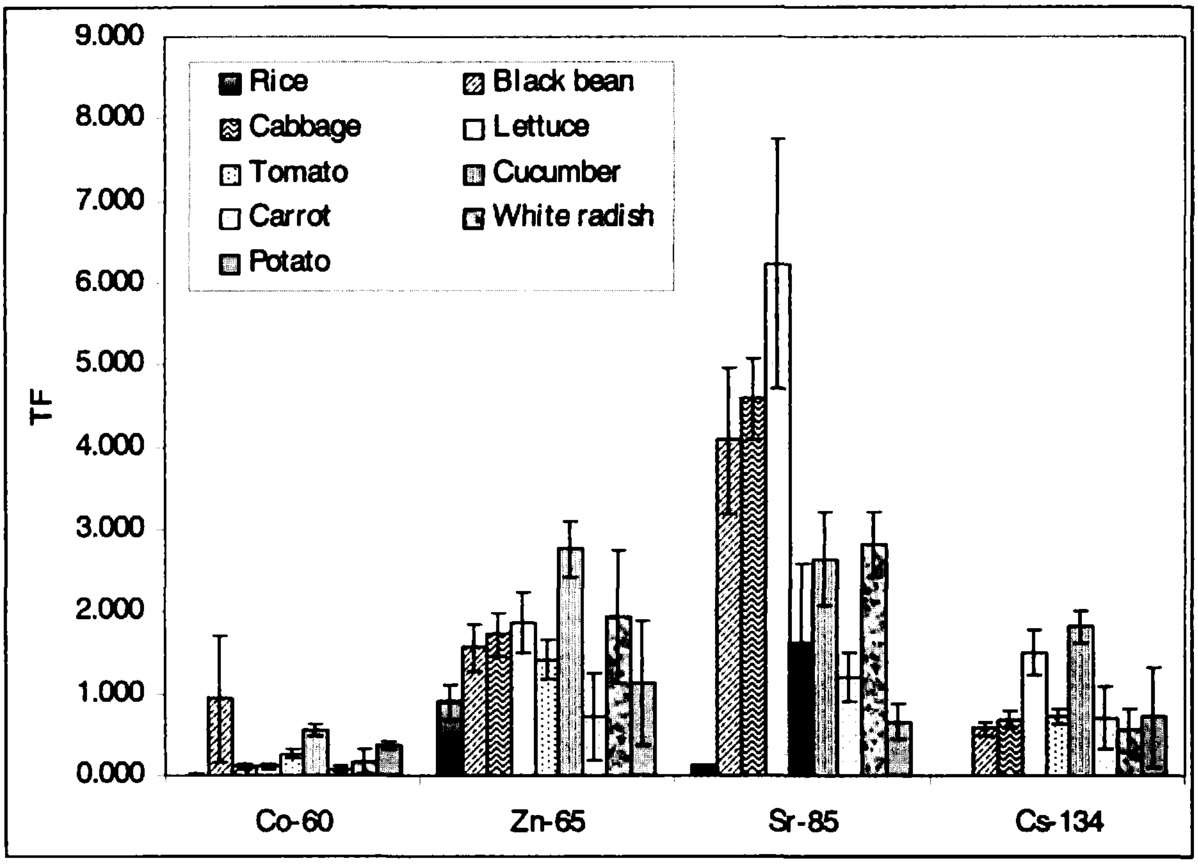


Fig. 3. Average TF derived from pot experiments.

Factor analysis and stepwise multiple regression analysis:

Factor analysis and stepwise multiple regression analysis have been performed to understand some correlations between the measured TF and soil parameters. The results of the analysis are presented in Table 7.

Table 7. The fitted model equations of TF over each kind of soil, plant class and radionuclide.

Nuclide	Plant class	Type of soil	TF equation of the fitted model	R^2
^{60}Co	Black bean	Podzolic	-2.7786 - 5.0 × 10^{-5} × Nucl. Conc.- 0.084 × Ex.K + 0.747 × Ex.Ca	0.514
	Cabbage	Podzolic	-0.1893 - 0.1 × 10^{-5} × Nucl. Conc.- 0.002 × Ex.K + 0.066 × Ex.Ca	0.533
	Lettuce	Podzolic	-0.1813 + 0.8 × 10^{-5} × Nucl. Conc.- 0.014 × Ex.K + 0.040 × Ex.Ca	0.187
	Tomato	Podzolic	0.5612 - 0.9 × 10^{-5} × Nucl. Conc.- 0.004 × Ex.K - 0.009 × Ex.Ca	0.647
^{65}Zn	Black bean	Podzolic	-0.9866 - 0.1× 10^{-5} × Nucl. Conc.+ 0.022 × Ex.K + 0.416 × Ex.Ca	0.570
	Cabbage	Podzolic	1.6075 + 1.4 × 10^{-5} × Nucl. Conc.+ 0.118 × Ex.K - 0.095 × Ex.Ca	0.320
	Lettuce	Podzolic	0.3914 + 2.3 × 10^{-5} × Nucl. Conc.- 0.031 × Ex.K + 0.147 × Ex.Ca	0.659
	Tomato	Podzolic	1.8365 + 0.4 × 10^{-5} × Nucl. Conc.- 0.059 × Ex.K - 0.056 × Ex.Ca	0.196
^{85}Sr	Rice	Ferralitic Ferralic Acrisols Eutric Fluvisols Orthi-thionic Fluvisols	0.3464 - 0.2 × 10^{-5} × Nucl. Conc. + 0.006 × Ex.K - 0.008 × Ex.Ca - 0.017 x pH - 0.033 × OM	0.715
	Black bean	Podzolic	6.3262 - 1.7 × 10^{-5} × Nucl. Conc. + 0.525 × Ex.K - 0.581 × Ex.Ca	0.283
	Cabbage	Ferralitic Ferralic Acrisols Eutric Fluvisols Orthi-thionic Fluvisols	-7.2287 - 7.4 × 10^{-5} × Nucl. Conc. - 1.394 × Ex.K - 1.243 × Ex.Ca + 5.537 × pH - 3.140 × OM	0.743
	Lettuce	Podzolic	2.5236 + 16.7 × 10^{-5} × Nucl. Conc. + 0.247 × Ex.K + 0.006 × Ex.Ca	0.715
	Tomato	Podzolic	-6.9539 + 3.2 × 10^{-5} × Nucl. Conc. + 0.780 × Ex.K + 0.627 × Ex.Ca	0.521
^{134}Cs	Rice	Ferralitic Ferralic Acrisols Eutric Fluvisols Orthi-thionic Fluvisols	2.8553 - 2.3 × 10^{-5} × Nucl. Conc. + 0.052 × Ex.K - 0.016 × Ex.Ca - 0.288 × pH + 0.013 × OM	0.972

Nuclide	Plant class	Type of soil	TF equation of the fitted model	R^2
	Black bean	Podzolic	$0.2876 + 0.9 \times 10^{-5} \times$ Nucl. Conc. $+ 0.068 \times$ Ex.K $- 0.014 \times$ Ex.Ca	0.216
	Cabbage	Ferralitic Ferralic Acrisols Eutric Fluvisols Orthi-thionic Fluvisols	$-5.127 - 1.0 \times 10^{-5} \times$ Nucl. Conc. $+ 0.048 \times$ Ex.K $- 0.269 \times$ Ex.Ca $+ 0.803 \times$ pH $- 0.227 \times$ OM	0.575
	Lettuce	Podzolic	$-1.1848 + 5.4 \times 10^{-5} \times$ Nucl. Conc. $+ 0.058 \times$ Ex.K $+ 0.227 \times$ Ex.Ca	0.425
	Tomato	Podzolic	$1.4324 - 0.2 \times 10^{-5} \times$ Nucl. Conc. $- 0.056 \times$ Ex.K $- 0.077 \times$ Ex.Ca	0.450

In general, there is a strong relationship between TF and some soil parameters (exchangeable K, exchangeable Ca, CEC, OM, pH and radionuclide concentration). Many correlation coefficients are higher than 0.700.

4. CONCLUSIONS

The measured Vg values (cm/s) are in the range of 0.01 - 1.84 for ^{7}Be and 1.95 - 49.77 for ^{137}Cs. In general, the Vg values are higher than those obtained by other authors in other locations. This demonstrates one of the most relevant features of cold air masses during the winter monsoon periods at our observation location. The results of fitting in linear model described the relationship between Vg_^{7}Be & H_mean and between Vg_^{137}Cs & U_mean.

The obtained data on TF show that there are no systematic differences between TF in other countries and TF in Vietnamese tropical environment.

456 data on TF and relevant parameters collected in the past years in Vietnam could be suitable to insert into the TF data bank of the Region.

There are many strong relationships between TF values and soil parameters (exchangeable K, exchangeable Ca, CEC, OM, pH and radionuclide concentration) in the cases of ^{60}Co TF for black bean, cabbage, tomato; ^{65}Zn TF for black bean, lettuce; ^{85}Sr TF for rice, cabbage, lettuce, tomato; ^{134}Cs TF for rice, cabbage. There are some insignificant relationships between TF values and soil parameters in the cases of ^{60}Co TF for lettuce; ^{65}Zn TF for cabbage, tomato; ^{85}Sr TF for black bean; ^{134}Cs TF for black bean.

Because the TF often depend on a large number of environmental variables, the experiments on the TF should last for a longer time to generate a larger number of TF with the observations of additional parameters as the relevant stable isotopes and to form more suitable TF.

5. ACKNOWLEDGEMENTS

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