A STUDY ON THE TRITIUM BEHAVIOR IN THE RICE PLANT AFTER A SHORT-TERM EXPOSURE OF HTO

Dae-Sik Yook, and Kun Jai Lee
Department of Nuclear Engineering, Korea Advanced Institute of Science and Technology
373-1, Kusong-dong, Yusong-gu, Taejon, 305-701, Korea
(FAX) 82-42-869-3810 (TEL) 82-42-869-3858
linkone@nuchen.kaist.ac.kr, kunjailee@cais.kaist.ac.kr

Yong-Ho Choi
Nuclear Environment Research Division, Korea Atomic Energy Research Institute, P.O. Box 105, Yusong-gu, Taejon, 305-600, Korea

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. In Korea, there are four CANDU type reactors that release relatively large amounts of tritium into the environment. Since 1997, KAERI (Korea Atomic Energy Research Institute) has carried out the experimental studies to obtain domestic data on various parameters concerning the direct contamination of plant. In this study, the behavior of 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, tritium concentrations in the soil and in leaves to time were derived. If the effect of tritium concentration in the soil is considered, the tritium concentration in leaves is described as a double exponential model. On the other hand if the tritium concentration in the soil is disregarded, the tritium concentration in leaves is described by a single exponential term as other models (e.g. Belot's or STAR-H3 model). Also concentration of organically bound tritium in the seed is predicted and compared with measurements. The results can be used to predict the tritium concentration in the rice plant at a field around the site and the ingestion dose following the release of tritium to the environment.

INTRODUCTION

Tritium, the heaviest and only radioactive isotope of hydrogen, was discovered in 1939 by Alvarez and Cornog, and subsequent work established its physical half-life at 12.3 years. The nuclide, ^{3}H or T, decays to form ^{3}He by emission of a beta particle with a maximum energy of 18 KeV and an average energy of 5.7 KeV.(1) 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 in Korea, the portion of tritium has been considered to take about 70% of total exposure. Generally, tritium behavior in organisms is divided into two types, tissue free water tritium (TFWT) and organically bound tritium (OBT). (2) HTO is mostly relevant to the incorporation of tritium in living organisms and forming organically bound tritium. 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. (3) 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. (4) HTO is easily absorbed by plant leaves from soil and air, and 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 from the grain-filling period to harvest .(5) 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. (6)

CONCEPTUAL MODEL OF THE SOIL-PLANT-ATMOSPHERE HTO 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 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. 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. 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 can describe this system are as follows:

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

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

$$D \frac{d\theta}{dt} = R_i - \frac{\rho_{wv} - \rho_{wa}}{r} - \alpha\theta \quad \text{(Eq.3)}$$

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 ($g/cm^2 \cdot \sec$)

D : the rooting depth of the vegetation (cm)

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

Among the above equations of (Eq.1), (Eq.2), and (Eq.3) are derived based on the assumptions of the experiment which was carried out in KAERI.(7)

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, respectively.

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,

$$0 \le t < t_{ex} \quad C_a(t) = C_{a1}$$
$$t \ge t_{ex}, \qquad C_a(t) = 0 \tag{Eq.4}$$

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

$$R_i - \alpha\theta = \frac{\rho_{wv} - \rho_{wa}}{r} \tag{Eq.5}$$

and if this equation is put in (Eq.2)

$$\frac{dC_s}{dt} + \frac{R_i C_s}{D\theta} = \frac{\beta R_i}{D\theta} C_a \tag{Eq.6}$$

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

$$\frac{dC_s}{dt} + \frac{R_i}{D\theta} C_s = \frac{\beta R_i}{D\theta} C_{a1} \tag{Eq.7}$$

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

$$C_s(t) = \beta C_{a,1} + A_1 \cdot e^{-\frac{R_i}{D\theta} \cdot t} \tag{Eq.8}$$

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

$$\therefore C_s(t) = \left(1 - e^{-\frac{R_i}{D\theta} \cdot t}\right) \beta C_{a1} \tag{Eq.9}$$

② $t \ge t_{ex}, \quad C_a = 0$ so (Eq.7) is arranged as

$$C_s = A_2 \cdot e^{-\frac{R_i}{D\theta} \cdot t} \tag{Eq.10}$$

Here if the boundary condition($t = t_{ex}$, ①=②) is applied, tritium concentration in the soil to time becomes as

$$C_s(t) = \begin{cases} \beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta} \cdot t_{ex}}\right) & 0 \le t < t_{ex} \\ \beta C_{a1}\left(e^{\frac{R_i}{D\theta} \cdot t_{ex}} - 1\right) \cdot e^{-\frac{R_i}{D\theta} \cdot t} & t \ge t_{ex} \end{cases} \tag{Eq.11}$$

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

$$\frac{dC_v}{dt} = \left(\frac{A_v}{V_v}\right)\frac{C_a \rho_{wa}}{1.05r} - \left(\frac{A_v}{V_v}\right)\frac{0.9\rho_{wv}}{1.05r} \cdot C_v + \left(\frac{A_v}{V_v}\right) \cdot \left(\frac{\rho_{wv} - \rho_{wa}}{r}\right) \cdot C_s \tag{Eq.12}$$

and tritium concentration in the soil has been known as (Eq.11) so

③ $0 \le t < t_{ex}$, $\begin{cases} C_a(t) = C_{a1} \\ C_s(t) = \beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta}\cdot t}\right) \end{cases}$ (Eq.13)

Using (Eq.12) and (Eq.13) , (Eq.14) can be derived as

$$\frac{dC_v}{dt} + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{0.9\rho_{wv}}{1.05r}\right)\cdot C_v = \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wa}}{1.05r}\right)\cdot C_{a1} + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wv}-\rho_{wa}}{r}\right)\left\{\beta C_{a1}\left(1 - e^{-\frac{R_i}{D\theta}\cdot t}\right)\right\} \quad \text{(Eq.14)}$$

Where

$$K_1 = \left(\frac{A_v}{V_v}\right)\left(\frac{0.9\rho_{wv}}{1.05r}\right), \quad K_2 = \left(\frac{A_v}{V_v}\right)\left(\frac{\rho_{wa}}{1.05r}\right)C_{a1}, \quad K_3 = \left(\frac{A_v}{V_v}\right)\left(\frac{\rho_{wv}-\rho_{wa}}{r}\right)\cdot\beta C_{a1} \quad \text{(Eq.15)}$$

and then (Eq.14) is solved by using integral factor as

$$C_v(t) = \frac{K_2}{K_1} + \frac{K_3}{K_1} - \frac{K_3}{\left(K_1 - \frac{R_i}{D\theta}\right)}\cdot e^{-\frac{R_i}{D\theta}\cdot t} + A_3\cdot e^{-K_1\cdot t} \quad \text{(Eq.16)}$$

A_3 is obtained by the initial condition ($t = 0, \quad C_v = 0$) and (Eq.16) is expressed as

$$C_v(t) = \left(\frac{K_2 + K_3}{K_1} - \frac{K_3}{\left(K_1 - \frac{R_i}{D\theta}\right)}\right)\left(e^{-\frac{R_i}{D\theta}\cdot t} - e^{-K_1\cdot t}\right) \quad \text{(Eq.17)}$$

④ $t \ge t_{ex}$, $\begin{cases} C_a(t) = 0 \\ C_s(t) = \beta C_{a1}\left(e^{\frac{R_i}{D\theta}\cdot t_{ex}} - 1\right)\cdot e^{-\frac{R_i}{D\theta}\cdot t} \end{cases}$ (Eq.18)

Using (Eq.12) and (Eq.18), TFWT concentrations is derived as

$$\frac{dC_v}{dt} + \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{0.9\rho_{wv}}{1.05r}\right)\cdot C_v = \left(\frac{A_v}{V_v}\right)\cdot\left(\frac{\rho_{wv}-\rho_{wa}}{r}\right)\beta C_{a1}\left(e^{\frac{R_i}{D\theta}\cdot t_{ex}} - 1\right)e^{-\frac{R_i}{D\theta}\cdot t} \quad \text{(Eq.19)}$$

Where

$$K_4 = K_3\cdot\left(e^{\frac{R_i}{D\theta}\cdot t_{ex}} - 1\right) \quad \text{(Eq.20)}$$

And then (Eq.21) is solved as

$$C_v(t) = \frac{K_4}{\left(K_1 - \frac{R_i}{D\theta}\right)}\cdot e^{-\frac{R_i}{D\theta}\cdot t} + A_4\cdot e^{-K_1\cdot t} \quad \text{(Eq.21)}$$

A_4 is obtained by using the boundary condition ($t = t_{ex}$, ③=④)

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

(Eq.22)

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

If the soil effect is not considered, (Eq.1) is rearranged as

$$\frac{dC_v}{dt} = \left(\frac{A_v}{V_v} \right) \cdot \left(\frac{\rho_{wa}}{1.05r} C_a - \frac{0.9\rho_{wv}}{1.05r} C_v \right) \quad \text{(Eq.23)}$$

⑤ $0 \le t < t_{ex}$, (Eq.23) is solved as

$$C_v(t) = \frac{K_2}{K_1} + A_5 \cdot e^{-K_1 \cdot t} \quad \text{(Eq.24)}$$

Here,

$$\frac{K_2}{K_1} = \frac{\left(\frac{A_v}{V_v} \right) \cdot \left(\frac{\rho_{wa}}{1.05r} \right) \cdot C_{a,1}}{\left(\frac{A_v}{V_v} \right) \cdot \left(\frac{0.9\rho_{wv}}{1.05r} \right)} = \frac{1}{0.9} \cdot \frac{\rho_{wa}}{\rho_{wv}} \cdot C_{a1} \quad \text{(Eq.25)}$$

Supposing that leaf temperature is equal to air temperature and initial condition is applied, (Eq.24) becomes as

$$C_v(t) = \frac{RH}{0.9} \cdot C_{a1} \cdot \left(1 - e^{-K_1 \cdot t} \right) \quad \text{(Eq.26)}$$

Where, $RH =$ Relative Humidity

⑥ $t \ge t_{ex}$, (Eq.23) is solved as

$$C_v(t) = A_6 \cdot e^{-K_1 \cdot t} \quad \text{(Eq.27)}$$

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

$$C_v(t) = \frac{RH}{0.9} C_{a1} \left(e^{K_1 \cdot t_{ex}} - 1 \right) \cdot e^{-K_1 \cdot t} \quad \text{(Eq.28)}$$

(Eq.26) and (Eq.28) which are solved by excluding the soil effect are described by a single exponential term as other models (e.g. Belot's or STAR-H3 model). (8,9,10)

The Prediction of OBT concentration in leaves

HTO is short-lived in plants and much that enters vegetation never reaches human diet. Plants, however, convert it to organic compounds in which form it can be longer lasting and available to humans. Rates of formation of OBT and the environmental factors controlling them are important in determining doses to humans following releases of tritium to the environment. OBT once formed, is subsequently lost by oxidation. In addition to actual loss of tritium with time, remaining OBT is being diluted by aggregation of new plant material. Conversion of HTO to OBT will be described by simple model as followings;

$$\text{leaf HTO} \xrightarrow{K_t} \text{leaf OBT} \xrightarrow{K_e} \text{Loss} \qquad \text{(Eq.29)}$$

so that

$$\frac{dC_{obt}(t)}{dt} = \mathrm{K}_t C_v(t) - K_e C_{obt}(t) \qquad \text{(Eq.30)}$$

Because the equation for TFWT concentration in leaves with time is (Eq.17) and (Eq.21) respectively, the equation for OBT concentration in leaves also should be calculated separately with time.

⑦ when $0 \le t < t_{ex}$

In equation of (Eq.17), constant term is very complex so it can also be replaced as following;

$$K_5 = \left(\frac{K_2 + K_3}{K_1} - \frac{K_3}{\left(K_1 - \frac{R_i}{D\theta} \right)} \right) \qquad \text{(Eq.31)}$$

if this equation of (Eq.17) is inserted into (Eq.30) and the initial condition is used, the equation for OBT concentration are obtained as follows;

$$C_{OBT}(t) = \frac{K_t \cdot K_5}{\left(K_e - \frac{R_i}{D\theta} \right)} \cdot \left(e^{-\frac{R_i}{D\theta} \cdot t} - e^{-K_e \cdot t} \right) - \frac{K_t \cdot K_5}{(K_e - K_1)} \cdot \left(e^{-K_1 \cdot t} - e^{-K_e \cdot t} \right)$$

(Eq.32)

⑧ when $t \ge t_{ex}$, The equation for OBT concentration in leaves are also can be calculated by using the equation (Eq.21) and (Eq.30).

$$C_{OBT}(t) = \frac{K_t \cdot K_4}{\left(K_1 - \frac{R_i}{D\theta} \right)\left(K_e - \frac{R_i}{D\theta} \right)} \cdot e^{-\frac{R_i}{D\theta} \cdot t} + \frac{K_t \cdot A_4}{(K_e - K_1)} \cdot e^{-K_1 \cdot t} + A_8 \cdot e^{-K_e \cdot t}$$

(Eq.33)

Where

$$A_6 = \frac{K_t}{\left(K_e - \frac{R_i}{D\theta}\right)} \cdot e^{\left(K_e - \frac{R_i}{D\theta}\right) \cdot t_{ex}} \left(K_5 - \frac{K_4}{\left(K_1 - \frac{R_i}{D\theta}\right)} \right) - \frac{K_t}{\left(K_e - K_1\right)} \cdot e^{(K_e - K_1) \cdot t_{ex}} \cdot (K_5 + A_4) + K_t \cdot K_5 \left(\frac{1}{\left(K_e - K_1\right)} - \frac{1}{\left(K_e - \frac{R_i}{D\theta}\right)} \right)$$

(Eq.34)

RESULTS AND CONCLUSION

As shown in (Eq.15), K_1 is a 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. (Eq.26) which is solved by excluding the soil effect is described by a single exponential term as other models e.g. Belot's or STAR-H3 model .(8,9,10) The results derived by (Eq.17) and (Eq.21) are shown from Fig. 1 to Fig. 2, and the input data used to the calculation are shown in Table I. D3, D5, and N1 represent the time of which tritium exposure experiment was carried out by KAERI.(7) Also D means day exposure and N means night exposure. When the elimination rate(K_1) of the day exposure which was obtained through above equations is about 1.6 hr^{-1}, the results is relatively well agree with measurement. And the elimination rate of night exposure is about 0.33 hr^{-1}. Also when the infiltrated precipitation (R_i) value is within the range of 10^{-5}~10^{-6} ($g/cm^2 \cdot sec$), the results are well agreed with measurement. Actually, at the early stage of tritium exposure, the major cause of tritium concentration in leaves is the effect of tritium diffusion between the air and the leaves but the residual tritium concentration in 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.1 or 2, 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.

(Eq.32) and (Eq.33) show the OBT concentration that is converted from HTO and lost. These equations are described by three exponential terms and the results are shown from Fig.3 to Fig.4. Because measured OBT concentration in leaves had not measured except exposure experiments of D1 and D, only measurement values of D1 and D2 are shown in Fig.3 and Fig.4. But these values can be used to compare with predicted value in order to confirm a decrease tendency and order of magnitude of OBT concentration in leaves. As shown in Fig.3 and Fig. 4, OBT concentration in leaves is not dependent on the tritium concentration in soil but dependant on the transfer rate (k_t) and loss rate (k_e), and these rates of the day exposure are 10 times higher than that of night exposure. The obtained transfer rate of day exposure in this study has a similar value to compare with other literature cited. (11)

While the TFWT concentration in leaves is decreased at about $10^{-4} \sim 10^{-5}$ compared with initial concentration in leaves, OBT concentration is only decreased at about 10^{-1} compared with initial concentration. Although the produced quantity of OBT in leaves is small, its residence time in organisms is relatively longer than that of TFWT. So in view of radiological impact assessment in the environment, the effect of OBT concentration in leaves will be very larger than that of TFWT.

ACKNOWLEDGEMENT

This work was supported by the Korea Science and Engineering Foundation (KOSEF) through the Innovative Technology Center for Radiation Safety (iTRS), Hanyang University.

REFERENCE

1. NCRP, "Tritium in the environment", No. 62, (1979)
2. Y.BELOT, "Tritium in plants: a review." Radiation Protection Dosimetry, p101-105, (1986)
3. Silvia Diabate, and Siegfried Strack, "Organically Bound Tritium." Health Physics, **65**(6), p.698-712, (1993)
4. Y.H.Choi, S.B.Kim, and K.M. Lim, "Incorporation into organically bound tritium and the underground distribution of HTO applied to a simulated rice field." journal of Environmental Radioacitivity, **47**,p.279-290, (2000)
5. Sang-Bog Kim, Myung-Ho.Lee, and Gun-Sik Choi, "Investigation into trtium Behaviour in Chinese cabbage and Rice after a Short-term Exposure of HTO" J.Korean Asso. Radiat.,**23**(2),p.75-82,(1998)
6. Charles E. Murphy, Jr., "Tritium Transport and Cycling in the Environment" Health Physics, **65**(6):p.683-697, (1993)
7. KAERI, "Development of Environmental Radiation Protection Technology-Radioecological Studies on Terrestrial Food Chain Analysis for Accidental Release"(in Korea), (1999)
8. W.Raskob, "Description of the New Version 4.0 of the Tritium Model UFOTRI Including User Guide, KERNFORSCHUNGSZENTRUM KARLSRUHE -PUBLICATIONS- KFK",(1993)
9. A.J.P. Brudenell, C.D. Collins,and G.Shaw, "Dynamics of Tritiated Water (HTO) Uptake and Loss by Crops After Short-Term Atmospheric Release", J. Environmental Radioactivity,**36**,p.197-218, (1997)
10. Y.BELOT, D.G., H.CAMUS, and C.CAPUT, "Prediction of the Flux of Tritiated Water from Air to Plant Leaves" Health Physics,**37**,p.575-583.(1979)
11. Peter Barry et al, "BIOMOVS II (BIOspheric MOdel Validation Study – Phase II)", **8**

Table I. Input Data used for Calculation

	D3			D5			N1		
V_v	1.0			1.0			1.0		
A_v	100			100			100		
C_{a1}	8.1310×10^7			7.4173×10^7			8.1310×10^7		
Temp.	30.4			30.2			22.1		
r	6	9		6	9		15	18	
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			89.7			91.9		
K_t	7.0×10^{-4}			7.0×10^{-4}			7.0×10^{-5}		
K_e	3.0×10^{-4}			3.0×10^{-4}			3.0×10^{-5}		

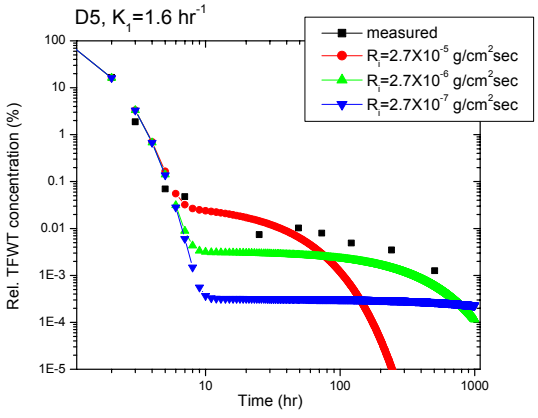

Fig. 1. Changes of TFWT concentrations in the different R_i with the lapse of time after its daytime exposure to atmosphere HTO

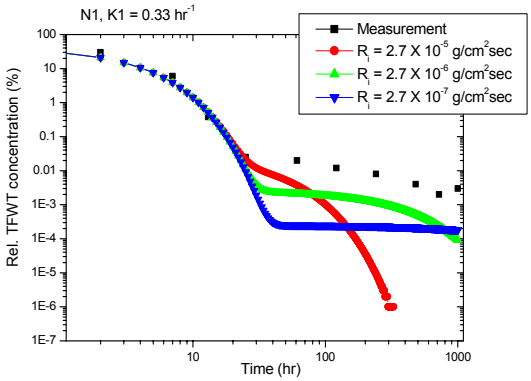

Fig. 2. Changes of TFWT concentrations in the different R_i with the lapse of time after its night time exposure to atmosphere HTO

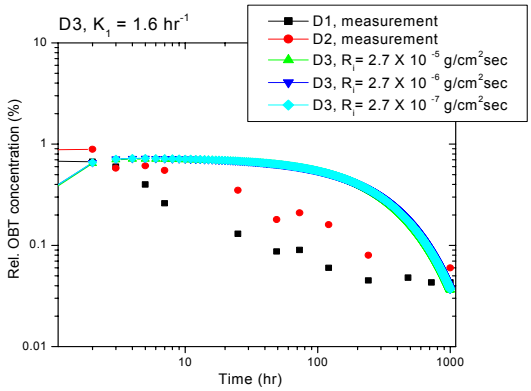

Fig. 3. Changes of OBT concentrations in the different R_i with the lapse of time after its daytime exposure to atmosphere HTO

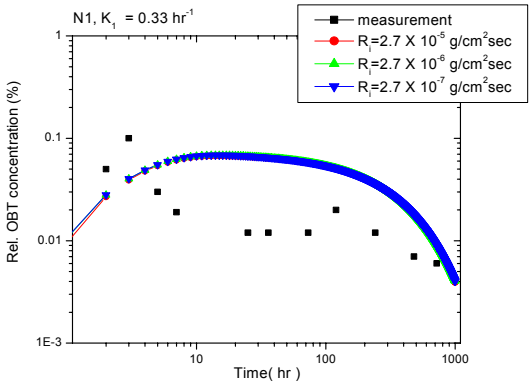

Fig. 4. Changes of OBT concentrations in the different R_i with the lapse of time after its night time exposure to atmosphere HTO