

## Overview and Dynamics of Iodine and Bromine in the Environment

### 2. Iodine and bromine toxicity and environmental hazards

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#### Abstract

Flooding results in the dissolution of I and Br in soil, enabling rice plants to absorb these elements easily. Therefore the use of forest land and upland field soils for paddy rice cultivation under flooding conditions could induce the widespread occurrence of Reclamation-Akagare disease of rice plants, due to excessive absorption of I. In addition, under these circumstances, rice grain pollution by Br may occur, in which the Br content could exceed the allowable residue level (50 mg/kg air dry wt.). Soil fumigation with methylbromide (MB) resulted in the increase of the Br content in vegetables: in greenhouse cucumbers, the average Br content reached 73 mg/kg fresh wt. (1,660 mg/kg dry wt.) which exceeded the allowable residue level of Br in crops. Application of potassium chloride fertilizer (Br 950 mg/kg dry wt.) increased the Br content of crops to the maximum value of 200 mg/kg dry wt. which was much lower than 5,000 mg/kg dry wt. associated with the application of pesticides (MB, EDB). The Br concentration of the well water in areas with heavy MB and EDB application reached 2.3 mg/L, a value 250 times higher than that recorded in areas with lower doses of the chemicals. Influence of potassium chloride fertilizer on the Br concentration of the well water appeared to be negligible. Findings on the behavior of stable I in the soil-plant system were applied for the estimation of the dynamics of long-lived  $^{129}\text{I}$  (half-life,  $1.7 \times 10^7$  years) in the system and for its radiological assessment.

**Discipline:** Agricultural environment/Soils, fertilizers and plant nutrition

**Additional key words:** chlorine, ground water, iodine-129, methylbromide, Reclamation-Akagare disease of rice

#### Excessive absorption of I and Br by paddy rice

The authors cultivated paddy rice from the transplanting to harvesting stages under flooded and non-flooded conditions in soil culture pot experiments using soils of forest land and then a comparison was made between the

contents of I and Br in the top of rice plants<sup>40)</sup>.

As shown in Table 1, the average content of I in straw was 290 mg/kg dry wt. under flooded conditions and 0.37 mg/kg dry wt. under non-flooded conditions, and the ratio of flooded/non-flooded was 784. The average content of Br in straw was 398 mg/kg dry wt. under flooded conditions and 38 mg/kg dry wt. under non-flooded conditions and the ratio of

flooded/non-flooded was 10.5. The higher value of the I ratio under flooded/non-flooded conditions in plant compared with that of Br in plant may be ascribed to the differences in the ratios of both elements in a soil solution<sup>42,44</sup> under flooded/non-flooded conditions.

Paddy rice cultivated under flooded conditions in both soils showed severe symptoms of Reclamation-Akagare disease due to excessive absorption of I, and the yield of rice decreased remarkably compared with paddy rice cultivated under non-flooded conditions.

Hitherto, Reclamation-Akagare physiological disorder of rice plant had often occurred in paddy fields developed on reclaimed land, consolidated farmlands, and in paddy fields amended by using soils transferred from forestlands. As an example, the conditions of paddy field areas subjected to farmland consolidation in Toyama Prefecture are shown in Table 2. This table indicates that the Reclamation-Akagare disease always occurred when the I content exceeded 59 mg/kg dry wt. in leaves or 22 mg/kg dry wt. in soils<sup>15</sup>.

The average content of I in soils of forest land and upland fields of Japan was both 43 mg/kg dry wt.<sup>40,44</sup>, suggesting that the Reclamation-Akagare disease occurs in most of the forest land and upland field soils of Japan when paddy rice is cultivated under flooded conditions.

On the other hand, damage from excessive amount of Br could not be confirmed in these soil culture pot experiments (Table 1). However, the Br content in unpolished rice cultivated on Tochigi Ando soil was 57 mg/kg air dry wt., a value higher than the Br residue tolerance level of 50 mg/kg air dry wt.

The average Br content in soils of forest land and upland fields in Japan was approximately 100 mg/kg dry wt.<sup>40,44</sup>, a value higher than 57 mg/kg dry wt. recorded in Tochigi Ando soil. Therefore, when paddy rice is cultivated in these soils under flooded conditions, the Br

content in unpolished rice may exceed the Br residue tolerance level.

### **Influence of pesticides and fertilizers containing Br on Br residues in crops, soils and ground water**

In Japan, MB amounting to about 3,000 t/year is used for storehouse fumigation of imported agricultural crops, etc. and about 5,000 t/year for soil fumigation to prevent injury during growth due to continuous cropping. These amounts account for about 12% of the whole amount used in the world. Ethylene dibromide (EDB:  $C_2H_4Br_2$ ) had been widely used as a nematicide in vegetable farms with mainly root crops and in orchards. However, the use was discontinued through administrative guidance in 1985, due to possible carcinogenic effects<sup>37</sup>.

Authorized tolerance level of Br residue (ppm:mg/kg on fresh weight basis of edible parts) in foodstuffs in Japan was 50 for wheat in 1970, 50 for rice in 1984, 180 for buckwheat, 80 for corn, 30 for kiwi and 60 for other fruits, in 1992, and thus, the number of crops subjected to regulation has increased.

Analytical research on the conditions of Br residues from pesticides containing Br is limited<sup>27-29,34,37</sup>. The Br content of crops and soils, regardless of the use of MB and EDB in greenhouse and outdoor cultivation of vegetables in farms, is shown in Fig. 1.

Fumigation with MB in a greenhouse is performed 4-5 days before planting of vegetables (on an average, 40 kg/10 a as MB, 33 kg/10 a as Br) and almost all MB may have changed into inorganic Br in the plowed soil at the time of planting. The vegetables absorb these inorganic Br ions through their roots.

The total average content of Br in cucumber regardless of the use of MB was as follows: leaves and stems (mg/kg dry wt.); used 2,290, not used 108; fruits [mg/kg dry wt. (mg/kg fresh wt.)]; used 1,660 (73), not used 81 (3.4).

**Table 1. Contents of I and Br in rice plant cultivated in soil pots under flooded and non-flooded conditions of soils**

Soil of forest land (I, Br, mg/kg dry wt.)	I						Br					
	Straw (mg/kg dry wt.)			Unpolished rice (mg/kg air dry wt.)			Straw (mg/kg dry wt.)			Unpolished rice (mg/kg air dry wt.)		
	Non- flooded (N)	Flooded (F)	(F/N)	Non- flooded (N)	Flooded (F)	(F/N)	Non- flooded (N)	Flooded (F)	(F/N)	Non- flooded (N)	Flooded (F)	(F/N)
Tochigi Ando soil (I 26) (Br 57)	0.36	300	(830)	0.025	0.48	(19)	48	660	(14)	4.2	57	(14)
Brown Forest soil (I 15) (Br 26)	0.37	280	(770)	0.027	0.58	(21)	27	135	(5.0)	3.4	8.7	(2.6)
Average	0.37	290	(784)	0.026	0.53	(20)	38	398	(10.5)	3.8	33	(8.6)

Average of two replicates<sup>40)</sup>.**Table 2. Content of I in rice plants and soils experiencing Reclamation-Akagare disease<sup>15)</sup>**

Presence of Reclamation-Akagare disease		Straw (mg/kg dry wt.)			Plowed soil (mg/kg dry wt.)		
		No. of samples	Average	Min.-Max.	No. of samples	Average	Min.-Max.
Paddy field <sup>a)</sup> with Reclamation-Akagare	Symptoms of Akagare	20	75	37-162	15	15	2.8-31
	No symptoms of Akagare <sup>b)</sup>	15	28	3.7-59	15	15	3.6-22
Control paddy field (basin of the Nagara river)		19	0.55	0.12-2.5	18	1.7	0.48-5.3

a): All the cases of Akagare disease appeared in the 1- to 4-year period after farmland consolidation for paddy fields in Toyama Prefecture.

b): Rice plants and plowed soils were sampled in the same paddy field where Akagare disease occurred.

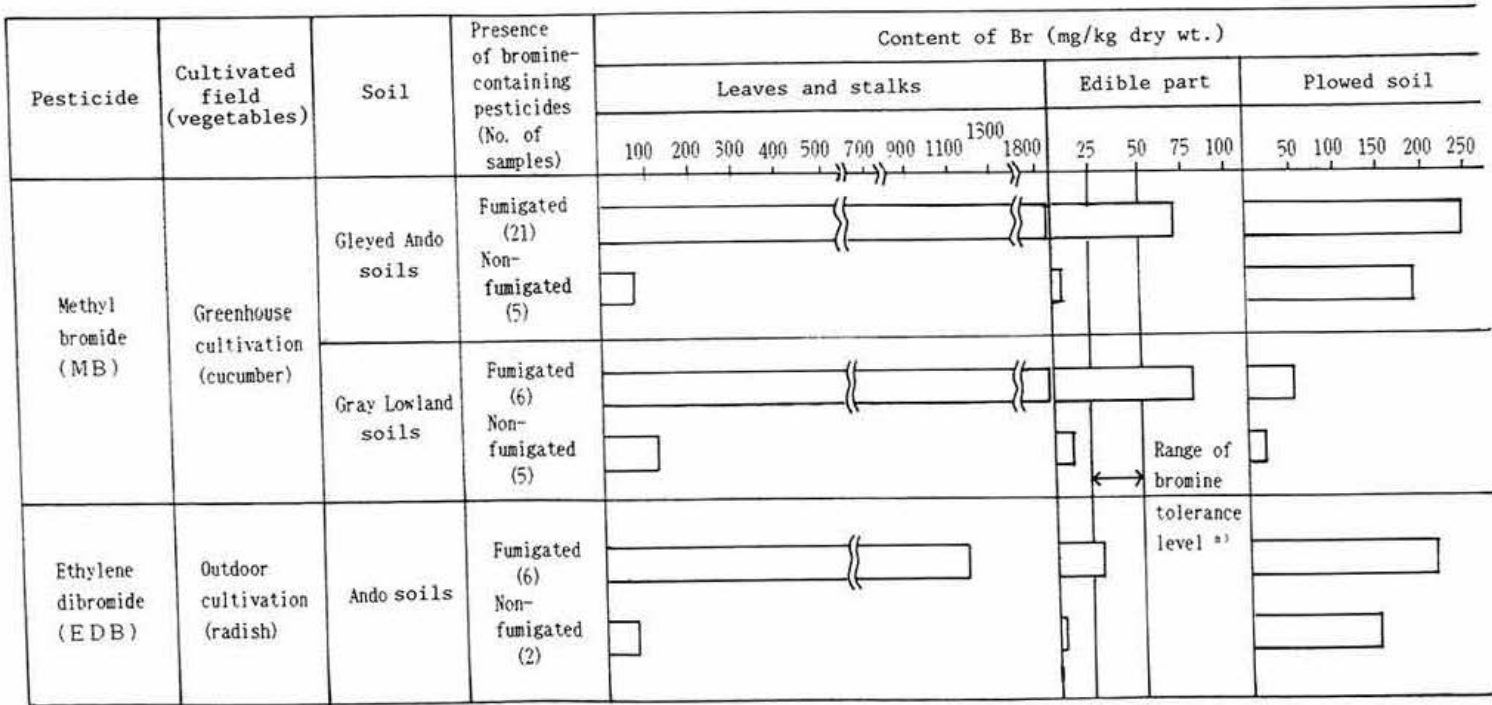


Fig. 1. Influence of Br-containing pesticides on Br residues in vegetables and soils<sup>37)</sup>  
 a): Range of bromine residue tolerance level in the world.

The content was more than 20 times higher when MB was used than in the absence of MB use, in all cases.

Also, in the EDB-treated upland fields, the Br content of vegetables was different among the vegetables, but generally was not lower than that of vegetables in the MB-fumigated greenhouse fields.

Though there is no Br residue tolerance level for vegetables such as cucumbers in Japan, the

average Br content of vegetables exceeded the tolerance level of fruits and crops in Japan and in the world.

In Fig. 2, the input and output of Br derived from the soil fumigated with MB in greenhouse cultivation of cucumbers are shown. By the end of the cucumber cultivation, 3% of the Br originating from the applied MB was absorbed by the top of cucumber plant and 17% remained in the plowed soil. The value of 17%

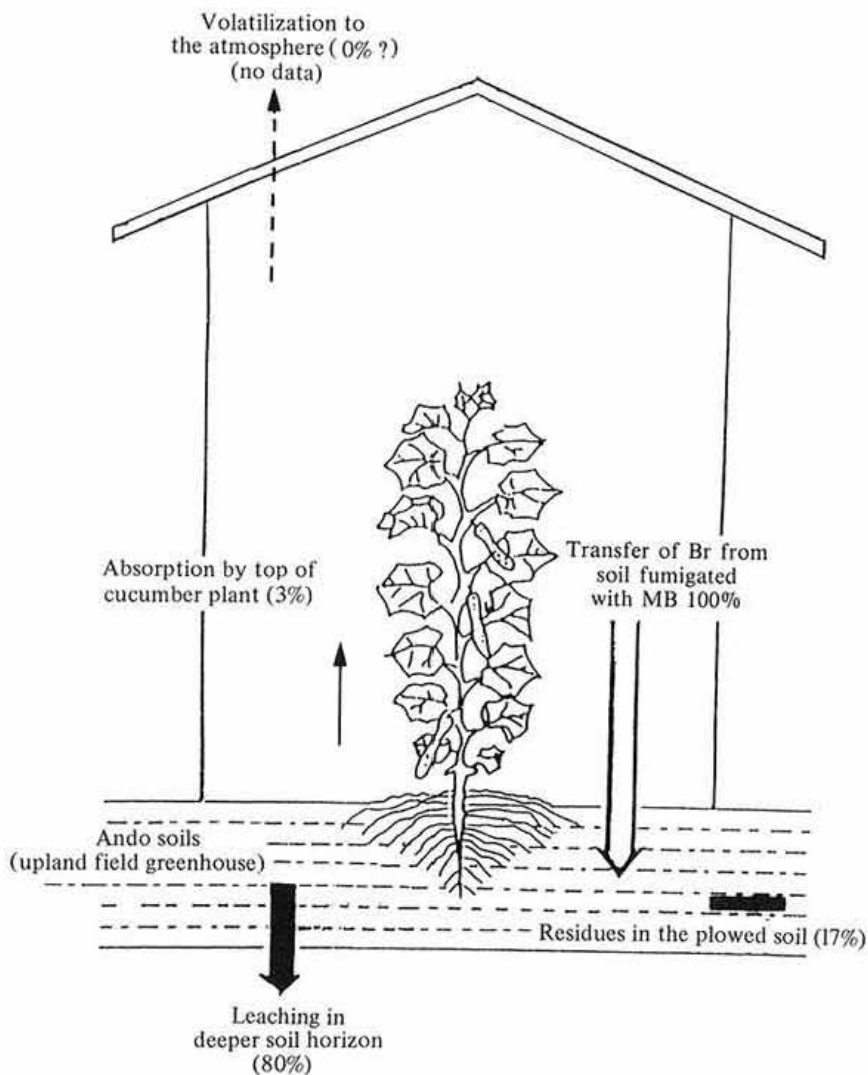


Fig. 2. Input and output of Br derived from soil fumigated with MB in greenhouse cultivation of cucumber<sup>37)</sup>

was calculated from the difference in Br content in the plowed soil when MB was used or not applied. Br residue percentage was different depending on the kinds of soils (Gleyed Ando soils 24%, Gray Lowland soils 15%)<sup>37)</sup>.

As the greenhouse was tightly closed before the planting of cucumbers, there was almost no volatilization to the atmosphere, and therefore, the residual amount of 80% may have leached into the deeper soil horizon with irrigated water.

Br atom originating from the breakdown of MB, halons, etc. exerts a destructive effect on the ozone layer (about 40 times that of Cl originating from the breakdown of flon gas) and it is estimated that more than 50% MB production is volatilized into the atmosphere<sup>1)</sup>.

Restriction of MB production and consumption amount in the world to the level of 1991 from 1995 onward was promulgated by the 4 parties in the contract for the Montreal protocol in Nov. 1992. The authors assumed that the amount of volatilized MB in the atmosphere from the greenhouse was nearly 0<sup>37)</sup>, but this value should be confirmed by actual measurements. It is preferable to measure the amount of volatilized MB originating from natural

generation in the soil and sea surface water.

Results of the analytical survey on the ground water pollution by Br originating from the Br-containing pesticides are shown in Table 3. The average concentration of Br in the ground water in the areas where a large quantity of Br-containing pesticides was used was 2.29 mg/L, a value 250 times higher compared with the average value of 0.0092 mg/L in the ground water in the areas where a smaller amount of Br-containing pesticides was used, and 7 times compared with the average value of 0.32 mg/L in the areas where an amount in the intermediate range was used (upland fields and greenhouses where Br-containing pesticides were applied were scattered).

The Br concentration in ground water (well water) used for drinking water in the areas in which a large quantity of Br-containing pesticides was applied was 4.26 mg/L, and it became equivalent to 9.4 mg of Br intake when 2.2 L/day of this well water was absorbed. This intake of Br is very large, and almost at the same level as the 10 mg of Br intake when humans absorb 200 g/day of rice with 50 mg/kg of Br, which corresponds to the tolerance level of Br in rice.

Table 3. Influence of bromine-containing pesticides on Br concentration in ground water<sup>37)</sup>

Amount of bromine-containing pesticides applied	Region (No. of sampling sites)	Sampling date (Sampling times)	Depth of well (m)	Average concentration (mg/L) (Min. - Max.)
Large	Miyazaki Pref. (5)	May 2, 1979 ~ April 18, 1980 (6 times at intervals of 2 months)	4-8	2.29 (0.57-5.87)
Medium	Kansai region (5)	March 30, 1984 (1 time)	5-12	0.32 (0.05-1.0)
Small	Kanagawa Pref. (21)	June 30, 1983 ~ July 1, 1983 Nov. 1, 1983 ~ Nov. 2, 1983 (2 times)	4-80	0.0092 (0.0023-0.030)

Furthermore, when the concentration of bromide in untreated water for city water increased, the total generation of trihalomethane including bromide trihalomethane during the chlorination process increased remarkably<sup>3)</sup>. We must pay attention to Br pollution of untreated water such as lake water, river water and ground water.

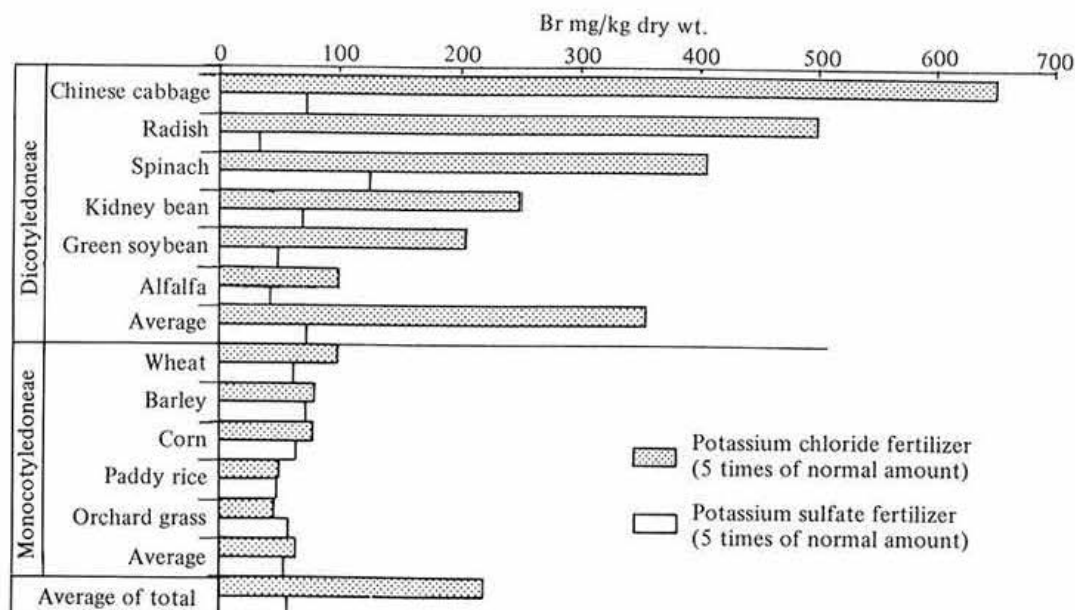
KCl fertilizer contains 950 mg/kg dry wt. of Br and K<sub>2</sub>SO<sub>4</sub> fertilizer contains 51 mg/kg dry

**Table 4. Br content in chemical fertilizers and reagents<sup>37)</sup>**

Chemical fertilizer	Reagent	Br (mg/kg dry wt.)
Potassium chloride	Fertilizer	950
Potassium chloride	Purified reagent	110
Potassium sulfate	Fertilizer	51
Potassium sulfate	Purified reagent	0.28
Ammonium sulphate	Fertilizer	0.36
Superphosphate	Fertilizer	0.95
Calcium hydroxide	Fertilizer	3.2
Potassium nitrate	Purified reagent	0.31

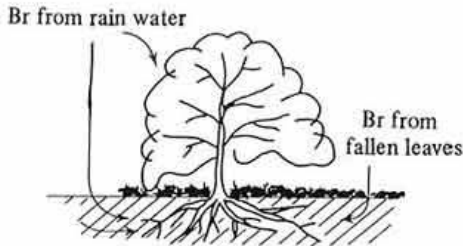
wt. of Br (Table 4)<sup>37)</sup>. When large amounts of the KCl fertilizer were applied, the Br content in the leaves and stalks of the crops increased by the application of potassium chloride fertilizer, especially in Dicotyledoneae (Fig. 3). However, the degree of increase was much lower than when pesticides containing bromine were used. It was estimated that the values in farms were at most 200 mg/kg dry wt.<sup>37)</sup> which were less than 1/10 of those of crops cultivated in MB-fumigated soil. Based on these results, the author classified the Br content level in plant leaves into four groups according to the main sources of absorption of Br (Fig. 4)<sup>34)</sup>. (1) 15 mg/kg dry wt.: Natural sources. (2) 50 mg/kg dry wt.: Chemical fertilizer (potassium chloride) application. (3) 500 mg/kg dry wt.: Dissolved soil Br under flooded conditions (Reclamation paddy fields). (4) 2,000 mg/kg dry wt.: Pesticide (MB, EDB) application.

Influence of fertilization with KCl on the concentration of Br in the ground water appears to be negligible. For example, the concentration of Br in the ground water of the



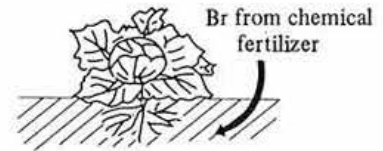
**Fig. 3. Influence of potassium chloride and potassium sulfate fertilizer application to the soil on the Br content in crops (Neubauer's pot experiment)<sup>37)</sup>**

- 1) 15 (1~100) mg/kg dry wt.  
Natural sources  
 Uncultivated (forest) land  
 - Wild plants -



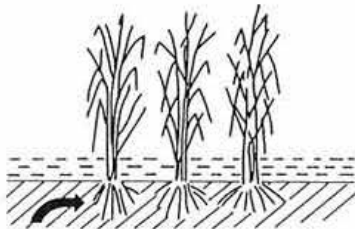
Soil Br (Br in soil is present in forms not readily absorbed by plants under non-flooded conditions)

- 2) 50 (10~200) mg/kg dry wt.  
Chemical fertilizer (Potassium chloride) application  
 Cultivated (upland fields, paddy fields, grasslands) land  
 - Many crops -



Potassium chloride fertilizer Br (Br 950 mg/kg dry wt.)

- 3) 500 (100~2000) mg/kg dry wt.  
Dissolved soil Br under flooded conditions  
 Reclamation, farmland consolidation  
 paddy fields  
 - Rice plants -



Soil Br (Large amounts of Br in soil which is dissolved into soil solution under flooded conditions, are easily absorbed by plant)

- 4) 2000 (200~5000) mg/kg dry wt.  
Pesticide (MB, EDB) application  
 Greenhouse and upland fields  
 - Vegetables, tree crops -

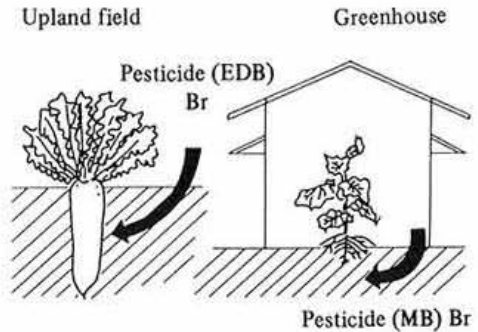


Fig. 4. Classification of Br content level in plant leaves according to main sources of absorption of Br<sup>34)</sup>

Ashigara plains in Kanagawa Pref. where paddy fields and tree gardens are spread out widely in this basin was very low (Table 3), and the amount of Br applied per unit area of KCl fertilization was less than 1/1,000 of the amount of MB fumigation<sup>37)</sup>.

### Analysis of accumulation in soil and transfer to plants of long-lived <sup>129</sup>I

The reprocessing plants of spent nuclear fuel

are operated mainly in developed countries where a large number of nuclear reactors are present and nuclear weapons are produced.

In Japan, already the Tokaimura/Ibaraki fuel reprocessing plant has been in operation and the Rokkashomura/Aomori large plant is under construction. In the reprocessing plants, spent uranium fuel rods are gathered from nuclear reactors in various areas, and the extraction of radioactive <sup>239</sup>Pu generated during the operation of the reactor and of the remaining <sup>235</sup>U,



as well as the removal of nuclear fission products requires care.

As the fuel rods in the reprocessing plants are cooled for more than one year, the short-lived nuclides have already undergone decay, while the long-lived nuclides still remain. Therefore, the release of long-lived nuclides into the environment, especially volatile nuclides during subsequent chemical reprocessing of spent nuclear fuel rods may occur.

Among all the nuclides released from the stacks into the atmosphere during the normal operation of the reprocessing plants,  $^{129}\text{I}$  shows the maximum contribution ratio of all the nuclides in the internal radiation dose for the public through food ingestion (Fig. 5)<sup>5)</sup>. It is necessary to obtain quantitative data on the transfer and accumulation of  $^{129}\text{I}$  from the atmosphere to the soil-plant system from a long-term perspective.

However, the current level of  $^{129}\text{I}$  in the environment is still low<sup>8)</sup> and it is difficult to determine  $^{129}\text{I}$  in soils and plants except for those in the vicinity of reprocessing plants, and few data are available<sup>6)</sup>. Especially, no studies on the transfer of  $^{129}\text{I}$  to rice plant which is the staple food of the Japanese people are available in the literature. Furthermore, because it was considered that there were large differences

in the movement of  $^{129}\text{I}$  between the paddy soil-paddy rice system and the upland soil-crop system, Japan was requested to undertake a study independently of other nations. Therefore, it is important to develop a prediction method based on the quantitative data of stable I ( $^{127}\text{I}$ ) in the environment to analyse the dynamics of radioactive  $^{129}\text{I}$ .

Analysis of the behavior of stable I in the soil-plant (paddy rice) system was used for acquisition of basic data for the radiological assessment of the radiation dose and nuclear safety inspection<sup>26,35,38,39)</sup>.

The transfer pathway of  $^{129}\text{I}$  (identical with that of stable  $^{127}\text{I}$  and radio  $^{131}\text{I}$ ) to the rice grains is shown in Fig. 6. As data about the direct deposition of I in the atmosphere onto leaves and ears of rice, especially the wet deposition of I into rainwater are very limited<sup>11,12)</sup>, further studies concerning these aspects should be carried out.

In the investigation on the Chernobyl nuclear reactor accident, it was demonstrated that the deposition of  $^{131}\text{I}$  onto the wheat tops was due to the direct deposition of gaseous and particulate  $^{131}\text{I}$  in the air, and not due to the direct wet deposition of  $^{131}\text{I}$  in rainfall<sup>41)</sup>. After the completion of analyses on the direct deposition, the contribution of three transfer path-

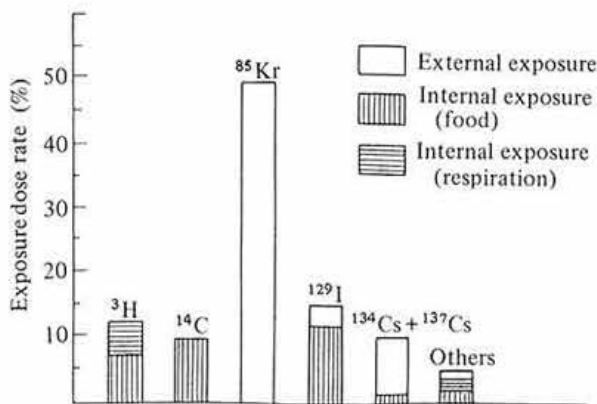


Fig. 5. Contribution ratio to human population exposure doses of each nuclides emitted from the stack of reprocessing plants during normal operation<sup>5)</sup>

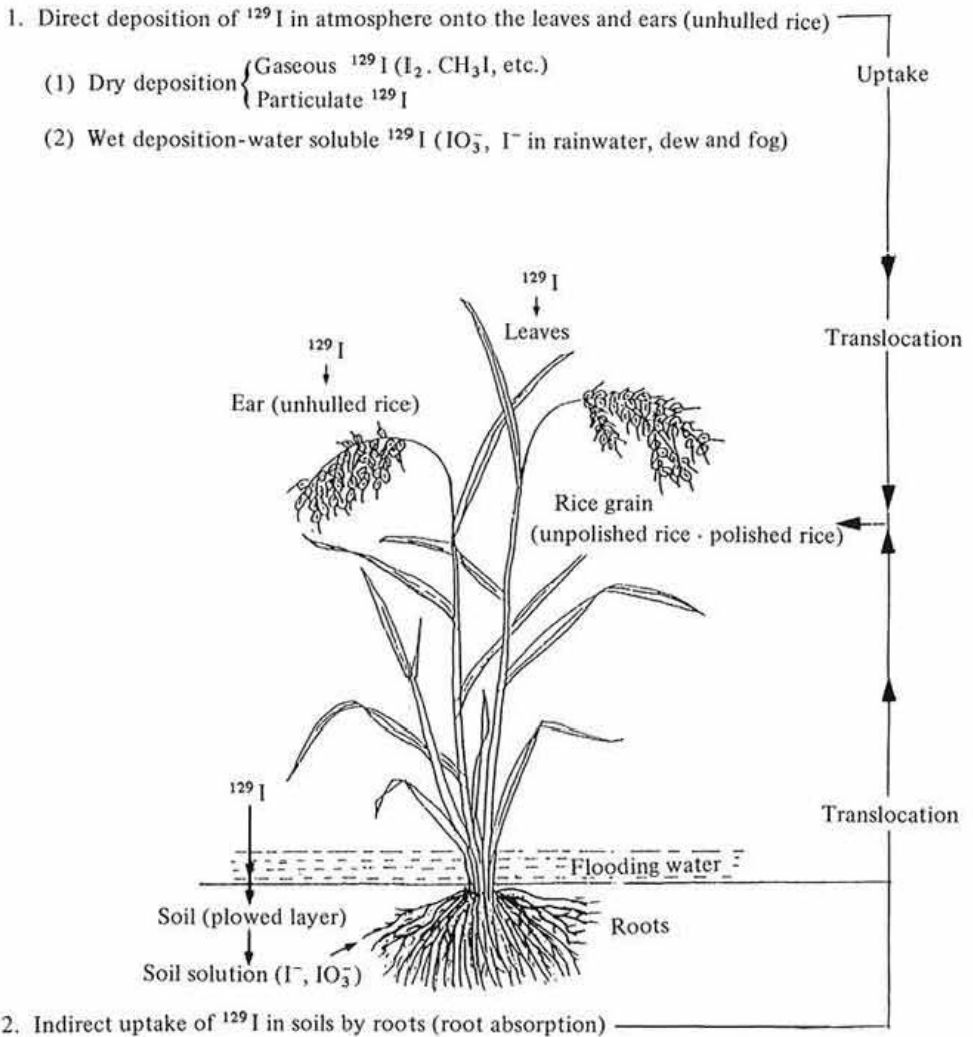


Fig. 6. Transfer pathway of  $^{129}\text{I}$  in the atmosphere to rice grain<sup>38)</sup>

ways, i.e., dry deposition, wet deposition and indirect uptake by roots to the total  $^{129}\text{I}$  content in rice grains should be studied quantitatively.

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