J. RADIAT. RES., 24, 326–338 (1983)

Influence of Chemical Forms on Iodine Uptake by Plant

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(Received May 29, 1983)

Iodine uptake/Chemical form/Plant

Uptake of iodide and iodate by komatsuna, *Brassica Rapa var. pervidis*, was studied using culture solution containing radioiodine as a tracer. After 24 hours' exposure, accumulation of iodide in the plant was several times higher than that of iodate. The concentrations of both iodide and iodate in roots were much higher than those in shoots. It was estimated that high accumulation of iodide in roots was due to the adsorption of the element on the root surface. The plant of the earlier growth stage absorbed iodide selectively from the solution. On the contrary that of the older growth stage, more than 40 days after seeding, showed less absorption of the element.

Influence of stable iodine content on the uptake of radioiodine was also examined. The transfer rate of radioactive iodide to roots decreased remarkably with increasing amount of stable iodide in the culture solution, whereas that to shoots remained relatively constant indicating that the element content in shoots increased almost proportionally with the increasing concentration of stable iodine in the solution.

A significant amount of iodate in the culture solution was transformed to iodide in the presence of plant. The dominant chemical form of iodine in the plant was thought to be iodide.

INTRODUCTION

Iodine is highly concentrated in the human thyroid gland. Therefore, from the radiation protection point of view, radioiodine is one of the notable radionuclides released from the stacks of nuclear facilities to the environment. The important pathways of radioiodine in the terrestrial environment to the human bodies are as follows:

1) atmosphere — pasture — cow — milk — man
2) atmosphere — leafy vegetable — man
3) atmosphere — soil — plant (agricultural products) — man

It is recognized that the former two pathways are significant for ^{131}I. Only recently attention has been payed to the artificially produced ^{129}I, which is introduced into the environment by low level chronic releases from spent fuel reprocessing plant. Because of its long half-life (1.6×10^7 years), it is expected that the nuclide will accumulate in soils and enter into plants. Therefore, the pathway, atmosphere — soil — plant — man, should not be ignored when estimating the amount of the nuclide ingested orally. However, only a little is known about the uptake of iodine by plant from soil.

The chemical composition of soil varies widely, and it is difficult to control the experimental conditions. Therefore we have used solution cultures for the preliminary experiment of iodine uptake through roots. Uptake of iodine by plants may be influenced by chemical, biological and meteorological conditions during their cultivation period. The chemical form of the element is thought to be one of the most important factors, because of the variety of its valence states and chemical instability[1]. It is commonly accepted that the major chemical forms in the terrestrial environmental water are iodide and iodate.

In this paper, uptake of iodide and iodate by plants has been studied using radioisotope tracer techniques. Studies on the chemical forms of the element in the culture solutions and in the plants were additionally carried out.

MATERIALS AND METHODS

Cultivation of the plant prior to the exposure experiments

Komatsuna, *Brassica Rapa var. pervidis*, was chosen for the current experiments, because this vegetable is widely consumed in Japan and it is suitable for growing in solution culture. The Hy-lizer, a commercially available nutrient solution, was used. The solution contains 98 mg total N/l, 15 mg P/l, 120 mg K/l, 24 mg Mg/l, 70 mg Ca/l, 32 mg S/l and small quantities of trace elements such as Fe, B and Mn[2]. The iodine content of this solution is less than 0.5 ngI/ml. Prior to the exposure experiments, the plants were cultivated in a growth chamber under an artificial day — night rhythm (9 — 15 hours). Illumination was provided by a combination of fluorescent lamps and electric bulbs, and the light intensity of daytime was about 15000 lux at leaf level. Temperature and humidity in the chamber were maintained at 25 ± 2℃ and 60 ± 10% during daytime, and 15 ± 2℃ and 60 ± 10% during nighttime.

Preparation of radioiodine stock solutions

Carrier free ^{131}I (NEZ-035A, lmCi ^{131}I in NaOH solution) was used for the exposure experiments as a tracer. The radioactive iodide($^{131}I^-$) stock solution (about 100 nCi/ml) was prepared by adding a small amount of sodium sulfite (about 0.1 ml of a 100 ppm solution) to the ^{131}I solution to reduce the various chemical forms of iodine to iodide, then it was diluted with de-ionized water. To prepare the radioactive iodate($^{131}IO_3^-$) stock solution (about 100 nCi/ml), 2 ml of bromine water (3%) were added to the ^{131}I solution for oxidation. It was then heated to remove excess of bromine, afterwards diluted.

Iodine exposure experiment

Parts of the radioactive iodide or iodate stock solutions were added to 300 ml of the culture solution in 300 ml conical beakers that served as culture vessels. The activities in the solutions were adjusted to about 1000 cpm/ml with a NaI scintilation-counter. Pairs of the prepared Komatsuna plant were transplanted to the vessels, which were covered with aluminum foil to shield the roots from the light.

These plant samples were then placed in a growth chamber, which was specially designed for preventing a release of gaseous radioiodine outside[3]. In the exposure experiments, temperature and humidity in the chamber were maintained at 26 ± 2°C and 50 ± 15% during daytime, and 14 ± 2°C and 70 ± 15% during nighttime. Here, the illumination was provided by a combination of fluorescent lamps and mercury lamps, and the light intensity of daytime was 8000 lux at leaf level.

After 24 hours' exposure, the plant samples are taken out of the chamber. The roots were rinsed quickly by the fresh culture solution which was free from radioiodine, superficial water on the roots was removed with paper tissues. The plants were divided into shoots and roots, and each part was homogenized in a mortar. Radioactivities were measured with a NaI scintillation-counter.

Determination of chemical forms of iodine in the solution

The probable chemical forms of iodine in the culture solution are iodide, iodate, elemental iodine and organic iodine. In this study the chemical forms of radioiodine (*I) were determined by using the isotope exchange reactions:

$$*I^- + 1/2\, I_2 \rightleftharpoons I^- + 1/2\, *I_2 \quad \text{(a)}$$

$$*IO_3^- + 1/2\, I_2 \rightleftharpoons IO_3^- + 1/2\, *I_2 \quad \text{(b)}$$

It is recognized that the reaction (a) proceeds very fast, whereas the reaction (b) is extremely slow[4,5]. Therefore each chemical species is separately determined.

A part of the culture solution was transferred to a separatoly funnel, mixed with carbon tetrachloride and shaken. For the measurement of radioactive elemental and organic iodine the organic phase was separated. A carbon tetrachloride containing stable elemental iodine (about 0.2 mg I_2/ml) was added to the aqueous phase, shaken vigorously, then separated. These procedures were repeated 3 or 4 times. During this process, the complete exchange between radioactive iodide (*I^-) and stable iodine (I_2) in the organic phase is accomplished. The activity of the aqueous solution was counted to determine radioactive iodate, which remained in the solution because of the slow exchange reaction (b). The amount of the radioactive iodide was calculated by subtracting the activities of the elemental and organic iodine and iodate from the initial activity.

RESULTS AND DISCUSSIONS

(1) Uptake of iodide and iodate by plants

Fig. 1 shows the uptake of iodide and iodate by Komatsuna plants in different growing stages of 24, 35 and 40 days after seeding. The accumulation of iodide both in roots and shoots was several times higher than that of iodate. The radioactivity per unit weight of root was much higher than that of shoot. These findings were similar to the results reported by Whitehead[6] using clovers grown in solution cultures. Tensho and Yeh[7] investigated the uptake of iodine by wheat seedlings from soil, showing that iodide was the most available form, followed by elemental iodine and iodate.

The activity ratios of iodide for the shoots of 24, 35 and 40 days old Komatsuna plants were 5.6, 2.1 and 0.94, respectively. The activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution. For reference, the growth curve obtained for the plants under the present experimental conditions is shown in Fig. 2. From Figs. 1 and 2 a reverse

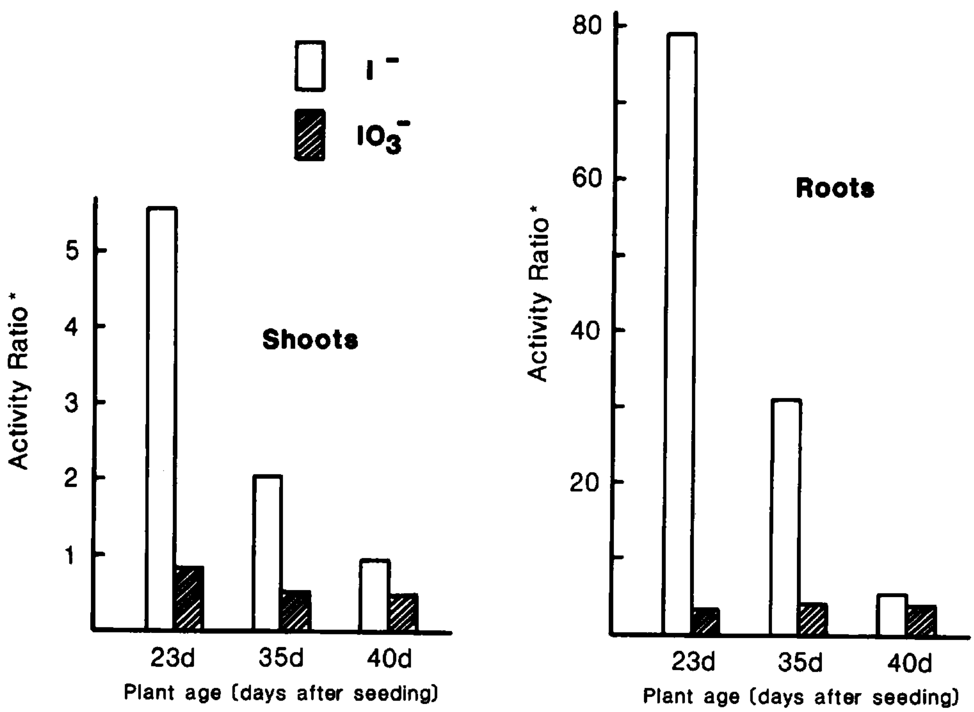

Fig. 1. One day uptake of iodide and iodate by Komatsuna, *Brassica Rapa var. pervidis*, of different growth stage.

(Note)
*Activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution.
No stable iodine was added to the culture solution as carrier.

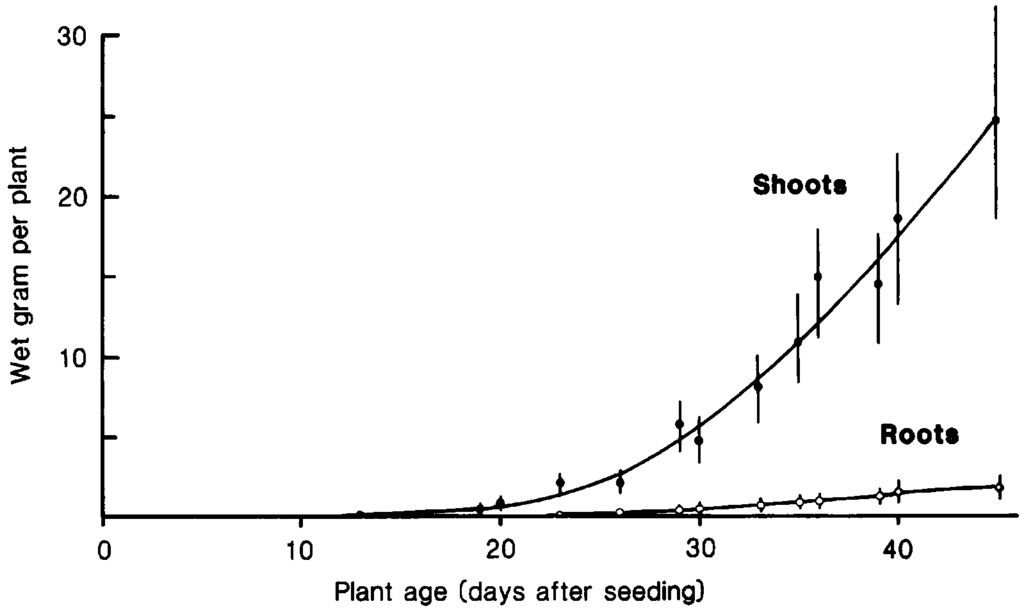

Fig. 2. Growth curve of Komatsuna, *Brassica Rapa var. pervidis.*

correlation between the activity ratio and the plant age can be deduced.

The water consumption of the Komatsuna plants during 24 hours was about 2 to 3 times more than the fresh weight of the plants in this experiment. If radioiodine is taken up by the plant at the same speed as water and wholly retained in the plant, the activity ratio of plant/culture-solution after 24 hours' exposure should be nearly 2 to 3. The activity ratio of iodide in the shoots of 35 days old plants was comparable to this value. The ratio in the younger plants (24 days old) was remarkably higher, whereas that in older one (40 days old) was lower than this value. It can be said that the plants in the earlier growth stage absorb iodide selectively from the solution. On the contrary, inactive transport of iodide to the shoots of 40 days old plants was observed. Decreasing uptake of the nutrients such as N, P, K, Ca and Mg with increasing plant age was also reported by Barber[7].

The activity ratios of iodate for shoots of 24, 35 and 40 days old plants were 0.83, 0.53 and 0.49, showing no significant differences among those growing stages.

(2) Influence of stable iodine content on the uptake of radioiodine

Variable amount of stable iodide or iodate carrier were added to the culture solutions containing a constant amount of radioactive iodide or iodate and then the 24 hours' uptake of radioiodine by plants was examined. Fig. 3 shows the influence of the carrier on the uptake of those radioisotopes. The activity ratio of radioactive iodide for roots decreased remarkably with increasing amount

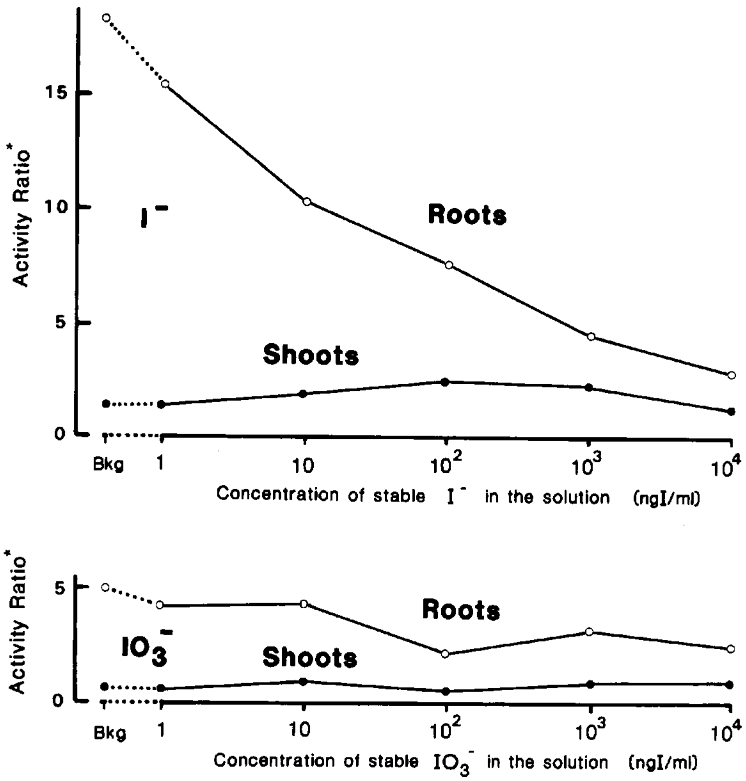

Fig. 3. Influence of stable iodine content on the uptake of radioiodine by Komatsuna, *Brassica Rapa var. pervidis.*

(Note)
*Activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution.
Plant age: 36 days after seeding.
Bkg: Background level of stable iodine in the solution was less than 0.5 ngI/ml.

of stable iodide, whereas that of iodate showed a slight decrease. On the other hand, the activity ratios for shoots were almost constant, though the values were different between iodide and iodate.

The remarkable decrease of iodide in roots can be explained by the isotope dilution effect. The almost constant concentration of radioiodine in shoots indicates that the stable iodine content in shoots increased almost proportionally with the increasing concentration of stable iodine in the solution. A similar result was reported by Whitehead[6)] using clovers cultivated in culture solution containing stable iodine. He mentioned that the quantities of stable iodine taken up by clovers increased with increasing concentration of iodide in the solution. Significant uptake of iodide by agricultural products grown on soil solution which had a high iodine concentration was also found by Tensho and Yeh[9)] and

Yuita[10] specifically in relation to the "Reclamation-Akagare" disease of lowland rice.

Information on iodine concentration of soil solution is quite limited. We adopted tentatively the value described by Bowen[11], 0.01 ppmI for soil solution, and used the culture solutions containing stable iodine(I^- or IO_3^-), 10 ng per ml as I in the following experiments.

(3) Uptake of iodine as a function of time

Fig. 4 shows the uptake of radioactive iodide and iodate by shoots during 1, 2, 3 and 7 days. The culture solutions were renewed daily to reduce the altering of radioiodine concentration and its chemical forms during the exposure. It is expected that both chemical forms of iodine are increasingly concentrated in the shoots with increasing time though the slopes are different each other.

(4) Distribution of radioiodine in Komatsuna plants

Table 1 shows the distribution of radioiodine in the different fractions of the plants. Both iodide and iodate were increasingly accumulated in the order

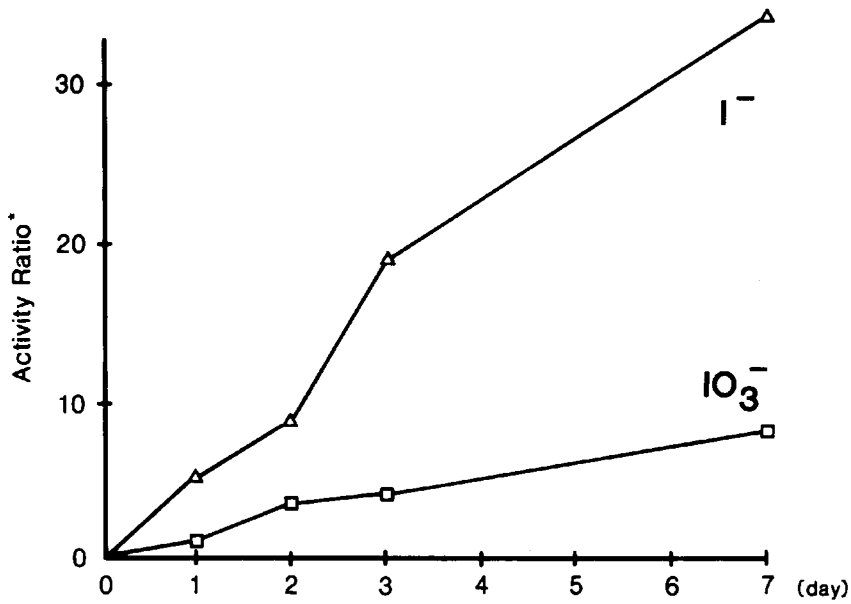

Fig. 4. Accumulation of iodine (iodide and iodate) in shoots of Komatsuna, *Brassica Rapa var. pervidis*, as a function of time.

(Note)
*Activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution.
Stable iodine (iodide or iodate) concentration in the culture solution: 10 ng/ml as I.
Plant age at the beginning of the experiment: 30 days after seeding.

Table 1. Distribution of radioiodine in Komatsuna, *Brassica Rapa var. pervidis.*

	Part	Activity ratio* I^-	Activity ratio* IO_3^-
I-a	Leaves	2.9	0.49
	Stems	4.9	0.77
	Roots (upper part)	2.7	0.88
	Roots (lower part)	41	10
I-b	Leaves	1.4	0.48
	Stems	1.7	0.65
	Roots (upper part)	1.8	1.4
	Roots (lower part)	9.7	6.0

(Note)
*Activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution.
Concentration of stable iodine (iodide or iodate) in the culture solution: 10 ngI/ml
Plant age; I-a: 23 days after seeding, I-b: 36 days after seeding.

Table 2. Alteration of radioiodine concentrations in the culture solutions before and after the exposure experiment as a function of plant age.

	Age of Komatsuna plants	Ratio of iodine activity in the culture solution before and after the exposure experiment*
Culture solution containing I^-	23	0.89
	29	0.89
	36	0.95
	40	0.99
Culture solution containing IO_3^-	23	1.06
	29	1.04
	36	1.10
	40	1.09

(Note)
*Radioiodine concentration in the solution after 24 hours uptake was divided by the concentration in the solution before the exposure experiment; Smaller than 1.0 indicates the selective uptake by Komatsuna plants.
Concentration of stable iodine (iodide or iodate) in the culture solution: 10 ngI/ml

leaves, stems, upper part of roots and lower part of roots. The activities of the lower part of roots were extremely high, but the differences between leaves and stems were small. Not shown in the table, the iodine content in the lower parts of the leaves were somewhat higher than that of the upper parts.

In order to compare the amount of radioiodine in the liquid phase of the plant with that in the residual one, the shoots were crushed and then filtered through 1.2 μm glass filter (Watman GF/C) to separate the plant juice and the residue. The residue was further pressed to extract the water. A higher activity of radioiodine was found in the liquid part than in the residual plant materials.

Distribution of radioiodine in a plant showed that about 30 — 50% of the activity was found in the root, though the percentage weight of the part was about 10%. The high uptake rate by roots might be due to the adsorption of iodine on the surface of the root hairs which came in contact with the solution. This is partly explained by the fact that there was not so much difference in the activity between the roots with shoots and the excised ones.

(5) Radioactivity of iodine in the culture solution after the 24 hours' exposure

Alterations of radioiodine concentrations in the culture solutions were measured before and after the exposure experiment, and the results are shown in Table 2. If iodine is taken up by plants passively with water being transpired, the concentrations of the nuclide in the culture solution would be always constant. It can be seen from the table that the activity of iodide in the solution decreased, but only under the presence of younger plants. On the other hand, the activity of iodate always increased. This suggests iodide was taken up selectively by the younger plants, whereas iodate was poorly absorbed.

For reference, no adsorption of radioiodine on the glass wall inside the vessel was observed.

(6) Transformation of chemical forms of iodine in the culture solution

Fig. 5 shows the transformation of iodate to iodide in the culture solution with and without plant and also with and without stable iodine carrier. It was found that the transformation rate with plants was considerably higher than that without plant. The transformation rate without carrier was higher than that with carrier. Little transformation of iodide to iodate was observed. Elemental or organic iodine was produced though very small in quantities. It seems that the transformation of iodate to iodide might be accelerated by the plant activity.

In order to know the net uptake of IO_3^- by plants it is required to keep the I^-/IO_3^- ratio in the culture solution as low as possible during the uptake experiment. This is partly accomplished by growing the plants in different volume of the culture solution. Supposing a plant could change a fixed amount of iodate to iodide, increasing volume of the culture solution leads to the decrease of

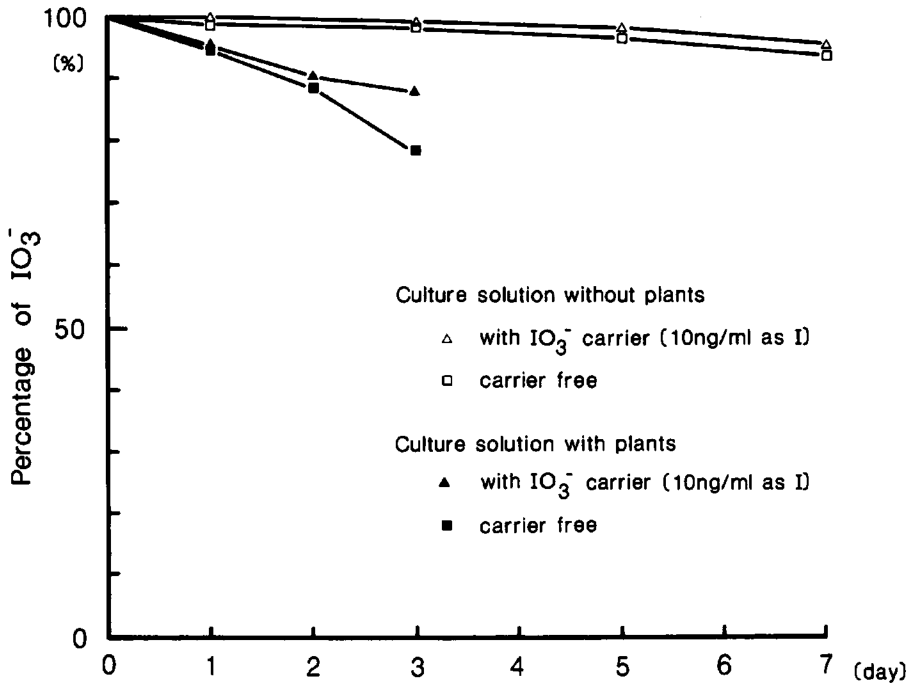

Fig. 5. Transformation of iodate to iodide in the solution with and without plant and also with and without stable iodine carrier.

(Note)
Plant age at the beginning of the experiment: 30 days after seeding.

Table 3. Activity ratios and iodide to iodate ratios in the culture solution.

	Part	Activity ratio*	Ratio of I^-/IO_3^- in the solution after the 24 hrs' exposure
A.	Shoots	0.94	0.16
	Roots	2.7	
B.	Shoots	0.63	0.053
	Roots	1.8	

(Note)
Ratio of I^-/IO_3^- in the solution before the experiment: less than 0.01
Volume of the culture solution; A: 300 ml, B: 900 ml.
Concentration of stable iodate in the culture solution: 10 ngI/ml
Plant age: 40 days after seeding.
*Activity ratio is defined as activity per unit weight of plant sample divided by the activity of the culture solution.

I^-/IO_3^- ratio in the solution. As shown in Table 3 after 24 hours' exposure experiment the I^-/IO_3^- ratio in the small container (A) was higher than that in the larger one (B). The uptake of iodate by plants was lower in the larger container, which has the low I^-/IO_3^- ratio. This result indicates that even in the iodate solution the plants absorb preferably iodide which has been formed from iodate. Therefore the net uptake rate of iodate should be smaller than the values given, for example, in Fig. 1.

Tsunogai and Sase[11)] reported the process of iodate reduction to iodide by organisms utilising the enzyme nitrate reductase. They also described that iodide was generally more enriched in the sea water having higher biological activity. But the transformation mechanisms in the terrestrial water still remains to be studied.

(7) Chemical form of iodine in Komatsuna plants

The chemical forms of the radioiodine in the plant juice are shown in Table 4. Iodide is dominant even in the plants grown in the culture solution containing iodate. The ratio of iodide to iodate in the root juice was higher than that in shoot juice. These might be due to the high adsorption of iodide on the roots.

In order to know the stability of iodate in the plants, radioactive iodate was added to filtered plant juice free from radioiodine and the chemical forms were determined as a function of time. The results are presented in Table 5. This table shows that iodate was converted readily to iodide in the plant juice, and after 24 hours about a half of it was converted to iodide. This suggests that the chemically stable form of iodine in the plant is iodide.

Table 4. Chemical form of iodine in the plant juice from Komatsuna, *Brassica Rapa var. pervidis*, which was grown on the iodate solution.

Plant juice from;	Proportion of radioactive iodate and iodide in the plant juice (%)*	
	IO_3^-	I^-
Shoots	10.3	89.7
Roots	4.8	95.2

(Note)
The chemical forms were determined after 24 hours' exposure experiment.
*Proportion of radioactive iodate and concentration of stable iodate in the culture solution before the exposure were 100% and 10 ngI/ml, respectively.
Plant age: 38 days after seeding.

Table 5. Transformation of iodide to iodate and iodate to iodide in the plant juice of Komatsuna, *Brassica Rapa var. pervidis.*

Chemical form of radioiodine added	Proportion of radioactive iodide and iodate					
	after 10 min.		after 1 day		after 2 days	
	I^-	IO_3^-	I^-	IO_3^-	I^-	IO_3^-
I^-	98	2	97	3	97	3
IO_3^-	1	99	56	44	85	15

(Note)
Radioactive iodide or iodate was added to the plant juice, and then the transformation of chemical forms in the plant juice were examined as a function of time.

CONCLUSION

The results obtained can be summarized as follows:

1. Iodide was taken up by Komatsuna plants much more readily than iodate. Both iodide and iodate were higher concentrated in roots than in shoots.
2. The uptake rate of iodide in the younger plants was higher than that in the older ones, whereas that of iodate varied not so much with plant age. The approximate activity ratios of iodide for shoots of 24, 35 and 40 days old plants were 5.6, 2.1 and 0.94, and those of iodate were 0.83, 0.53 and 0.49, respectively.
3. High uptake rate of iodine, specifically iodide, by roots may be due to the absorption of the element on the root surface. The plant of the earlier growth stage absorb iodide selectively from the solution.
4. The transfer rate of radioactive iodide to roots decreased remarkably with increasing amount of stable iodide in the culture solution, whereas that to shoots remained relatively constant indicating that the stable iodine content in shoots increased almost proportionally with the increasing concentration of stable iodine in the solution.
5. A significant amount of iodate in the culture solution was reduced to iodide in the presence of the plants.
6. It is expected that both iodide and iodate are increasingly concentrated in the shoots with increasing time though the slopes are different each other.
7. The dominant chemical form of iodine in the plant juice was iodide.

ACKNOWLEDGEMENTS

The authors wish to express their thanks to Dr. M. Saiki, National Institute of Radiological Sciences, and Prof. H. Glubrecht, University of Hannover, for their help and encouragement. The second author (D.C.) was the recipient of a fellowship from the Science and Technology Agency of Japan.

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