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青菜-土壤生态系统中氚水的迁移与分布动态

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摘要：采用模拟污染物的同位素示踪技术研究了两种引入方式(模拟灌溉和降雨)下氚水在青菜-土壤模拟生态系统中的迁移与分布动态。测定了植物和土壤样品中两种形态氚(自由水氚和结合态氚)的比活度。以探明氚水在青菜-土壤系统中的行为特性、不同引入方式对青菜吸收和积累氚水的影响。结果显示：引入土壤中的氚水，不仅在系统各分室间转移和分配，而且迅速向系统外散逸；以自由水氚和结合态氚形式存在于青菜组织中，以吸湿性水氚和结晶水氚存在于土壤，其中自由水氚(或吸湿性水氚)远大于结合态氚(或结晶水氚)；青菜茎中的总氚比活度高于叶和根中的比活度；运用示踪动力学分室模型原理对实验数据进行拟合得：土壤中的比活度 C_s(模拟灌溉) $=91.59e^{-0.1002t}+99.91e^{-9.1105t}$ 和 C_s(模拟降雨) $=78.09e^{-0.5115t}+48.27e^{-0.0475t}$；青菜植株中的比活度 C_v(模拟灌溉) $=543.52(e^{-0.1002t}-e^{-9.1105t})$ 和 C_v(模拟降雨) $=647.07e^{-0.5115t}+241.28e^{-0.0475t}$；方差分析结果表明各回归方程较好地反应了氚水在青菜-土壤系统中的变化动态。

关键词：放射生态学；氚水；青菜；迁移与分布；非线性回归

The Migration and Distribution of Tritium Water in Chinese Cabbage (*Brassica chinensis*)-Soil Ecosystem

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Abstract: In this study, tritium water was administered into simulated Chinese cabbage (*Brassica chinensis*)-soil system in mode of simulated irrigation and rainfall to study its migration and distribution in each component of the system. The objective of this study are: (1) to evaluate accumulation and distribution of tritium water in Chinese cabbage when treated with different method; (2) to clarify distribution of tritium water in soil profile.

Four concrete pools with dimensions of 1m×1m×0.3m were constructed outdoor. Two hundred ninety kg of soil with moisture content of 21.74% was filled into each pool. And soil depth was about 24 cm. Thirty Chinese cabbage seedlings, 8～10 cm in height, were transplanted into each pool. 3200 mL of tritium water with a specific activity of 1.7355×10^4 Bq/mL was introduced into each pool 30 days after transplanted. In two pools, tritium water was sprayed on foliage surface to simulate rainfall; and in the other two pools, it was sprayed on soil surface to simulate irrigation. The samples were collected at the time intervals of 0.25, 1, 3, 7, 14, 21 and 28 days after treatment. Three seedlings were collected from each pool, the roots were rinsed with tap water, and the water remaining on their surface was removed with an absorbent paper. They were divided into root, petiole and foliage. And then they were cut into small pieces after measuring the weight of each part. Twenty grams samples from each part were put into 100ml matrass for free water tritium distilled. In the meantime, three soil columns were collected from

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each pool with soil sampler. Each soil column was sectioned into three equal parts, each part was pulverized, mixed thoroughly, and then 30 g of soil sample was weighed into 100ml matrass for tritium of hygroscopic distilled. Every sample had 3 replications.

The free water tritium and tritium of hygroscopic were distilled using constant temperature distillation, And the bound tritium and crystalline water using oxidation burning method. The radioactivity was measured with a liquid scintillation spectrometer (Wallac 1400DSA). The counting error was controlled to be lower than 5%. The counting data were calibrated with counting efficiency, distilling reclaim ratio, burning reclaim ratio and any other factors.

The open two-compartment system model and non-linear regression analysis method were used to simulate the dynamics of tritium water in Chinese cabbage-soil system, and fitting equations were established at 95% confidence level.

The results showed that when tritium water administered into simulated Chinese cabbage-soil system in mode of simulated irrigation and rainfall, it not only was transferred to other component of the system, but also vaporized into atmosphere rapidly. Both free water tritium and bound tritium were found in Chinese cabbage. Tritium of hygroscopic and crystalline water consist in the soil. The specific activity of free water tritium(or tritium of hygroscopic water) was bigger than that of bound tritium(or tritium of crystalline water). The specific activity of total tritium in root, petiole and foliage of Chinese cabbage reached 305.66 Bq/g, 553.98 Bq/g and 439.00Bq/g only 6 hours after administered with simulated irrigation method. The specific activity increased with time, then it decreased gradually when it reached the maximum value(338.24Bq/g, 650.15Bq/g and 439.00Bq/g). The specific activity in petiole was the biggest in Chinese cabbage. In case of simulated rainfall, tritium water was not only taken up via stoma of above-ground parts, but also could be taken up by roots from soil. Thus, accumulation of tritium water in Chinese cabbage resulted in a greater specific activity than that in case of simulated irrigation. Variation in specific activity showed a similar tendency as that in simulated irrigation condition.

The compartment model of tracer kinetic was applied to imitate the experimental data. For dynamics of specific activity in whole plant and soil, it could be described with the following exponential regression equations respectively: the specific activity of soil C_s(simulate irrigation) $= 91.59e^{-0.1002t} + 99.91e^{-9.1105t}$ and C_s(simulate rainfall) $= 78.09e^{-0.5115t} + 48.27e^{-0.0475t}$; and the specific activity of Chinese cabbage C_v (simulate irrigation) $= 543.52(e^{-0.1002t} - e^{-9.1105t})$ and C_v (simulate rainfall) $= 647.07e^{-0.5115t} + 241.28e^{-0.0475t}$ were gained. The results of analysis of variance showed that each regression equation could described the dynamics of accumulation and disappearance of tritium water in Chinese cabbage-soil ecosystems preferably.

Key words: radioecology; tritium water; Chinese cabbage; migration and distribution; nonlinear regression

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有关核电站放射性排放物^{89}Sr、^{134}Cs、^{60}Co等核素在生态环境中的行为,尤其是迁移和累积等行为特性前人已作了较多的研究[1~5]。但对氚水在农业生态系,尤其是作物体中行为的研究国内外研究报道甚少[6,7],在青菜中行为的动态研究尚未报道。然而,为了实现核电的持续发展和保持高质量环境的完美统一,研究氚水的环境行为是一项极待开展的工作,本试验采用同位素示踪技术研究了氚水由模拟灌溉和模拟降雨两种方式引入青菜-土壤生态系统,其在系统各组份中的消长与分布动态,并运用具有相互交换的开放二分室模型[8]和非线性回归方法建立其动态行为的数学模型,以探明氚水在青菜-土壤系统中的行为特性、不同引入方式对青菜吸收和积累氚水的影响。为评价氚水对生态环境和农作物的影响提供科学依据。

1 材料与方法

1.1 氚水与土壤

氚水由中国原子能研究院同位素研究所提供，放射性比活度为 5.5536×10⁷Bq/ml，放射化学纯度大于95%，使用前稀释 10 倍得 5.5536×10⁶Bq/ml 的氚水。供试土壤（小粉土）采自浙江大学实验农场，使用前经粉碎，去除石块、植物根系等杂物。其主要理化性质见表 1。

表 1 供试土壤的主要理化性质

Table 1 Main Physico-chemical properties of the soil studied

有机质 Organic matter(g/kg)	$pH(H_2O)$ pH	阳离子交换量 CEC (cmol/kg)	砂粒 Sand(%)	粉粒 Silt(%)	粘粒 Clay(%)
7.9	6.5	3.28	66.3	31.2	2.5

1.2 试验方法

试验在 4 只 1m×1m×0.3m 的模拟池内进行。每池装小粉土 290kg（湿土，含水量 21.74%），土层厚度约 24cm，每池移栽青菜苗（品种抗热 605，株高约 8～10cm，由上海市蔬菜良种综合技术开发部提供）5×6株。待青菜苗成活长大（移栽后 1 个月）分别向叶表面（模拟降雨，2 只池重复）和土壤表面（模拟灌溉，2 只池重复）均匀喷洒比活度为 1.7355×10⁴Bq/ml 的氚水各 3200ml。于氚水引入后 0.25、1、3、7、14、21、28d 采样。试验期间的温度约为 25～30℃。

1.3 采样

每次采样从每池随机取青菜 3 株（同一处理合并共 6 株），根部用清水冲洗后用吸水纸吸去表面水，然后分根部、茎和叶 3 个部分，分别剪碎装入 100ml 梨形蒸馏瓶中（每种样品 3 个重复）秤量后待处理。用半筒式取土器每池随机取全土柱 3 条，均等分割为 3 段，分别装入 100ml 梨型瓶中（每种样品 3 个重复）。梨型瓶皆及时用塞子塞紧，以防蒸发损失。

1.4 自由水氚或吸湿性水氚提取

自由水氚或吸湿性水氚采用恒温蒸馏法提取[9]，方法如下：在 100ml 梨型瓶中装入待提取的样品（湿土约 30g，植物约 20g），然后在 130℃油浴（液状石腊）上蒸馏 8～10h，收集馏出水并分别吸取 0.8ml 注入装有 8ml 闪烁液和 2.5ml 乙二醇乙醚的测样瓶中。馏出水量由蒸馏前后样品的重量差得到。闪烁液配方为：7gPPO＋0.5gPOPOP＋乙二醇乙醚 300ml＋二甲苯 700ml。

1.5 结合态氚和结晶水氚提取

将蒸馏后的样品（蒸渣）置于 100℃的烘箱中以彻底去除残存的自由水氚或吸湿性水氚，然后称取一定量的蒸渣（植物 0.200g，土壤 1.000g）在 OX-600 型生物氧化燃烧仪上于 850℃下燃烧 3min，通氧量和清洗氮气的通气量皆约 350ml/min，接收液为上述闪烁液，以提取样品中的结合态氚和结晶水氚[9]。

1.6 测量

上述所有样品的放射性测量皆在 Wallac 1400DSA 液闪仪上测定，测量误差控制在 5%以内，测量结果经蒸馏或燃烧回收率校正[9]后换算成样品鲜样的比活度。

2 结果与分析

2.1 氚水在青菜-土壤系统中的消长与分布动态

氚水在青菜-土壤系统中的分布动态如表 2、表 3 所示。由表 2 可见，从表土引入（模拟灌溉）的氚水迅速向青菜迁移，并以自由水氚和结合态氚形式存在，结合态氚的形成和存在表明氚水与青菜组织中的氢发生了交换以及由于光合作用形成了氚标光合产物（结合态氚产物）。引入氚水后 6h，青菜根、茎和叶中的总氚（自由水氚和结合态氚之和）比活度便分别达到 305.66 Bq/g 、553.98 Bq/g 和 439.00Bq/g，其中根和茎中的比活度随时间逐渐增加，至第 3 天达最大值（338.24Bq/g 和 650.15Bq/g）后又趋下降；而叶中的比活度在 6h 时已达最大值，而后随时间逐渐降低。由于氚水的生物学性质类似于普通水，青菜通过根系的摄取而进入植株，又由于挥发和向土壤深层迁移，使得表层土壤中的氚水比活度逐渐降低，向青菜植株的转移速率相应减小，而青菜又具有较大的叶表面积和高的含水量，叶面水分蒸发量大，致使氚水在植株中积累

到最大值后又逐渐下降。从青菜各部位总氚比活度分布看，在初始阶段（0～1d），茎中最高，菜叶次之，根中最小，而至试验后期根中的比活度又超过了叶中的比活度。试验后期由于土壤中氚水的比活度已经较低，从根、茎输送到叶面的氚水远少于叶面蒸发掉的氚水，致使叶中的比活度下降速度大于根和茎，导致了在试验后期叶中的比活度低于根中的比活度。土壤中的氚水由于青菜吸收和自然挥发总的随时间逐渐下降；

表 2　青菜-土壤系统中氚水的比活度分布动态（模拟灌溉）

Table 2　The distribution dynamic of HTO specific activity in Chinese cabbage-soil ecosystem (Simulated irrigation)

时间 Time(d)	青菜 Chinese cabbage (Bq/g fresh sample)			土壤层 Soil profile (Bq/g wet soil)		
	根 Root	叶柄 Petiole	叶 Foliage	0～8cm	8～16cm	16～24cm
0.25	303.87	552.39	433.72	287.66	18.56	1.18
	(1.79)	(1.59)	(5.28)	(0.49)	(0.00)	(0.00)
1	329.30	581.62	391.66	213.88	72.33	4.33
	(3.63)	(2.95)	(4.52)	(0.78)	(0.00)	(0.00)
3	332.77	648.47	106.43	156.11	32.99	4.47
	(5.47)	(1.68)	(3.46)	(1.02)	(0.01)	(0.00)
7	167.74	285.62	89.76	98.30	33.99	6.15
	(5.48)	(4.35)	(4.23)	(1.14)	(0.00)	(0.00)
14	83.89	89.61	41.98	32.87	30.32	7.48
	(6.07)	(3.90)	(3.37)	(1.03)	(0.02)	(0.00)
21	51.30	81.91	40.40	20.51	15.58	4.52
	(4.43)	(3.78)	(3.22)	(1.49)	(0.02)	(0.00)
28	35.22	65.23	31.09	16.20	10.54	3.95
	(5.23)	(3.90)	(4.25)	(1.83)	(0.05)	(0.00)

* 表中数据为自由水或吸湿性水氚，括号内的数据为结合态或结晶水氚　Tritium of free or hygroscopic water in the table, Combined tritium or tritium of crystalline water between brackets

表 3　青菜-土壤系统中氚水的比活度分布动态（模拟降雨）

Table 3　The distribution dynamic of HTO specific activity in Chinese cabbage-soil ecosystem (Simulated rainfall)

时间 Time(d)	青菜 Chinese cabbage (Bq/g fresh sample)			土壤层 Soil profile (Bq/g wet soil)		
	根 Root	叶柄 Petiole	叶 Foliage	0～8cm	8～16cm	16～24cm
0.25	484.54	888.95	610.20	268.07	27.28	3.44
	(3.17)	(2.20)	(4.96)	(0.21)	(0.00)	(0.00)
1	353.26	733.07	446.52	209.34	68.84	4.53
	(5.13)	(1.80)	(5.41)	(0.85)	(0.05)	(0.00)
3	287.91	455.95	121.28	130.12	61.57	5.86
	(5.27)	(3.40)	(5.25)	(0.66)	(0.10)	(0.00)
7	123.11	257.86	82.58	61.85	33.84	6.46
	(3.96)	(3.10)	(7.13)	(1.33)	(0.22)	(0.00)
14	68.62	104.61	30.52	34.97	32.97	4.51
	(5.14)	(1.93)	(4.33)	(1.58)	(0.17)	(0.00)
21	64.37	96.85	24.40	27.30	21.92	2.59
	(7.45)	(2.05)	(3.21)	(2.26)	(0.21)	(0.00)
28	42.35	70.25	18.92	18.43	13.52	3.33
	(6.24)	(3.01)	(5.29)	(2.54)	(0.25)	(0.00)

* 表中数据为自由水或吸湿性水氚，括号内的数据为结合态或结晶水氚　Tritium of free or hygroscopic water in the table, Combined tritium or tritium of crystalline water between brackets

表土（0～8cm）中的总氚比活度随时间呈下降趋势；中层土壤（8～16cm）开始时随时间逐渐增加，至 1d 达最大值后又逐渐降低；底层（16～24cm）土壤的趋势与中层土壤相似，但迟第 14 天才达最大值。这是因为氚水引入表土后，由于不断向深层迁移、青菜吸收以及挥发，使得表土中的氚水也随之下降，而中层和底层土壤的比活度反而增加，达峰值后又趋下降。总体上看，自由水氚（或吸湿性水氚）的比活度远大于结合态氚（或结晶水氚）的比活度，表明氚水进入青菜和土壤后主要以自由水氚（或吸湿性水氚）的形式存在，只有很少量转化为结合态氚（或结晶水氚）。氚水在青菜中形成的结合态氚多于在土壤中形成的结晶水氚，其原因主要是氚水中的氚不仅与青菜组织中的氢发生了同位素交换，而且还参于光合作用形成同化产物而生成结

合态氚。

由表 3 可见，模拟降雨情况下，由于青菜不仅从根系而且还从叶面吸收氚水，使得其根、茎和叶中的比活度在 6h 时已达最大值，且在初始阶段（0～1d）其比活度大于模拟灌溉处理的青菜相应部位的比活度。土壤中氚水的比活度总体上略低于模拟灌溉时土壤中的比活度，这是由于在氚水引入总量相同时，实际进入土壤的氚水比模拟灌溉时略少一些。其他特性（比如氚水在青菜各部位的分布以及在土层中的变化动态等）与模拟灌溉处理的情况相类似。

2.2 氚水在青菜-土壤系统中的迁移和消长规律

氚水在青菜-土壤生态系统中的迁移与消长行为可运用具有相互交换的开放二分室模型描述，如图 1 所示，令各分室中总氚（自由水氚和结合态氚之和）的比活度对时间的变化率服从一级速率过程[8]。若以 q_s、C_s、m_s 和 q_v、C_v、m_v 分别表示土壤和青菜中总氚的总活度、比活度和质量；k_{sv}和 k_{vs}分别为氚水由土壤分室向青菜分室和由青菜分室向土壤分室转移的速率常数，k_{sa} 或 k_{va}为土壤或青菜分室氚水挥发损失的速率常数。则各分室中氚水总活度对时间的变化率为：

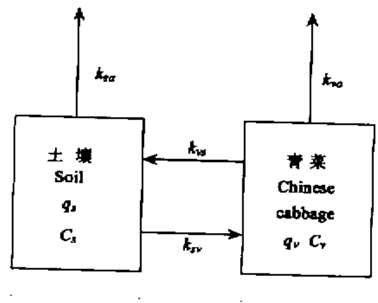

图 1　具有相互交换的开放二分室模型

Fig. 1　The open two-compartment system with interchange

$$\frac{\mathrm{d}q_s}{\mathrm{d}t} = k_{vs}q_v - (k_{sv} + k_{sa})q_s$$

$$\frac{\mathrm{d}q_v}{\mathrm{d}t} = k_{sv}q_s - (k_{vs} + k_{va})q_v$$

解此微分方程组，并注意到 $q_s = m_sC_s$、$q_v = m_vC_v$，并令 $k_1 = k_{sv} + k_{sa}$，$k_2 = k_{vs} + k_{va}$，便得：

$$C_s = \frac{m_vk_{vs}C_v(0)}{m_s(\delta - \gamma)}(e^{-\gamma t} - e^{-\delta t}) + \frac{C_s(0)}{\delta - \gamma}[(k_2 - \gamma)e^{-\gamma t} - (k_2 - \delta)e^{-\delta t}]$$

$$C_v = \frac{m_sk_{sv}C_s(0)}{m_v(\delta - \gamma)}(e^{-\gamma t} - e^{-\delta t}) + \frac{C_v(0)}{\delta - \gamma}[(k_1 - \gamma)e^{-\gamma t} - (k_1 - \delta)e^{-\delta t}]$$

式中，

$$\gamma = \frac{1}{2}\left[k_1 + k_2 + \sqrt{(k_1 + k_2)^2 - 4(k_1k_2 - k_{sv}k_{vs})}\right]$$

$$\delta = \frac{1}{2}\left[k_1 + k_2 - \sqrt{(k_1 + k_2)^2 - 4(k_1k_2 - k_{sv}k_{vs})}\right]$$

根据上述推导结果，表明土壤和青菜分室中的总氚比活度服从二项指数规律。对于模拟灌溉引入的生态系统，土壤中的初始比活度 $C_s(0) = 191.5$(Bq/g)，青菜中的初始比活度 $C_v(0) = 0$；而模拟降雨引入的系统，则有：$C_s(0) + (m_v/m_s)C_v(0) = 195.5$(Bq/g)。运用表 4、表 5 数据进行指数回归分析[10]，得青菜-土壤系统两分室总氚比活度的变化动态拟合方程式，对拟合方程进行方差分析[11]，计算在 95%置信度下的置信区间，结果列于表 6。从各拟合方程式与实验值的标准误差 σ 看（其值分别为 43.83Bq/g、6.59Bq/g、31.65Bq/g 和 8.28Bq/g），与青菜峰值比活度和土壤起始比活度的比率分别为 8.9%、3.4%、4.1%和 6.6%），表明各拟合方程较好地反应了氚水在青菜土壤系统中的消长动态。

表 4　生态系统中各分室总氚比活度（模拟灌溉）(Bq/g)

Table 4　The specific activity of each compartment total tritium in the ecosystem(Simulated irrigation)(Bq/g)

分室 Compartment	时间 Time(d)							
	0	0.25	1	3	7	14	21	28
青菜 Chinese cabbage	0.00	483.73	491.18	374.39	186.75	74.77	69.01	50.86
土壤 Soil	191.50	102.63	97.11	64.86	45.53	23.91	14.04	10.82

表 5 生态系统中各分室总氚比活度(模拟降雨)(Bq/g)

Table 5 The specific activity of each compartment total tritium in the ecosystem(Simulated rainfall)(Bq/g)

分室 Compartment	时间 Time(d) 0	0.25	1	3	7	14	21	28
青菜 Chinese cabbage	—	769.50	610.38	311.47	199.78	82.72	74.35	56.71
土壤 Soil	—	99.67	94.52	66.10	34.57	24.73	18.09	12.68

表 6 青菜-土壤系统中氚水的消长动态拟合方程

Table 6 Regression equation of accumulation and disappearance of tritium water in Chinese cabbage-soil ecosystem

引入方式 Induction method	分室 Compartment	拟合方程 Regression equation	置信水平 Believe level	置信区间 Believe interval
模拟灌溉 Simulated irrigation	青菜 Chinese cabbage	$C_v(t)=543.52(e^{-0.1002t}-e^{-9.1105t})$	95%	$C_v\pm87.66$
	土壤 Soil	$C_s(t)=91.59e^{-0.1002t}+99.91e^{-9.1105t}$	95%	$C_s\pm13.18$
模拟降雨 Simulated rainfall	青菜 Chinese cabbage	$C_v(t)=647.07e^{-0.5115t}+241.28e^{-0.0475t}$	95%	$C_v\pm63.30$
	土壤 Soil	$C_s(t)=78.09e^{-0.5115t}+48.27e^{-0.0475t}$	95%	$C_s\pm16.56$

3 结论

上述实验结果显示:(1) 氚水在引入青菜-土壤系统后,由于挥发有较大部分进入空气,其余部分以自由水氚和结合态氚形式存在于青菜组织中,以吸湿性水氚和结晶水氚存在于土壤,其中自由水氚(或吸湿性水氚)远大于结合态氚(或结晶水氚),结合态氚的形成和存在表明氚水与青菜组织中的氢发生了交换以及由于光合作用形成了氚标光合产物(结合态氚产物)。(2) 两种引入方式(模拟灌溉和模拟降雨),对氚水在青菜-土壤生态系统中行为特性的影响,除青菜组织中比活度达峰值的时间不同外,其余性质基本相拟。(3) 氚水在青菜-土壤系统中的消长行为可以采用具有相互交换的双库室开系统模型描述,经计算机拟合、方差分析,表明各回归方程较好地反应了氚水在青菜-土壤系统中的消长与迁移动态。

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