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Comparison of the Binding Strength of Hydrogen in Grain Near Wolsung Nuclear Power Plants

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ABSTRACT

Because the Tissue Bound Tritium of food irradiates the organic tissues of a man during a longer time than the Tissue Free Water Tritium, we found the ratio of labile and bound hydrogen, which is the direct source of TBT concentration, in grain such as rice and barley.

Tissue free water was extracted from rice and barley sampled, adjacent to Wolsung nuclear power plants of CANDU type, by freeze-drying. Tissue bound water was taken from some of the dried samples by high-pressure combustion. The other of the samples was washed by tritium-free water for 2-3 hours, and dried again by freeze-drying. Tissue bound water was taken again from some of the second dried samples by the combustion. The extracted tissue free and bound waters were distilled and TFWT and TBT concentrations of them were counted by a liquid scintillation counter.

Through alternating washing, drying and combustion until the concentration of TBT would be constant, the tritium concentration existing as bound hydrogen was found. The ratios of labile and bound hydrogen of rice and barley were determined by TFWT concentration, initial TBT concentration and bound tritium concentration. The ratios of bound hydrogen of rice and barley were 0.55, 0.60 relatively.

I. INTRODUCTION

In general, food consists of free water and organic component. Hydrogen isotopes in food are distributed in two compartments. Tritium of tissue free water is called TFWT. Tritium of organic component is called TBT, Tissue Bound Tritium, or OBT, Organically Bound Tritium. Recently, interest has been focused on TBT of food, because TBT irradiates the organic human tissue for a longer time than TFWT (Etnier et al. [1], Diabate and Strack [2], Inoue et al. [3]).

Hydrogen in organic component is separated of labile hydrogen, by external water vapor or washing water, and bound hydrogen (Travis et al. [4]). The effect of dose of tritium will not be equal according to the state of food. For instance, the cases of uptake directly the fresh food, uptake the food washed several times, and uptake the food cooked are not equal relatively because of the differences of tritium existing in the food.

II. MODEL

The state of existing of hydrogen atoms in food can be expressed with the figure 1. The hydrogen in food is divided the hydrogen in the free water of food, the labile hydrogen and the bound hydrogen. The free water is easily movable into the inside or the outside of food. Of the organic components, the labile hydrogen generally is bound to O, N and S atoms and exchangeable with hydrogen isotopes in the external water. The bound hydrogen of them is bound to C atom and not exchangeable except metabolism.

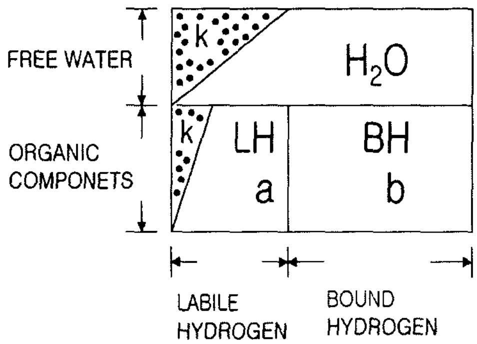

Fig. 1. Concept of taking up tritium into food compartments from the tritiated water vapor in the air. A and b is relative ratio of LH and BH in the figure, a + b = 1.

When food is exposed the tritiated water vapor, it will diffuse into food or be taken up to food by exchange the tritium with labile hydrogen in food. In this case, the gradient of tritium concentration is established from the surface of food to the center. Otherwise, when food is exposed the tritium-free water vapor, the exchangeable tritium comes out rapidly from food.

To simplify these phenomena, we assume that the weight of food is not exchanged during uptake. The proportion of exchanging hydrogen isotopes with labile hydrogen in the organic component or with tissue free water is representative as k (<1) in the figure 1. The tritium concentrations of food or T/H atom ratios before taking up tritium are denoted C_{W0}, C_{L0} and C_{B0} for the free water, labile and bound hydrogen in food. We assume that the tritium concentrations of the compartments before uptake are equal.

$$C_{W0} = C_{L0} = C_{B0} \quad (1)$$

, where C_{W0} is TFWT concentration of food. When the ratios of labile and bound hydrogen in dried food are denoted a and b,

$$a + b = 1 \quad (2)$$

The TBT concentration of food is that

$$C_{D0} = a\, C_{L0} + b\, C_{B0} = (a + b)\, C_{W0} = C_{W0} \quad (3)$$

The tritium concentrations of free water and labile hydrogen in food after taking up the tritiated water vapor from the outside are that

$$C_{W1} = k\, C_A + (1\text{-}k)\, C_{W0} \tag{4}$$

$$C_{L1} = k\, C_A + (1\text{-}k)\, C_{L0} \tag{5}$$

, where C_A is the tritium concentration of tritiated water vapor in the air. If the concentration of TBT taken through high pressure combustion of dried food after uptake is denoted C_{D1}, then, from equations (3), (4) and (5),

$$\begin{aligned} C_{D1} &= a\, C_{L1} + b\, C_{B1} \\ &= a\, C_{W1} + b\, C_{B1} \end{aligned} \tag{6}$$

Thus

$$a = (C_{D1} - b\, C_{B1}) / \quad C_{W1} \tag{7}$$

After washing the dried sample several times and drying, the concentration of TBT taken is denoted C_{DW}. Then C_A can be changed to C_{LO} in equation (5), and

$$\begin{aligned} C_{DW} &= a\, C_{LO} + b\, C_{B1} \\ b\, C_{B1} &= C_{DW} - a\, C_{LO} \end{aligned} \tag{8}$$

From equations (7) and (8),

$$a = (C_{D1} - C_{DW}) / (C_{W1} - \quad C_{LO})$$

, where $C_{LO} = C_{WO}$ from equation (1), and if $C_{W1} \gg C_{WO}$, then

$$a = (C_{D1} - C_{DW}) / \quad C_{W1} \tag{9}$$

In this equation, we can measure the concentration of TFWT in food, C_{W1}, the concentration of TBT in dried food before washing, C_{D1} and the concentration of TBT in dried food gotten rid of the tritium existing as labile hydrogen through washing several times, C_{DW} in the experiments. Therefore, the ratios of labile and bound hydrogen, a and b can be taken by equations (2) and (9) (Inoue et al., [3]).

III. EXPERIMENT

In this paper, rice and barley was sampled from Naa-ri, in which Wolsung nuclear power plant of CANDU type is located. The free water of samples was extracted by freeze-drying apparatus. It consists of a rotary pump for making vacuum of 1×10^{-2} Torr and a freezer for cooling a cold trap under -40 ℃ to trap water vapor of them. The freeze-dried samples were powdered by a grinder. The diameter is about 1 mm. The tissue bound hydrogen of dried samples was extracted as water form from some of them (~ 50 g) by high pressure combustion equipment, Parr 1121, which bomb them under atmosphere of oxygen of 300 psi (~ 20 atm).

The other of them was mixed with tritium-free water, which is under background level of 1 cpm, 1 equally using a stainless spoon and left for 2~3 hours under seal. After this washing process of them, they were filtered with a glass micro fiber filter and dried by freeze-drying again. The tissue bound water of some of them was extracted by high-pressure combustion again. By repeating the washing process several times, the TBT of samples in the washing step was extracted.

The tissue free water of 50 ml was distilled simply with mixing $KMnO_4$ 0.1 g and Na_2O_2 0.1 g. The tissue bound water of ~20 ml was mixed with $KMnO_4$ ~0.04 g and neutralized with Na_2O_2. Then, It was purified by distillation. The distillates weighing 8 g were put into Teflon vials of 20 ml and mixed with 12 g of liquid scintillator, HiSafe 3 (Wallac co.) to make the total volume of 20 ml. The concentrations of TFWT and TBT of samples were counted for 500 minutes respectively by a liquid scintillation counter. It was Quantulus 1220 (Wallac co.), its counting efficiency was 26 % and the background level was 1 cpm.

IV. RESULTS AND DISCUSSION

The TBT concentrations of rice and barley sampled adjacently to Wolsung nuclear power plant were measured with repetition of washing with tritium-free water. The TFWT concentrations of rice and barley were 88.16 Bq/L, 129.4 Bq/L. The TBT concentration of the dried rice and the dried barley before washing in the figure 2 was 122.4 Bq/L, 92.22 Bq/L. The TBT concentration of the dried rice after the first washing was decreased nearly to a constant level in the figure 2. This means that the tritium existing as labile hydrogen in the dried rice was exchanged almost with tritium-free water used for the first washing. The TBT concentrations of rice after the first washing were included the range of the relative error of 7.7 %. The TBT concentration of the dried barley was decreased nearly to a constant level after the second washing. The TBT concentrations of barley after the second washing were included the range of the relative error of 5.6 %. From the figure 2, the concentrations of bound tritium of rice and barley, $C_{DW, RICE}$, $C_{DW, BARLEY}$ were 83.12 Bq/L, 41.12 Bq/L.

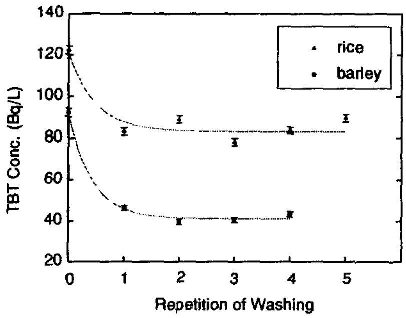

Fig. 2. Concentrations of TBT with repetition of washing with tritium-free water

From the results, the Specific Activity Ratio, which is the ratio of TBT concentration to TFWT concentration, of rice and barley was 1.4 and 0.71. This is included the range of 0.7~1.7 of the average SAR in food of several countries (Hisamatsu et al. [5]; Hisamatsu et al. [6]).

The ratios of labile and bound hydrogen of them were determined from the concentrations of TFWT, of the initial TBT and of TBT of the dried samples washed completely tritium in labile hydrogen of organic component. In the table 1, the ratio of labile hydrogen of rice was 0.446. This is alike to the results of labile hydrogen of rice, which were 0.40 (from U.S.A.) and 0.38 (from India), of the other paper. The ratio of the labile and bound hydrogen of barley were 0.395, 0.605. These results can be the basic data for determining the internal dose of TBT in rice and barley.

Table 1. Ratios of labile and bound hydrogen of rice and barley.

Samples	Cwl (Bq/L)	CD1 (Bq/L)	CDW (Bq/L)	A	B
Rice	88.16	122.4	85.71	0.446	0.554
Barley	129.4	92.22	41.16	0.395	0.605

V. CONCLUSION

In this experiment, we found the ratio of labile and bound hydrogen, which is the direct source of TBT concentration which irradiates the organic tissues of a man during a longer time than TFWT when a man take food, in grain such as rice and barley.

The rice and barley, which were the major diet of Korean, were sampled adjacently to Wolsung nuclear power plant of CANDU type and measured the TFWT and TBT concentration of them. The TBT concentrations were determined after washing completely tritium existing as labile hydrogen of food by washing effect. From the results, the SARs of rice and barley was 1.4 and 0.71. The ratios of the labile and bound hydrogen were 0.446 and 0.554 for rice, and 0.395 and 0.605 for barley.

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