

# Biogeochemical transfer and dynamics of iodine in a soil–plant system

Huan-Xin Weng · Ai-Lan Yan · Chun-Lai Hong ·  
Ya-Chao Qin · Lehua Pan · Ling-Li Xie

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**Abstract** Radioactive iodide ( $^{125}\text{I}$ ) is used as a tracer to investigate the fate and transport of iodine in soil under various leaching conditions as well as the dynamic transfer in a soil–plant (Chinese cabbage) system. Results show that both soils (the paddy soil and the sandy soil) exhibit strong retention capability, with the paddy soil being slightly stronger. Most iodine is retained by soils, especially in the top 10 cm, and the highest concentration occurs at the top most section of the soil columns. Leaching with 1–2 pore volume water does not change this pattern of vertical distributions. Early breakthrough and long tailing are two features observed in the leaching experiments. Because of the relatively low peak concentration, the early breakthrough is really not an environmental concern of contamination to groundwater. The long tailing implies that the retained iodine is undergoing slow but steady release and the soils can provide a low but stable level of mobile iodine after a short period. The enrichment factors of  $^{125}\text{I}$  in different plant tissues are ranked as: root >

stem > petiole > leaf, and the  $^{125}\text{I}$  distribution in the young leaves is obviously higher than that in the old ones. The concentrations of  $^{125}\text{I}$  in soil and Chinese cabbage can be simulated with a dual-chamber model very well. The biogeochemical behaviors of iodine in the soil–cabbage system show that cultivating iodized cabbage is an environmentally friendly and effective technique to eliminate iodine deficiency disorders (IDD). Planting vegetables such as cabbage on the  $^{129}\text{I}$ -contaminated soil could be a good remediation technique worthy of consideration.

**Keywords**  $^{125}\text{I}$  · Biogeochemical transfer · Soil–plant system · Box model · IDD

## Introduction

Iodine is an essential microelement for synthesis of thyroid hormones in human body. The environmental iodine deficiency can cause a number of health problems known as iodine deficiency disorders (IDD) (Longombe and Geelhoed 1997). Iodine deficiency is the single most common cause of preventable mental retardation and brain damage in the world (Delange et al. 2001). It also decreases child survival, causes goiters, and impairs growth and development. Iodine deficiency has become a threat to the social and economic development of many countries (ICCIDD 1999). Globally, 2.2 billion people (38% of the world's population) live in areas with iodine

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H.-X. Weng (✉) · A.-L. Yan · C.-L. Hong ·  
Y.-C. Qin · L.-L. Xie  
Institute of Environment and Biogeochemistry, Zhejiang  
University, Hangzhou 310027, People's Republic  
of China  
e-mail: gswenghx@zju.edu.cn

L. Pan  
Lawrence Berkeley National Laboratory, Berkeley,  
CA 94720, USA

deficiency and risk its complications, with 650 million suffering from goiter (14% of the world population) (Delange 1998). Supplying iodized salt to communities at risk of the disorders is the most common practice worldwide (Johnson et al. 2003). However, although using iodized salt is a convenient way to supplement the diet, too much iodine in a deficient community can lead to problems of toxicity and this requires careful monitoring of the iodized salt distribution. Furthermore, 90% of iodine in the iodized salt is wasted during production, storage, transportation, and cooking (Chi 1993). On the other hand, IDD is closely related to the low iodine content of food. In fact, food accounts for over 90% of human iodine exposure in most circumstances, whereas water and air provide minimal input (Zheng et al. 2001). Iodine in foodstuffs can easily be assimilated and its bioavailability is up to 99%. Therefore, improving iodine content in foodstuffs by environmental controls as a more natural way to eliminate IDD has attracted much attention recently (Cao et al. 1994; DeLong et al. 1997; Jiang et al. 1997; Shaohua and DeLong 2000; Fordyce et al. 2003; Johnson et al. 2003). However, the biogeochemical behavior of iodine in soil–plant systems is not yet well understood, even though early study of iodine uptake by plant can be tracked back to a century ago (Mark 1936). As an element with multiple oxidation states, iodine exhibits complex biogeochemical behavior in environments (Whitehead 1984). Varied soil redox conditions and complex interactions between iodine species and soil organic matter and minerals make iodine behavior in soil–plant system more puzzling. Different or even conflicting mobility data of iodine in soils were observed (Whitehead 1984; Hu et al. 2005). Quite different transfer factors (TF), defined as the ratio of iodine content in plant to that in soil, which is often used to assess the iodine enrichment of plant, were also reported (Ng 1982; Frissel and van Bergeijk 1989; Robens et al. 1988; Muramatsu et al. 1989; Shinonaga et al. 2001). Few publications address the fate and dynamics of iodine transfer in soil–plant systems.

The objectives of this study are to understand the fate and transport of iodine in soil–plant system using a radioactive  $^{125}\text{I}$  tracer technique and to describe the dynamic transfer of iodine in the soil–vegetable system with mathematical model. Such knowledge is critical for developing environmentally friendly and

effective techniques of cultivating iodized organic crops as environmental controls to eliminate IDD. It is also useful for assessing the environmental impact of  $^{129}\text{I}$  released from nuclear facilities or for developing environmental remedy techniques for regions with toxic levels of iodine in soil or groundwater.

## Materials and methods

### $^{125}\text{I}$ transport experiments in soils

Two types of the local soils, paddy and sandy soils (Table 1), were chosen as experimental materials. They were retrieved approximately from the upper 2 to 20 cm of the soil layer and the topmost gravel layer was discarded. Then these soils were homogenized and sieved to remove stones and plant debris. The physicochemical properties of the soils are listed in Table 1. The soil was placed in a polyethylene column (27 cm in length by 8 cm in radius) and the column bottom was wrapped with gauze of three layers to prevent soil leakage. Two types of treatments were used in the transport experiments.

Treatment 1 (no leaching): Deionized water was added to the plastic column to keep soil moist after filling the soil into the plastic column slowly. A measure of 2 ml of  $\text{Na}^{125}\text{I}$  ( $22103 \text{ Bq ml}^{-1}$ ) was introduced into the column. After remaining for 20 days at  $30 \pm 4^\circ\text{C}$  in the laboratory, the column was sectioned into intervals at depths of 0.5, 1, 1.5, 2, 3, 4, 5, 7, 10, 15, and 20 cm, and each portion was subsequently dried at  $45^\circ\text{C}$ , crushed, and homogenized. A 1.0 g soil sample from each interval was used to measure the  $^{125}\text{I}$  radioactivity.

Treatment 2 (leached with various water): Deionized water was added into the column to keep soil moist after filling the soil into the plastic column slowly. A measure of 15 ml of  $\text{Na}^{125}\text{I}$  ( $22103 \text{ Bq ml}^{-1}$ ) was introduced into each column. Then the soil was leached continuously with 500, 750, and 1,000 ml deionized water, respectively, keeping the water surface 2 cm higher than the soil surface. Effluent water from the bottom was collected every 50 ml, and a 1 ml water subsample was measured for  $^{125}\text{I}$  radioactivity. After leaching, the soil column was sectioned into intervals at depths of 0.5, 1, 1.5, 2, 3, 4, 5, 7, 10, 15, and 20 cm, respectively. Each portion was subsequently dried at  $45^\circ\text{C}$ , crushed, and

**Table 1** Physicochemical properties of the soil used in the experiments

Soil type	Organic matter (g kg <sup>-1</sup> )	pH	Cation exchange capacity (cmol kg <sup>-1</sup> )	Clay (%)	Silt (%)	Sand (%)
Paddy	40.9	5.91	19.28	36.86	45.46	17.68
Sandy	7.9	6.5	3.28	2.5	31.2	66.3

homogenized. A 1.0 g soil subsample from each section was measured for radioactivity.

<sup>125</sup>I uptake experiments for vegetable (Chinese cabbage)

Chinese cabbage (*Brassica chinesis* L.) seeds were soaked into water at 55°C for 15 min and then disinfected in 1% KMnO<sub>4</sub> solution for 15 min followed by thorough rinsing with deionized water. These seeds were subsequently placed into a thermostostat at 30°C for germination using sterilized gauze. After 80% had germinated in the seedbed, the seedlings were carefully conveyed into quartz sand, and 4 days later appropriate 1/2 Hoagland nutrient solution (Table 2) was periodically fertilized for their growth. After cultivation for 25 days by nutrient solution, three seedlings were each transplanted into a PVC pot (12 cm by 10 cm) containing 0.5 kg of either of the two soils (paddy soil and sandy soil). Three days after transplanting, two holes with a depth of 3 cm were drilled symmetrically along the edge of each pot using a glass stick (4 mm o.d.). A measure of 2 ml of 31,372 Bq ml<sup>-1</sup> Na<sup>125</sup>I solution was slowly injected into each hole by using a pipette, then these holes were filled evenly. The same operation procedures described above were carried out three times and a total of 12 ml sodium iodide solution was injected. The temperature was controlled at 24 ± 4°C at all times. After 0.25, 0.5, 1, 2, 4, 6, 10, and 15 days, respectively, plant tissue samples in four categories (root, shoot, petiole, and leaf) were randomly

collected from six cabbages with identical treatment. The roots were rinsed with deionized water and dried with absorbent paper. All of these tissue samples were cut into small pieces and measured for their dry weights after drying at 45°C. A subsample of 100 g from each fraction was used to measure <sup>125</sup>I radioactivity.

Radioactivity measurement and radio autograph

<sup>125</sup>I radioactivity was measured with a multichannel γ spectrometer (model BH 1224, Beijing Nuclear Instrumentation Factory). This γ spectrometer is equipped with an NaI scintillation detector (70 mm in diameter), and is inverted in a lead-shielded chamber. Disposable plastic cups, 75 mm in diameter and 110 mm in height, were placed on the top of the scintillation detector during measurements. A home-made location device was used for fixing the counting position, so as to ensure homogeneity in the geometrical positioning of all samples. The counting error was controlled to be lower than 5%. All the samples were measured on the day of sampling to avoid errors due to water evaporation. The counting data were calibrated with counting efficiency, dead time, disintegration, and any other factors. The best-fit equations were established at a 95% confidence level.

A piece of leaf was picked out for radio autograph scanning from the cabbage cultivated for 15 days in the Na<sup>125</sup>I-fertilized paddy soil to illustrate the distribution of the iodine uptake in the leaf. The leaf outspread was fixed between two pieces of glass and

**Table 2** Nutrient composition of the Hoagland used in the experiments

Macronutrient	Concentration (mM)	Micronutrient	Concentration (μM)
KNO <sub>3</sub>	6.00	H <sub>3</sub> BO <sub>3</sub>	10.00
Ca(NO <sub>3</sub> ) <sub>2</sub>	3.50	MnSO <sub>4</sub> · H <sub>2</sub> O	0.50
KH <sub>2</sub> PO <sub>4</sub>	1.33	ZnSO <sub>4</sub> · 7H <sub>2</sub> O	0.50
MgSO <sub>4</sub> · 7H <sub>2</sub> O	0.50	CuSO <sub>4</sub> · 5H <sub>2</sub> O	0.20
NaCl	0.48	(NH <sub>4</sub> ) <sub>6</sub> Mo <sub>7</sub> O <sub>24</sub>	0.01
		Fe-EDTA	200

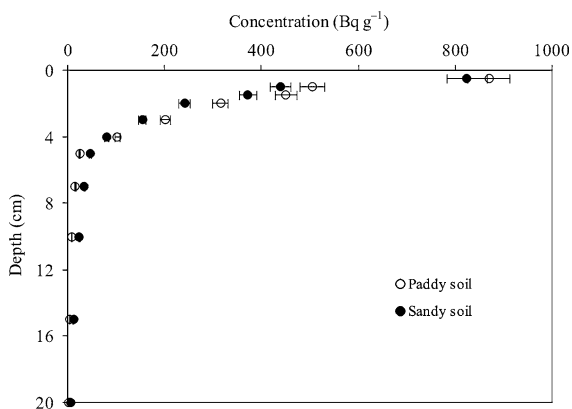
air dried for 2 days. After exposure in a darkroom for 2 days, the film was scanned by using a TYPHOON-9200 fluorescent and a radioisotope image analyzer.

## Results and discussion

### Retention and migration of $^{125}\text{I}$ in soils

The vertical  $^{125}\text{I}$  distributions in the two soil profiles after 20 days of diffusion dominant migration (no leaching treatment) are shown in Fig. 1. Clearly, most of the  $^{125}\text{I}$  remained in the upper 10 cm of the soil column for both soils (97.73% in the paddy soil and 93.98% in the sandy soil). The mean position (the first moment,  $\mu_c$ ) of the plume moved 1.98 cm in paddy soil and 2.04 cm in sandy soil, respectively, while the variance (the second moment,  $\sigma^2$ ) was 3.89  $\text{cm}^2$  in paddy soil and 5.70  $\text{cm}^2$  in sandy soil. These results suggest that both soils have a strong retention capability for iodine, though the paddy soil was stronger than the sandy soil. Such a strong retention capability can be further observed from the results of the leaching experiments with varying amounts of leaching water (Table 3). Only about 4.3% and 5.96% of the fertilized radioactivity of  $^{125}\text{I}$  was leached from the paddy and the sandy soil columns, respectively; these values are on the same order of magnitude as the volatilization loss (2.3% for the paddy soil and 4.7% for the sandy soil). The majority of the applied iodine (82.48% for the paddy soil and

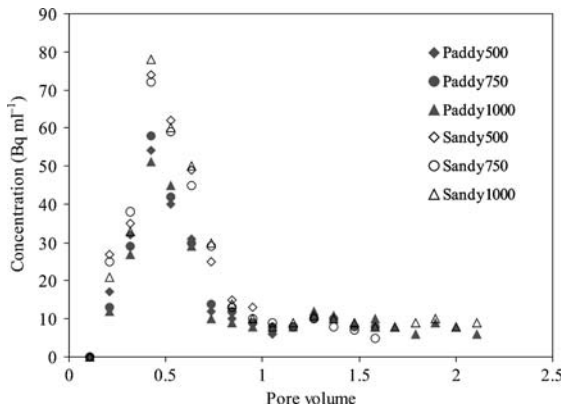
78.43% for the sandy soil) remained in the 20 cm column of soil. Doubling the leaching water (from 500 to 1,000 ml) only decreases the residual iodine by about 1.5% of the applied iodine. Note that the applied iodine in these experiments is iodide, which is often considered as a nonreactive tracer (Turin et al. 2002) because iodide sorption onto many types of geologic media is extremely limited (Whitehead 1984; Turin et al. 2002). Although significant iodide sorption to the soils has been reported (Hu et al. 2005), mostly with significant positively charged surfaces, the retention rate of 80% observed in this study cannot be explained by the sorption of iodide onto soils. In many column experiments of various soils, the majority of iodide can be recovered from the effluent with two or more pore volume (PV) (Hu et al. 2005). We notice that, in contrast with many soil column transport experiments in which the system is not open to air (Hu et al. 2005), the column experiments in this study were conducted under a condition similar to nature: soil during rain (or irrigation) or between rain events. This condition facilitates oxidization of iodide into other iodine species that are more active with soil organic matter and soil minerals. The transformation of iodide into other reactive species must have been taking place under the experimental conditions. For example, in a system open to air, iodide ( $\text{I}^-$ ) can be oxidized to the more reactive iodate ( $\text{IO}_3^-$ ) (Fuhrmann et al. 1998). In addition, iodine was found to remain chemically stable for 1000 years in natural peat land once it was transformed from an inorganic form to organoiodine compounds (Keppler et al. 2003). Fixation of inorganic iodine onto macromolecular humic substances, which greatly changes the physiochemical characteristics of iodine, has also been reported (Tikhomirov et al. 1980). Because radioactivity measurement of  $^{125}\text{I}$  does not distinguish the chemical status of iodine in these experiments, we cannot tell which transformation(s) had taken place in the soils. However, the long tails of the breakthrough curves from the leaching experiments (Fig. 2) show similar transport behavior to that of 4-iodoaniline, an organoiodine compound, observed in a Savannah River Site surface soil (Hu et al. 2005). This similarity implies that at least some organoiodine compounds have formed, either by fixation or other transformations. As a result, the paddy soil with higher organic matter has stronger iodine retention capability than the sandy



**Fig. 1** The profile of  $^{125}\text{I}$  in soils 20 days after application of 2 ml iodide solution on the top of the columns without additional leaching. The majority of applied iodine remains in the top 10 cm of the soils

**Table 3** Fate of iodine in soils under various leaching conditions

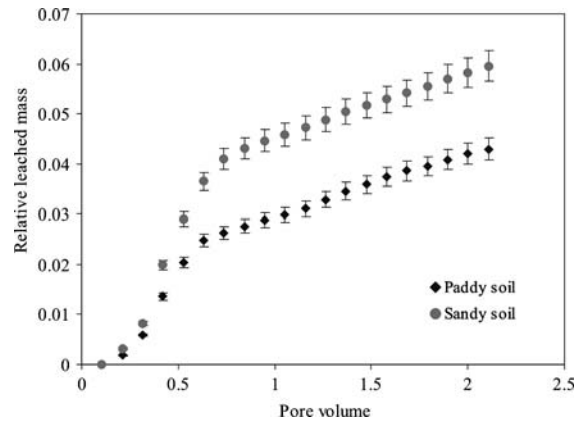
Soil	Leaching amount (ml)	Residual iodine (%)	Leached iodine (%)	Volatilization loss (%)	Decay (%)
Paddy	500	83.93	3.18	1.98	10.91
	750	83.13	3.91	2.05	10.91
	1,000	82.49	4.30	2.30	10.91
Sandy	500	80.39	4.52	4.18	10.91
	750	79.60	5.10	4.39	10.91
	1,000	78.43	5.96	4.70	10.91



**Fig. 2** The breakthrough curves of iodine through soil columns with different amounts of leaching water applied after injection of 15 ml iodide solution. The label “Paddy500” means paddy soil leached with 500 ml water, etc.

soil because the organic matter should play an important role in these transformation processes. Identifying these iodine species and the transform between them in natural soils is important to understanding terrestrial iodine cycling and could be a good research topic for the future.

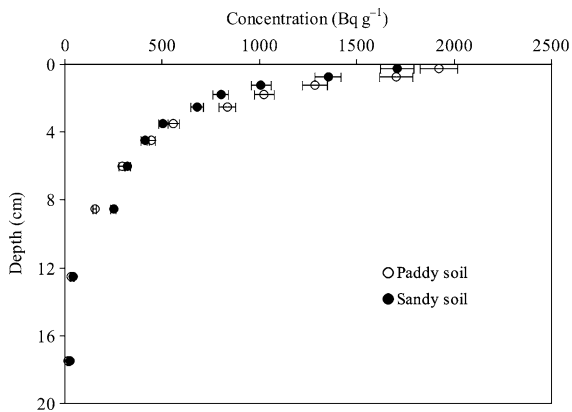
Early breakthrough of mobile iodine occurred in both soils (Fig. 2). The peak concentration occurred at 0.4 PV for both soils with all three leaching amounts. This consistent early breakthrough of mobile iodine indicates that both the paddy soil and the sandy soil have good structures so that there is significant immobile water in dead-end pores, e.g., the pores in soil granular. However, for the possible environmental impact of iodine leaching into the groundwater due to the iodine fertilization, this early breakthrough or preferential flow phenomenon should not be a concern because the peak concentration is very low (50 Bq ml<sup>-1</sup> for the paddy soil and 70 Bq ml<sup>-1</sup> for the sandy soil) as compared with the applied concentration of 22,103 Bq ml<sup>-1</sup>. On the other hand, the long tails of



**Fig. 3** Cumulative iodine mass leached from two soils with 1,000 ml water. The relative leached mass is the ratio of the cumulative mass in the leach solution to the applied mass of iodine

the breakthrough curves have a more complicated environmental significance. As shown in Fig. 3, although only about 3–4% of the applied iodine has been leached out of the soil columns at about 0.7 PV leaching, the cumulative mass of the leached iodine increases linearly thereafter at a rate of 1% of applied iodine for every additional 1 PV of leaching. This implies that the soils can provide a low but stable level of mobile iodine after a short period. In terms of cultivating iodized crops, this is a preferable buffering feature of the soil with which the plant can grow in an iodized soil but not toxic for a long period, probably many reasons. However, in terms of remediation of soils contaminated by the long-half-time isotope, <sup>129</sup>I, this is an unfavorable feature that could lead to exposure to an overdose of <sup>129</sup>I over the long term and that the common wash and treat approach would not be effective.

It is interesting to observe that the final iodine profiles in the soil columns after leaching with 1,000 ml water (Fig. 4) exhibit similar patterns to those in Fig. 1



**Fig. 4** The profile of  $^{125}\text{I}$  in soils after leaching with 1,000 ml water. The majority of the applied iodine still remains in the top 10 cm of the soils

(the cases without leaching), i.e., that the concentration decreases rapidly with depth. The spatial moment analysis of the residual iodine in soil columns shows that the effects of the leaching duration on those moments are not significant (Table 4). Note that the highest concentration still remains in the top 0.5 cm of the soil column after leaching with 1,000 ml (2.1 PV) water. This implies that the retention of iodine in these soils takes place very quickly comparing to the downward flow of the iodine solution driven by the leaching water. Once the iodine was retained by the soil components, it can barely migrate except for a small portion that may be released into pore water in some mobile forms. In addition, the upward migration of the volatilized iodine could also contribute to the final vertical distribution pattern of the iodine in the soils by cancellation of the downward migration of the iodide.

In general, the paddy soil showed stronger retention capability for iodine than the sandy soil. As shown in Table 3, the percentages of leached iodine are smaller for the paddy soil than for the sandy soil for all the leaching treatments. In other words, the iodine leached from the paddy soil is about 70–76% of that leached from the sandy soil. Furthermore, the

final distributions of iodine in the paddy soil columns are less spatially spread out than those in the sandy soil columns (Fig. 4). This difference can also be seen in the difference of the second spatial moments between two soils: 5.68–5.81 cm for the paddy soil columns versus 6.55–7.03 cm for the sandy soil columns (Table 4). However, the breakthrough curves for the two soils are similar except that the paddy soil has a lower peak concentration before the first PV (Fig. 3). In particular, the tailing concentrations in the leached solution for the two soils are almost the same (Fig. 3), even though the spatial distributions within the soil columns are different (Fig. 4). A reasonable explanation for these apparently contradictory phenomena is that the stronger iodine retention capability shown by the paddy soil is mainly because the pore water moves more slowly in the paddy soil than it does in the sandy soil. In the paddy soil, because of the slower motion of the iodine solution, there was longer time for adsorption, fixation, or other transformation processes to take place per unit distance traveled by the iodine plume. Obviously, the more iodine removed from the pore water in the upstream, the less iodine will be available in the downstream. The paddy soil has more clay (Table 1), which usually leads to low hydraulic conductivity. As a result, the pore water in the paddy soil flows much slowly than that in the sandy soil under the same pond-leaching conditions used in these experiments. Note that this physical effect of this on the transport of iodine through the soils is not significant during the release-dominant phase. Therefore, the concentration difference in the leaching solution between the paddy soil and the sandy soil reduced rapidly after the peak concentration, and became zero in the long tailings.

#### $^{125}\text{I}$ uptake by Chinese cabbage

The  $^{125}\text{I}$  distribution in paddy soil–cabbage system after cultivation for 15 days is listed in Table 5. The

**Table 4** Spatial moment analysis of the iodine distribution in soils after different leaching amounts

Leaching amount (ml)	First moment (cm)		Second moment (cm)	
	Paddy soil	Sandy soil	Paddy soil	Sandy soil
0	1.98	2.04	3.89	5.70
500	2.46	2.47	5.73	6.55
750	2.49	2.50	5.81	7.03
1,000	2.45	2.51	5.69	6.94

<sup>125</sup>I uptake by the cabbage accounts for 10.1% of the applied <sup>125</sup>I, while most of the <sup>125</sup>I (73.63%) was retained in the soil. Consistent with the results from the leaching experiments discussed above, the majority of the applied <sup>125</sup>I remained in the upper 10 cm of the soil layer, accounting for 89.84% of the total soil-retained iodine, whereas only 11.16% migrated down below a depth of 10 cm. Most (80.20%) of the <sup>125</sup>I uptake migrated upwards after uptake. In particular, 39.79% of the <sup>125</sup>I accumulates in the stem, 27.25% in the petiole, and 13.16% in the leaf. These results suggest that iodine is biogeochemically active and tends to be transferred by the plant in a soil–plant system. In particular, the uptake of iodine (10.1%) by cabbage is significantly greater than that lost due to leaching or volatilization (0.39%) under the conditions used in these cultivation experiments.

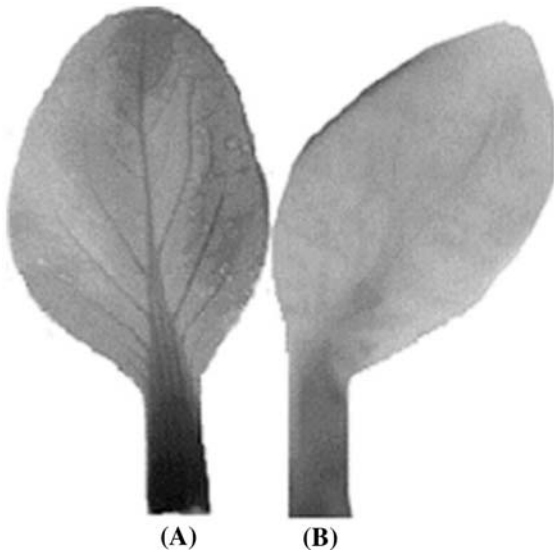
The migration of iodine in the soil–cabbage system can also be evaluated by the enrichment factor (CCs), defined here as the ratio of the specific radioactivity of <sup>125</sup>I of dry/raw tissue to that of dry soil. The enrichment factor of whole cabbage is 2.63, but different cabbage tissues have very different CCs (Table 5). The highest CCs value (13.85) occur in the root, 5.3 times as high as that of the whole plant, whereas the lowest CCs value (0.66) occurs in the leaf, only one-quarter of the whole-plant value. However, because the edible tissues compose the majority of the plant, the total iodine in the edible tissues is sufficient to make the cabbage an ideal candidate for iodized crops for the reduction of IDD.

As revealed by the results from the leaching experiments (i.e., the long-tailing breakthrough curves in Fig. 3), both soils have the ability to maintain a low but stable concentration of soluble iodine for a long period. Therefore, it is possible to grow iodized cabbage several times after one application of iodine fertilizer.

To investigate the distribution of iodine within the cabbage leaf, we took a radio autograph of the <sup>125</sup>I-fertilized cabbage leaf in comparison with a non-<sup>125</sup>I-fertilized one (Fig. 5). From the petiole to the main veins to the small branches, the darkness of the radio autograph gradually decreases. The concentration of <sup>125</sup>I along the leaf edge is lower than in other areas. In the mapping of iodine within the plant leaves, the images was more black and the nervure was more clear, the more iodine the more black in leaf but the mapping without iodine was indistinct. The spatial distribution of <sup>125</sup>I in the leaf (Fig. 5) is consistent with the results of in situ detection, which indicates a marked negative correlation of <sup>125</sup>I radioactivity with distance from the detection position to the main vein ( $r = -0.9752, P \leq 0.05$ ). This phenomenon illustrates a decreasing tendency of iodine accumulation from the main vein to branches and to leaf edge throughout the leaf, and is opposite to the case of seaweed (*Laminaria japonica*), where maximum iodine concentration was detected along the edges of seaweed leaves (Saenko et al. 1978). This difference suggests that different mechanisms are adopted by the two species for uptake of iodine.

**Table 5** Distribution of <sup>125</sup>I in the paddy soil–cabbage system

Item		Dry weight (g)	Specific activity (Bq g <sup>-1</sup> )	Activity (Bq)	Of the plant (%)	Of the initial (%)	Enrichment factor (CCs)
Cabbage	Root	1.08	1163.48	1256.6	19.80	10.1	13.85
	Stem	4.16	607.12	2525.6	39.79		7.23
	Petiole	8.42	205.42	1729.6	27.25		2.45
	Leaf	15.08	55.38	835.13	13.16		0.66
	Whole	28.74	220.52	6346.9	100		2.63
Soil layer	0–3 cm	165	59.35	9792.8	21.2	73.63	
	3–10 cm	165	189.39	31249	67.65		
	6–20 cm	220	23.43	5154.6	11.16		
	All	550	83.99	46197	100		
Auto disintegration		–	–	9981		15.91	
Other		–	–	219.1		0.37	

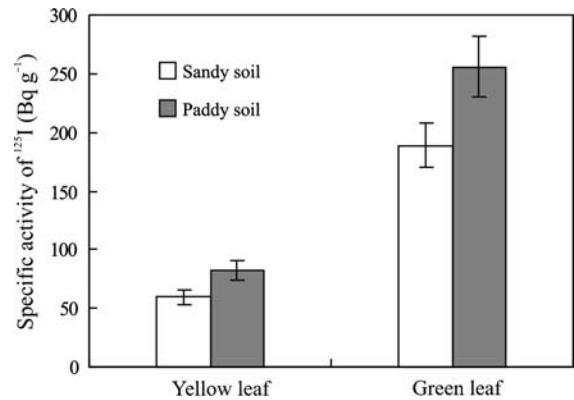


**Fig. 5** Radio autograph of cabbage leaves with (a) and without  $^{125}\text{I}$  fertilization (b)

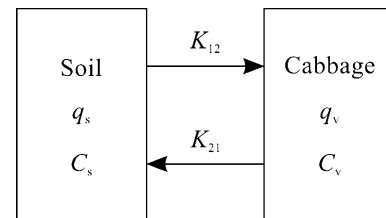
Iodine uptake from seawater by seaweed is active, and the iodine is conserved in the vacuole of specific cells that are mainly distributed along the leaf edge. On the other hand, iodine uptake from soil by cabbage seems to be a passive process and the iodine is stored along the way, especially in those veins that conduct solutions in the plant. In order to keep the cabbage growing, its roots take up water and nutrients, including the iodine, from the soil and transport them to the different parts of the leaves through the veins. For an old (yellow) leaf with less physiological activity, much less water and nutrients will be supplied. Consequently, much less iodine could be found there. Comparison of the iodine content between the green leaves and the yellow leaves confirmed this point (Fig. 6). The  $^{125}\text{I}$  content in green leaf was commonly 2–3 times higher than that in yellow leaf in both soils.

Modeling the dynamics of  $^{125}\text{I}$  in the soil–cabbage system

To describe the iodine behavior in the soil–vegetable system quantitatively, we propose to use a dual-chamber (soil and vegetable) model (Fig. 7) with first-order mass transfer to simulate the iodine transfer dynamics between the soil and vegetable as well as its loss to the environment. The dual chamber (soil and vegetable) models a closed system and the



**Fig. 6** Comparison of iodine concentration between yellow and green leaves planted in two soils



**Fig. 7** Schematic of the open chamber system with interchange

velocity of transfer and transportation of  $^{125}\text{I}$  in each chamber fits a first-order reaction.

For such a dual-chamber model of the soil–vegetable system, the mass balance equations of iodine can be written as following:

$$\begin{aligned} \frac{dm_s}{dt} &= -E_1 m_s + K_{21} m_v, \\ \frac{dm_v}{dt} &= K_{12} m_s - E_2 m_v, \end{aligned} \quad (1)$$

where  $m_s$  and  $m_v$  are the masses (Bq) of iodine in the soil and vegetable, respectively. The combined transfer rates ( $\text{day}^{-1}$ )  $E_1$  and  $E_2$  are defined as follows:

$$\begin{aligned} E_1 &= K_s + K_{12} \\ E_2 &= K_v + K_{21}, \end{aligned} \quad (2)$$

where  $K_s$ ,  $K_v$ ,  $K_{12}$ , and  $K_{21}$  are the loss rate ( $\text{day}^{-1}$ ) in soil, the loss rate ( $\text{day}^{-1}$ ) in the vegetable, the transfer rate ( $\text{day}^{-1}$ ) from soil to vegetable, and the transfer rate ( $\text{day}^{-1}$ ) from vegetable to soil, respectively. Note that both loss rates,  $K_s$  and  $K_v$ , include the natural decay rate of  $^{125}\text{I}$  ( $1.16300 \times 10^{-2} \text{ day}^{-1}$ ).



Let  $M_s$  and  $M_v$  be the total soil mass (g) and vegetable mass in the system, respectively. The concentrations ( $\text{Bq g}^{-1}$ ) of  $^{125}\text{I}$  in soil and vegetable,  $C_s$  and  $C_v$ , can thus be defined as:

$$\begin{aligned} C_s &= m_s/M_s \\ C_v &= m_v/M_v \end{aligned} \tag{3}$$

Combining Eqs. (1) and (3), we obtain the governing equations for  $^{125}\text{I}$  in the soil–vegetable system as follows:

$$\begin{aligned} \frac{dC_s}{dt} &= -E_1 C_s + K_{vs} C_v \\ \frac{dC_v}{dt} &= K_{sv} C_s + E_2 C_v, \end{aligned} \tag{4}$$

where  $K_{vs}$  ( $= K_{21}M_v/M_s$ ) and  $K_{sv}$  ( $= K_{12}M_s/M_v$ ) are the scaled transfer rates from cabbage to soil and vice versa, respectively.

Given the initial conditions of the system:

$$\begin{aligned} C_s &= C_0 \quad t = 0 \\ C_v &= 0 \quad t = 0 \end{aligned} \tag{5}$$

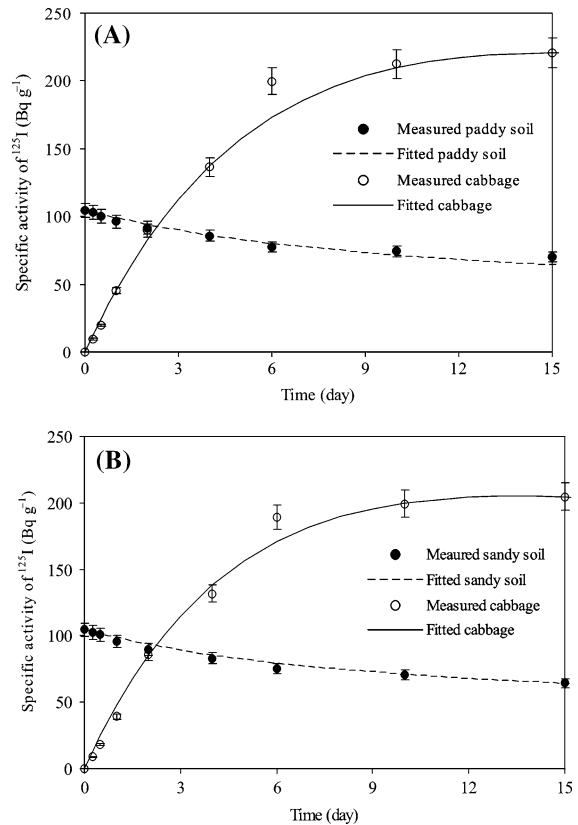
The solutions of Eq. (4) with the initial conditions (5) can be readily written as:

$$\begin{aligned} C_s &= \frac{C_0}{A_1 - A_2} [(E_2 - \lambda_2)e^{-A_2 t} - (E_2 - \lambda_1)e^{-A_1 t}] \\ C_v &= \frac{K_{sv} C_0}{A_1 - A_2} (e^{-A_2 t} - e^{-A_1 t}) \end{aligned} \tag{6}$$

where  $A_1$  and  $A_2$  are parameters, defined as follows:

$$\begin{aligned} A_1 &= \frac{1}{2} \left[ E_1 + E_2 + \sqrt{(E_1 - E_2)^2 + 4K_{sv}K_{vs}} \right] \\ A_2 &= \frac{1}{2} \left[ E_1 + E_2 - \sqrt{(E_1 - E_2)^2 + 4K_{sv}K_{vs}} \right]. \end{aligned} \tag{7}$$

As shown in Fig. 8, the model reproduced the observed iodine concentrations in soil and cabbage very well. The correlation coefficient between the simulated and the observed concentrations is very high ( $>0.99$ ) for all cases (Table 6). This good agreement between the simulated and the observed data indicate that the simple dual-chamber model is effective in simulating the biogeochemical transfer dynamics of iodine at the soil–vegetable system level. Furthermore, the fitted various transfer rates provide some important information about the iodine behavior in the soil–vegetable system (Table 6). First,  $K_v$  is



**Fig. 8** Measured and fitted  $^{125}\text{I}$  concentrations in (a) paddy soil and cabbage and in (b) sandy soil and cabbage

only slightly larger than the natural decay rate (i.e., the difference is  $5 \times 10^{-5} \text{ day}^{-1}$  for both soils), which indicates that the loss of iodine from the cabbage to the environment is negligible. However, the loss of iodine from the soil to the environment via volatilization or leaching is much higher, and the sandy soil is more vulnerable to such loss than the paddy soil, as indicated by its significant larger loss rate,  $K_s$  (i.e.,  $K_s$  minus the natural decay rate is  $2.4 \times 10^{-5} \text{ day}^{-1}$  for the paddy soil and  $4.1 \times 10^{-5} \text{ day}^{-1}$  for the sandy soil, respectively). Because the leaching is minimized in the experiments by proper irrigation, this loss of iodine is mainly because of volatilization. Obviously, larger leaching losses due to storm events are possible and the rate  $K_s$  could be larger for a soil–cabbage system in field conditions than for the well-controlled system used in these experiments. However, as revealed in the leaching experiments discussed above, the leaching loss is limited and would be on the same order as the volatilization loss

**Table 6** Fitting parameters of the dual-chamber model for the soil–cabbage system

System	$K_s$ (day <sup>-1</sup> )	$K_v$ (day <sup>-1</sup> )	$K_{sv}$ (day <sup>-1</sup> )	$K_{vs}$ (day <sup>-1</sup> )	$r_s^a$	$r_v^a$
Paddy soil–cabbage	0.0140	0.0117	0.4780	0.0120	0.988	0.996
Sandy soil–cabbage	0.0157	0.0117	0.5085	0.0144	0.994	0.997

<sup>a</sup>  $r$  is the correlation coefficient between the measured and the simulated data while the subscripts “s” and “v” indicate soil and cabbage, respectively. The natural decay rate of <sup>125</sup>I is 0.01163 day<sup>-1</sup> (half life = 59.6 days)

in the open system. Second, the scaled transfer rate from soil to cabbage,  $K_{sv}$ , is much higher than any of the other rates in both soils, which indicates effective uptake of iodine by the cabbage. In fact, more than 10% of the applied iodine was absorbed by the cabbage after 15 days of cultivation.

## Conclusions

The paddy and sandy soils exhibit strong retention capability for the fertilized <sup>125</sup>I, most of which remained in the upper 10 cm of soil layer, demonstrating relative immobility from downward diffusion and transfer. The breakthrough curves show features of early breakthrough and long tailing, which is common in a well-structured soil. The long tailing implies that the retained iodine is undergoing slow but steady release and that the soils can provide a low but stable level of mobile iodine after a short period. In terms of cultivating iodized crops, this is a preferable buffering feature of the soil with which the plant can grow in an iodized soil but not be toxic for a long period, probably many seasons.

<sup>125</sup>I can be enriched by Chinese cabbage, with most being transported to the aboveground tissues. The specific activity of <sup>125</sup>I in the cabbage tissues were ranked as root > stem > petiole > leaf. <sup>125</sup>I mainly accumulates in the stem, secondly in the petiole, followed by the leaf. The enrichment factor of the whole plant is 2.63, and the maximum value (13.85) occurs in the root, which is 5.3 times higher than that of the whole plant. Although the <sup>125</sup>I concentration in the stem and leaf are relatively lower, most of the iodine accumulates therein due to their large biomass proportion. The radio autograph shows that the iodine accumulation decreases from main veins to branches and to leaf edge throughout the leaf, which implies a different mechanism of iodine uptake and store in cabbage from that in seaweed. The fact that the <sup>125</sup>I contents in young

(green) leaves were obviously higher than those in old (yellow) ones indicates that iodine uptake by cabbage is highly related to its physiological status, especially to water consumption.

A simple dual-chamber model was used to describe the mass transfer of iodine in the soil–plant system. The model was calibrated using the observed dynamic variation of iodine concentrations in both the soil and the vegetable. The fitted transfer rates indicate that the iodine retained in soils has lower mobility than that in cabbage. The iodine loss to the environment from the soil–cabbage system is small under the cultivation conditions considered.

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