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Prediction of Tritium Behavior in Rice Plant
after A Short-Term Exposure of HTO

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Abstract

In many Asian countries including Korea, rice is a very important food crop. Its grain is consumed by humans and its straw is used to feed animals. Because four CANDU reactors are in operation in Korea, relatively large amounts of tritium are released into the environment and the dose by these tritium in the rice plant must be estimated. Since 1997, KAERI (Korea Atomic Energy Research Institute) has carried out experimental studies to obtain domestic data on various parameters related to the direct tritium contamination of plant. But the analysis of the tritium behavior in the rice plant has been insufficient. In this study, the behavior of the tritium in the rice plant is predicted and compared with the measurement performed at KAERI. Using the conceptual model of the soil-plant-atmosphere tritiated water transport system which was suggested by Charles E. Murphy, transient tritium concentrations in soil and leaves were predicted. If the effect of tritium concentration in the soil is taken into account, the tritium concentration in leaves can be described by a double exponential model, however if the tritium concentration in the soil is disregarded, the tritium concentration in leaves can be described by a single exponential term like other relevant models e.g. UFOTRI or STAR-H3 model. The results can be used to predict the tritium concentration in the rice plant near the plant site and to estimate the ingestion dose after the release of tritium to the environment.

1. Introduction

The two primary forms of gaseous tritium releases are tritium gas(HT) and tritiated water vapor(HTO). HTO has higher potential for human radiological dose than HT. Tritium is known to be rapidly transported through the environment and is rapidly taken up by various organisms. In evaluating the total dose assessment around the Wolsong NPP, the portion of tritium has been considered to take about 70% in total exposure. Generally, tritium behavior in organisms is divided into two types, tissue free water tritium (TFWT) and organically bound tritium (OBT) [1]. HTO is mostly relevant to the incorporation of tritium in living organisms and forming organically bound tritium(OBT). OBT exhibits longer residence time in organisms than tritiated water. Quantitatively, the most important process of OBT production after a tritium release into the environment is photosynthesis with HTO as a precursor [2]. So the understanding of tritium behavior in plants is very important for dose assessment in the environment.

In many Asian countries including Korea, rice is a major food crop. Its grain is consumed by humans and its straw is used to feed animals[3]. HTO is easily absorbed by plant leaves from soil and air, but it is also easily removed from them through leaching process with tritium free water or drying up.

The ingestion dose due to an accidental or incidental short-term tritium release can be dominated by the consumption of wheat and rice if tritium is absorbed to leaves and accumulated in grains during the grain-filling period until harvest [4]. Since 1997, KAERI (Korea Atomic Energy Research Institute) has carried out experimental studies to obtain domestic data on various parameters concerning the direct tritium contamination of plant. But the analysis of the tritium behavior in the rice plant has not been sufficient. So, in this study, using the conceptual model of the soil-plant-atmosphere tritiated water transport system which was suggested by Charles E. Murphy, transient tritium concentrations in the soil and in leaves were derived.

2. Conceptual model of the soil-plant-atmosphere tritiated water transport system

Tritiated water concentration in the vegetation is of interest because of the

potential of human uptake by consumption. Vegetation also serves as the primary path for the incorporation of tritium into organic compounds which can move up the food chain. The vegetation system, illustrated in Fig. 1, has three compartments: the atmosphere, the plant leaves, and the soil. Because bulk flow dominates the transport through the conducting system of the plant roots and stem, this part of the plant can be treated as a time delay in the path between the soil and the leaves. The flow of tritiated water can be described in terms of the concentrations in compartments and the transporting processes of tritiated water between compartments. Tritiated water in the soil moves into the leaf by bulk flow through the water-conducting vessels. This water comes into contact with the atmosphere at the thin film in the walls of the mesophyll cells lining the substomal cavity. Transport from inside of the leaf to the atmosphere is done by vapor diffusion through small pores(stoma) in the water-impervious cuticle covering the leaf. The flow toward the leaf is proportional to the tritiated water vapor concentration of the air and the diffusion resistances in the path through the atmosphere, stoma, and substomal cavity. Water flow from the soil into the plant takes place by absorption through the roots and is transported along a water potential gradient through the plant vascular system into the leaves. Tritium is transported by bulk flow with the water. The uptake is driven by evaporation within the plant leaves. Evaporation is essentially the same process as described for tritiated water exchange between the atmosphere and the wet surfaces of the leaf cells.

In most cases, the tritium concentration in the soil water is the result of washout of tritium from the atmosphere. Because of the relatively low rate of diffusion between the atmosphere and the soil, dry deposition of tritiated water or tritiated hydrogen to the soil only becomes significant when the atmospheric concentrations of these compounds are very high compared to the concentration in rainfall. Washout is proportional to the tritiated water vapor concentration and the diffusion resistances associated with the air and surface of the individual rain drops.

The system of equations which describe this system are as follows:

$$V_v \frac{dCv}{dt} = A_v \left(\frac{C_a \rho_{wa}}{1.05r} - \frac{0.9 \rho_{wv} C_v}{1.05r} + \frac{\rho_{wv} - \rho_{wa}}{r} C_s \right) \tag{1}$$

$$D \frac{d}{dt}(\theta C_s) = \beta C_a R_i - \alpha \theta C_s - \frac{\rho_{wv} - \rho_{wa}}{r} C_s \tag{2}$$

$$D \frac{d\theta}{dt} = R_i - \frac{\rho_{wv} - \rho_{wa}}{r} - \alpha \theta \tag{3}$$

Where

V_v : leaf water content (g)

A_v : leaf area (cm^2)

C_v : the tritiated water vapor concentration in the leaves (Bq/l)

C_a : the tritiated water vapor concentration in the air (Bq/l)

C_s : the tritiated water concentration in the soils (Bq/l)

ρ_{wv} : Saturation vapor concentration in the leaves (g/cm^3)

ρ_{wa} : Saturation vapor concentration in the air (g/cm^3)

r : diffusion resistance in the leaves (sec/cm)

α : The proportional constant of the soil water conductivity

β : washout coefficient

θ : the soil water content(g/cm^3)

R_i : the infiltrated precipitation ($g/cm^2 - sec$)

D : the rooting depth of the vegetation (cm)

For the soil-plant-atmosphere tritium transport, the average leaf tritium concentration for seasonal or annual periods can be estimated by the steady-state concentration under average air tritium concentration and climatic conditions. The steady-state concentration can be quantitatively described by setting the left hand sides of the equations (1), (2), and (3) to zero. Notice that the steady-state value of tirtiated water concentration does not depend on the volume of any part of the system, but only on the relative magnitudes of the inputs and outputs. This set of equations can be simplified to show the relationship between the air concentration and the leaf concentration of tritiated water [5].And the ratio of diffusion resistances for water and HTO is nearly constant at 0.95[6]

$$\frac{C_v}{C_a} = \frac{0.95\rho_{wa}}{0.9\rho_{wv}} + \frac{1}{0.9}\left(1 - \frac{\rho_{wa}}{\rho_{wv}}\right)\beta \tag{4}$$

The concentration of tritiated water in the leaf is somewhat lower than the atmospheric concentration. The ratio of leaf to atmospheric concentration depends on the ratio of the atmospheric water vapor concentration to the vapor concentration at the wet leaf cell surfaces. If leaf temperature is equal to atmospheric temperature, this vapor concentration ratio is identical to the relative humidity of the air. Using the equation of (4), the concentration of tritium in the pine needle around the Wolsung NPP was predicted and compared with the measurement. The results are shown in Table. 1

and Fig. 2, respectively. [7, 8]

3. The prediction of tritium concentration in leaves of rice plant including soil effect.

Above equations of (1),(2),and (3) are derived based on the assumption of the experiment which was carried out in KAERI.[9]

Assumptions

1. During a growing period, rice plant is covered by irrigation water so the soil water content θ is constant.

2. When the time is zero, concentrations of C_s and C_v are also zero

3. During a short- term exposure of HTO, tritium concentration in the air is constant and after a exposure its concentration is zero.

That is,

$$
\begin{aligned}
0 \leq t < t_{ex} \quad & C_a(t) = C_{a1} \\
t \geq t_{ex}, \quad & C_a(t) = 0
\end{aligned}
\tag{5}
$$

According to the assumption 1, the equation of (3) becomes as

$$
R_i - \alpha\theta = \frac{\rho_{wv} - \rho_{wa}}{r}
\tag{6}
$$

and if this equation is put in the equation of (2)

$$
\frac{dC_s}{dt} + \frac{R_i C_s}{D\theta} = \frac{\beta R_i}{D\theta} C_a
\tag{7}
$$

① $0 \leq t < t_{ex}, \quad C_a = C_{a1}$

$$
\frac{dC_s}{dt} + \frac{R_i}{D\theta} C_s = \frac{\beta R_i}{D\theta} C_{a1}
\tag{8}
$$

Using integral factor, above equation is solved and arranged as follows

$$
C_s(t) = \beta C_{a,1} + A_1 \cdot e^{-\frac{R_i}{D\theta} t}
\tag{9}
$$

when $t = 0, \quad C_s = 0$

$$
\therefore C_s(t) = \left(1 - e^{-\frac{R_i}{D\theta} t}\right) \beta C_{a1}
\tag{10}
$$

② $t \geq t_{ex}$, $C_a = 0$ so the equation of (8) is arranged as

$$C_s = A_2 \cdot e^{-\frac{R_i}{D\theta} \cdot t}$$ (11)

Here if the boundary condition($t = t_{ex}$, $j = k$) is applied, tritium concentration in the soil with time becomes as

$$C_s(t) = \begin{cases} \beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta} \cdot t_{ex}}\right) & 0 \leq t < t_{ex} \\ \\ \beta C_{a1}\left(e^{-\frac{R_i}{D\theta} \cdot t_{ex}} - 1\right) \cdot e^{-\frac{R_i}{D\theta} \cdot t} & t \geq t_{ex} \end{cases}$$ (12)

To obtain the tritium concentration in the leaves of rice plant, the equation of (1) is now rearranged as

$$\frac{dC_v}{dt} = \left(\frac{A_v}{V_v}\right)\frac{C_a \rho_{wa}}{1.05r} - \left(\frac{A_v}{V_v}\right)\frac{0.9\rho_{wv}}{1.05r} \cdot C_v + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wv} - \rho_{wa}}{r}\right)\cdot C_s$$ (13)

and tritium concentration in the soil has been known so

③ $0 \leq t < t_{ex}$, $$\begin{cases} C_a(t) = C_{a1} \\ C_s(t) = \beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta} \cdot t}\right) \end{cases}$$ (14)

Using the equation of (13) and (14) , the equation of (15) can be expressed as

$$\frac{dC_v}{dt} + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{0.9\rho_{wv}}{1.05r}\right)\cdot C_v = \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wa}}{1.05r}\right)\cdot C_{a1} + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wv} - \rho_{wa}}{r}\right)\left\{\beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta} \cdot t}\right)\right\}$$ (15)

Where

$$K_1 = \left(\frac{A_v}{V_v}\right)\left(\frac{0.9\rho_{wv}}{1.05r}\right), \quad K_2 = \left(\frac{A_v}{V_v}\right)\left(\frac{\rho_{wa}}{1.05r}\right)C_{a1}, \quad K_3 = \left(\frac{A_v}{V_v}\right)\left(\frac{\rho_{wv} - \rho_{wa}}{r}\right)\cdot\beta C_{a1}$$ (16)

and then the equation of (15) is solved by using integral factor so

$$C_v(t) = \frac{K_2}{K_1} + \frac{K_3}{K_1} - \frac{K_3}{\left(K_1 - \dfrac{R_i}{D\theta}\right)} \cdot e^{-\frac{R_i}{D\theta} \cdot t} + A_3 \cdot e^{-K_1 \cdot t} \tag{17}$$

A_3 is obtained by the initial condition ($t = 0$, $C_v = 0$) and the equation of (17) is expressed as

$$C_v(t) = \left(\frac{K_2 + K_3}{K_1} - \frac{K_3}{\left(K_1 - \dfrac{R_i}{D\theta}\right)} \right) \left(e^{-\frac{R_i}{D\theta} \cdot t} - e^{-K_1 \cdot t} \right) \tag{18}$$

④ $t \geq t_{ex}$,
$$\begin{cases} C_a(t) = 0 \\ C_s(t) = \beta C_{a1}\left(e^{-\frac{R_i}{D\theta} \cdot t_{ex}} - 1 \right) \cdot e^{-\frac{R_i}{D\theta} \cdot t} \end{cases} \tag{19}$$

Using the equation of (13) and (19)

$$\frac{dC_v}{dt} + \left(\frac{A_v}{V_v} \right)\left(\frac{0.9\rho_{wv}}{1.05r} \right) \cdot C_v = \left(\frac{A_v}{V_v} \right)\left(\frac{\rho_{wv} - \rho_{wa}}{r} \right)\beta C_{a1}\left(e^{\frac{R_i}{D\theta} \cdot t_{ex}} - 1 \right) e^{-\frac{R_i}{D\theta} \cdot t} \tag{20}$$

Here,

$$K_4 = K_3 \cdot \left(e^{\frac{R_i}{D\theta} \cdot t_{ex}} - 1 \right) \tag{21}$$

And then the equation of (20) is solved as

$$C_v(t) = \frac{K_4}{\left(K_1 - \dfrac{R_i}{D\theta}\right)} \cdot e^{-\frac{R_i}{D\theta} \cdot t} + A_4 \cdot e^{-K_1 \cdot t} \tag{22}$$

A_4 is obtained by using the boundary condition ($t = t_{ex}$, $l = m$)

$$\therefore A_4 = \frac{K_2 + K_3}{K_1} e^{K_1 \cdot t_{ex}} - \left(\frac{K_3 + K_4}{K_1 - \dfrac{R_i}{D\theta}} \right) e^{\left(K_1 - \frac{R_i}{D\theta}\right)t_{ex}} - \left(\frac{K_2 + K_3}{K_1} - \frac{K_3}{K_1 - \dfrac{R_i}{D\theta}} \right) \tag{23}$$

3. The prediction of tritium concentration in leaves of rice plant excluding soil effect.

If the soil effect is not considered, the equation of (1) is arranged as

$$\frac{dC_v}{dt} = \left(\frac{A_v}{V_v}\right) \cdot \left(\frac{\rho_{wa}}{1.05r}C_a - \frac{0.9\rho_{wv}}{1.05r}C_v\right) \tag{24}$$

⑤ $0 \leq t < t_{ex}$, the equation of (24) is solved as

$$C_v(t) = \frac{K_2}{K_1} + A_5 \cdot e^{-K_1 \cdot t} \tag{25}$$

Here,

$$\frac{K_2}{K_1} = \frac{\left(\frac{A_v}{V_v}\right) \cdot \left(\frac{\rho_{wa}}{1.05r}\right) \cdot C_{a,1}}{\left(\frac{A_v}{V_v}\right) \cdot \left(\frac{0.9\rho_{wv}}{1.05r}\right)} = \frac{1}{0.9} \cdot \frac{\rho_{wa}}{\rho_{wv}} \cdot C_{a1} \tag{26}$$

Supposing that leaf temperature is equal to air temperature and initial condition is applied, the equation of (25) becomes as

$$C_v(t) = \frac{RH}{0.9} \cdot C_{a1} \cdot \left(1 - e^{-K_1 \cdot t}\right) \tag{27}$$

where, $RH =$ Relative Humidity

⑥ $t \geq t_{ex}$, the equation of (24) is solved as

$$C_v(t) = A_6 \cdot e^{-K_1 \cdot t} \tag{28}$$

Here, A_6 is obtained by applying boundary condition so the equation becomes as

$$C_v(t) = \frac{RH}{0.9}C_{a1}\left(e^{K_1 \cdot t_{ex}} - 1\right) \cdot e^{-K_1 \cdot t} \tag{29}$$

4. Results and Conclusion

As shown in the equation (16), K_1 is the elimination rate, with dimension of inverse of time and the reciprocal of A_v/V_v is the amount of water per unit area of leaf. The equation (27) which is solved by excluding the soil effect is described by a single exponential term as other models e.g. UFOTRI or STAR-H3 model [10, 11, 12]. The results derived by equations of (18), (22), (27), and (29) are shown from Fig. 3 to Fig. 11, and the input data used to the calculation are shown in Table 2. D3, D4, and D5 represent the time of which tritium exposure experiment was carried out by KAERI. When the elimination rate(K_1) which was obtained through above equations is about 1.6 hr^{-1}, the results is relatively well agree with measurement. Also when the infiltrated

precipitation (R_i) value is within the range of $10^{-5} \sim 10^{-6}$ ($g/cm^2 - \sec$), the results can be well agreed with measurement. Actually, at the early stage of tritium exposure, the major cause of tritium concentration in the leaves is the effect of tritium diffusion between the air and the leaves but the residual tritium concentration in the leaves after about 10 hours originate in the soil. Eventually, the tissue free water tritium (TFWT) loss in the plant is well described by a double exponential function rather than a single exponential function. As shown in Fig.3 or 11, the prediction value is lower than that of the measurement. The tritiated water which was absorbed into the soil from the atmosphere cannot be sunk into the underwater because this experiment was carried out not in the field but in the greenhouse. So this tritiated water may affect tritium concentration in the leaves successively.

ACKNOWLEDGEMENT

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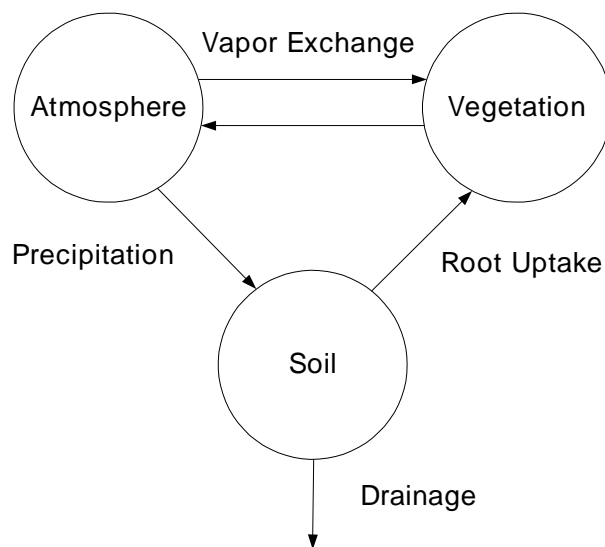
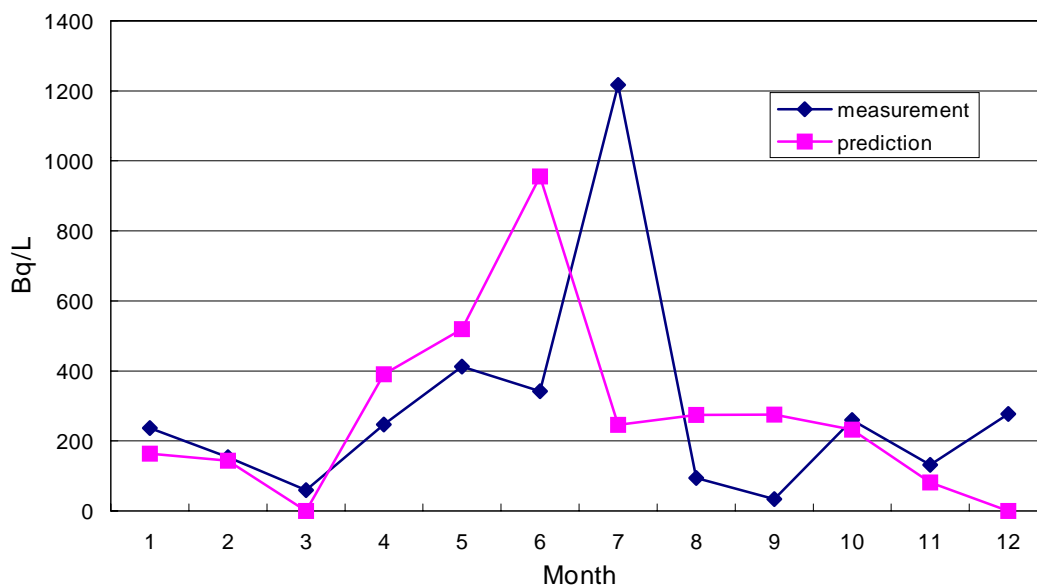Fig. 1. Conceptual model of the soil- plant- atmosphere tritiated water transport system

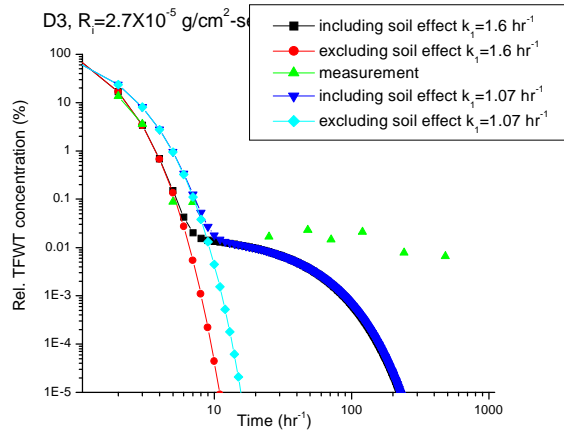Fig. 2.   Concentrations of TFWT around Choung Kyung company housing

Fig.3. Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D3)
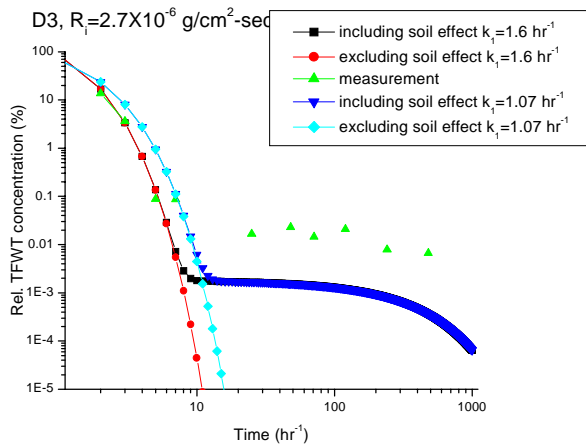

Fig.4. Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D3)
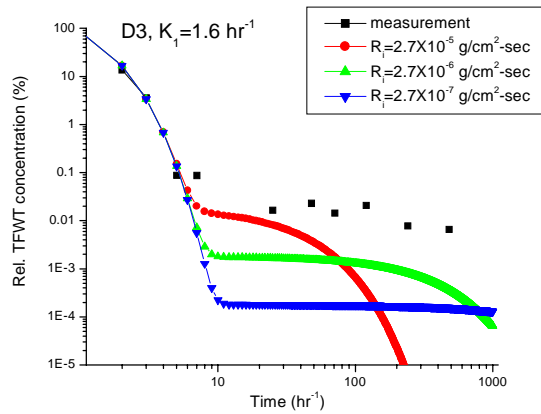

Fig.5. Changes of TFWT concentration in the different R_i with the lapse of time after its daytime exposure to atmosphere HTO (D3)
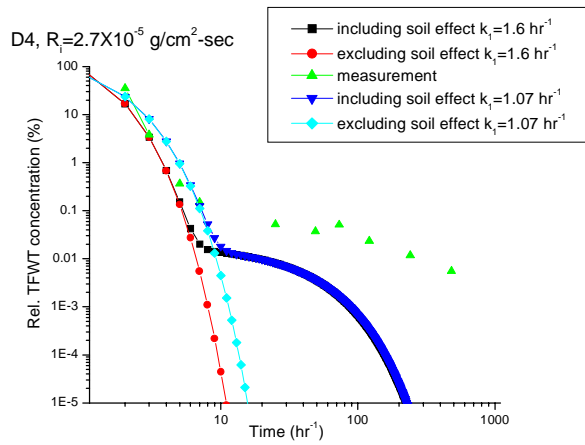
Fig.6.   Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D4)
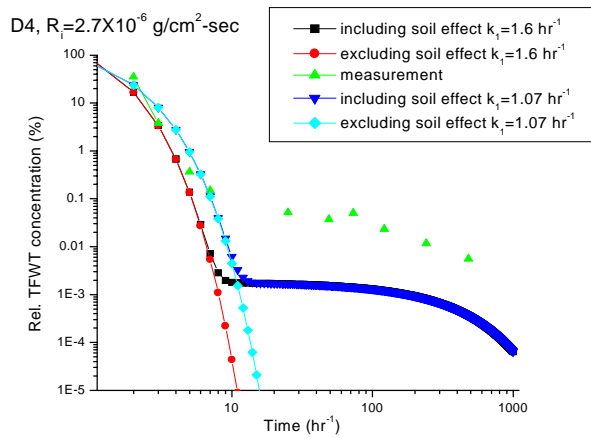

Fig.7.   Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D4)
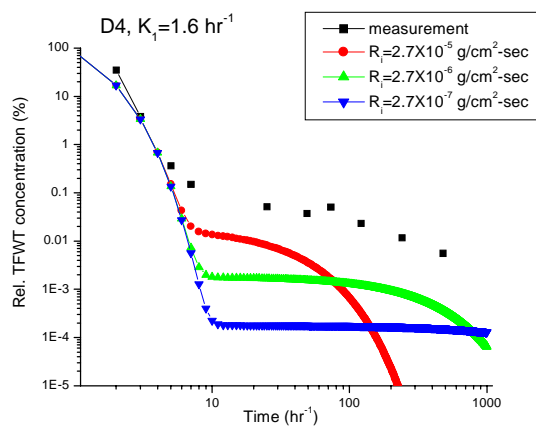

Fig.8.   Changes of TFWT concentration in the different R_i with the lapse of time after its daytime exposure to atmosphere HTO (D4)
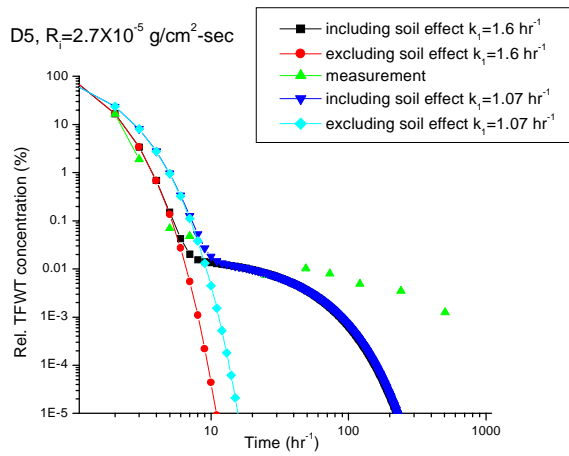
Fig.9. Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D5)
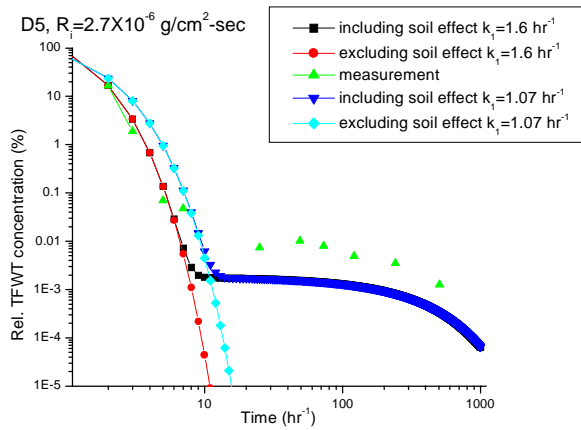

Fig.10. Changes of TFWT concentrations in the different K_1 with the lapse of time after its daytime exposure to atmosphere HTO (D5)
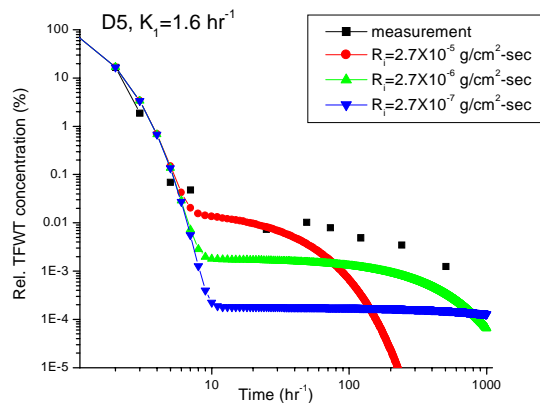

Fig.11. Changes of TFWT concentration in the different R_i with the lapse of time after its daytime exposure to atmosphere HTO (D5)

Table 1. Concentrations of TFWT in the pine needle around Choung Kyung company housing (Bq/L)

Month	Measured value	Predicted value
1	237	163.27
2	154	143.16
3	59.2	-
4	247	390.70
5	412	519.12
6	342	955.19
7	1217	246.14
8	94	274.54
9	33.7	274.81
10	260	231.91
11	132	81.13
12	277	-

Table 2. Input data used to the calculation

	D3			D4			D5		
V_v	1.0			1.0			1.0		
A_v	100			100			100		
C_{a1}	8.1310 X 10^7			5.5634 X 10^7			7.4173 X 10^7		
$Temp.$	30.4			30.7			30.2		
r	6		9	6		9	6		9
β	0.08			0.08			0.08		
θ	0.3			0.3			0.3		
R_i	2.7X10^{-5}	2.7X10^{-6}	2.7X10^{-7}	2.7X10^{-5}	2.7X10^{-6}	2.7X10^{-7}	2.7X10^{-5}	2.7X10^{-6}	2.7X10^{-7}
D	10			10			10		
RH	94.2			89.5			89.7		