# Iodine and Bromine in Soil-Plant System with Special Reference To "Reclamation-Akagare Disease" of Lowland Rice

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## Introduction

According to a series of study on radioactive fall-out in soil-plant system especially in connection with submerged soil and lowland rice, it was clarified by the author *et al.* that under submerged soil condition rice plant shows some characteristic features with regard to the root uptake of such fission products as $^{90}Sr$, $^{137}Cs$ and $^{131}I$.[1)-4)]

The preferential uptake of radio-iodine by lowland rice from submerged soil has been reported in 1963,[3)] and its mechanism was well explainable on the basis of the finding that the behavior of iodine in the system of soil and plant is primarily governed by oxidative retention and reductive release of iodine in soil in association with soil organic matter, as shown schematically in Fig. 1.[4)]

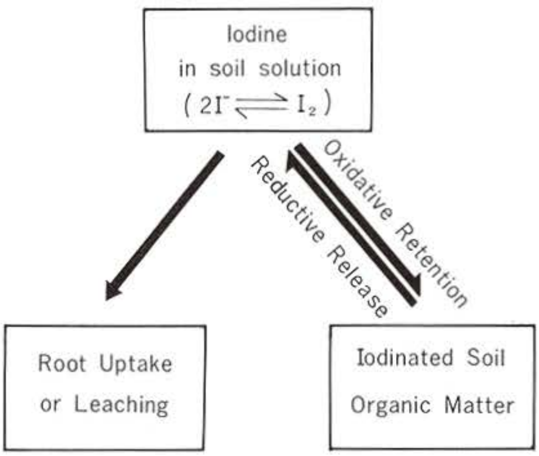


Fig. 1. Schematic mode of iodine behavior in soil-plant system.

Furthermore, it seemed to be noteworthy that the iodine toxicity in rice plant can be observed easily under submerged soil condition even at a minute dose of iodine application of about 10 ppm to the soil in the forms of iodide or elementary iodine. In addition, the symptoms due to iodine toxicity were similar to that of the so-called "Kaiden- or Reclamation-Akagare disease", a physiological disorder, of lowland rice.

Incidentally, "Kaiden-Akagare" means the withering with reddish-brown tint induced by the reclamation of upland into paddy field. Typical symptom in the initial stage of the disease is the appearance of small necrotic spots on leaf blades of the rice plant at usually 2 to 3 weeks after transplantation, and this disease occurred in some areas of newly-reclaimed paddy soils mostly in volcanic ash origin. Moreover, it has been noticed that the disease disappears usually within several years after the reclamation, and that rice plant tolerates differently among its varieties.

Meanwhile, during the course of a study on neutron activation analysis of such trace elements as strontium and barium in some plant samples originated from volcanic ash soils, bromine was found to be one of the most important interfering elements for the analysis.

Subsequently, it was confirmed that some upland soils mostly of volcanic ash origin along with the associated plant materials are remarkably rich in bromine at more than

50 ppm.[5),7)] Similar finding concerning volcanic ash soils rich in bromine has been reported also by Yamada,[10)] and the correlative occurrence of iodine and bromine in soil has been demonstrated by Vinogradov.[8)]

All the findings mentioned above as well as the related information led the author to study the behaviors of iodine and bromine in the soil-plant system by means of radioisotope techniques including neutron activation analysis and tracer method with $^{131}I$ and $^{82}Br$. This review includes mainly the iodine and bromine contents in the soil and plant with regard to the "Reclamation-Akagare disease" of lowland rice, and deals with some of the factors affecting their variable nature.

## Iodine and bromine in soil

Since the analysis of bromine by the neutron activation technique is easy compared to that of iodine, the data were obtained largely with regard to bromine. As for the analytical procedure,[7)] dried sample was irradiated in a reactor and decomposed with $H_2SO_4$-$HNO_3$ mixture after cooling for 2 to 5 days. The distillate was then subjected to gamma-spectrometry. In the case of iodine, without cooling the chemical separation of the iodine fraction in the distillate was made with alternate oxidation-reduction processes.

Bromine contents in various soil samples, which were collected mosty from the eastern parts of the Mainland, are shown in Table 1.[11)]

**Table 1. Bromine content in soils**

| Location | Field condition* | Soil horizon | Soil origin | Br ppm in dry basis |
|---|---|---|---|---|
| Tanashi, Tokyo | Upland | Surface | Volcanic ash | 85 |
| 〃 | 〃 | Subsoil | 〃 | 52 |
| Nishinasu | 〃 | Surface | 〃 | 139 |
| 〃 | 〃 | Subsoil | 〃 | 73 |
| 〃 | Paddy-R | Surface | 〃 | 119 |
| Mito, Ibaragi | 〃 -R | 〃 | 〃 | 115 |
| Chiba | Upland | 〃 | 〃 | 150 |
| 〃 | 〃 | Subsoil | 〃 | 33 |
| Kuriyagawa | 〃 | Surface | 〃 | 114 |
| Yatsugatake | 〃 | 〃 | 〃 | 55 |
| Morioka | 〃 | 〃 | 〃 | 92 |
| 〃 | Paddy-R | 〃 | 〃 | 90 |
| 〃 | 〃 -A | 〃 | 〃 | 26 |
| Utsunomiya | 〃 -A | 〃 | 〃 | 16 |
| Iwanuma | 〃 -A | 〃 | Alluvial | 23 |
| Chiba | 〃 -A | 〃 | 〃 | 12 |
| Nagano | 〃 -A | 〃 | 〃 | 8 |
| Yamaguchi | 〃 -A | 〃 | 〃 | 5 |
| Takashigahara | Upland | 〃 | Non-volcanic | 8 |
| Fujisawa, Iwate | 〃 | 〃 | 〃 | 10 |

* R and A denote newly reclaimed and aged fields respectively.

Bromine content in 20 soil samples varies in the range of 5 to 150 ppm. The average for 8 surface soils, including aged paddy and non-volcanic upland soils, amounts to 14 ppm while its value for 9 surface soils of volcanic ash origin sampled from upland or newly-reclaimed paddy fields is approximately 100 ppm. Vinogradov[8)] reported that the bromine content in about 50 samples of Russian soils varied in the range of 0.27 to 43 ppm with an average value of about 1 ppm.

Consequently, it can be pointed out probably that in normal soils the bromine content of more than 50 ppm is extraordinarily high. Further, it might be noteworthy that surface soils with high humus are rich in bromine, and its content is likely to decrease owing to the conversion from upland to paddy field, especially to the aging under submerged soil condition.

This correlative occurrence of bromine with soil humus was demonstrated more clearly by other investigators,[8),10)] and the trend of poor occurrence of bromine in paddy soils was pointed out also by Yamada.[10)]

Table 2 shows iodine contents in some volcanic ash soils. The iodine content in 5 soil samples ranged from 14 to 25 ppm and its relative abundance to bromine was from 0.12

**Table 2. Iodine content and its relative abundance to bromine in some volcanic ash soils**

| Location | Field Condition | I ppm in dry basis | I/Br |
|---|---|---|---|
| Tanashi, Tokyo | Upland | 14.1 | 0.17 |
| Nishinasu | 〃 | 17.1 | 0.12 |
| 〃 | Paddy-R | 14.7 | 0.12 |
| Mito, Ibaragi | 〃 -R | 17.4 | 0.18 |
| Yatsugatake | Upland | 22.6 | 0.41 |

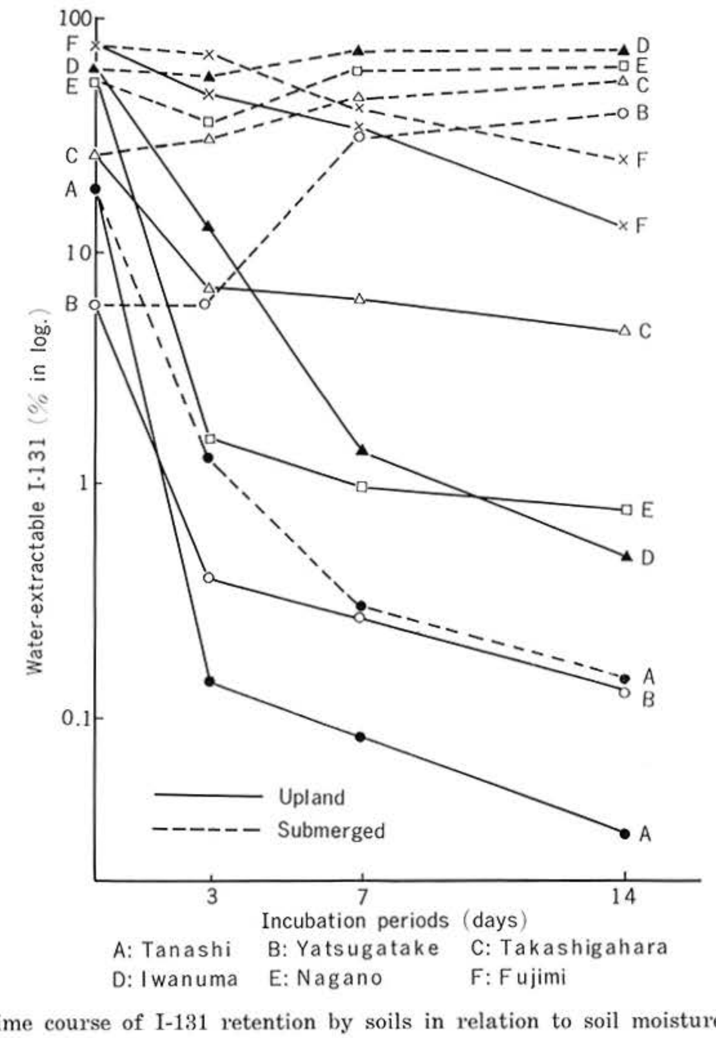


Fig. 2. Time course of I-131 retention by soils in relation to soil moisture condition.

to 0.41. Analysis of the soil iodine has been made mainly from the viewpoint of endemic goiter, and Vinogradov has reported on the basis of the compilation of world-wide data of about 2,000 soil samples that the iodine content in soils varies in the range of 0.1 to 50 ppm with an average value of 5 ppm.

Therefore, the iodine values found in Table 2 are also remarkably high, and the relative abundance of iodine to bromine is rather low in these soils in comparison with that of more than unity reported by Vinogradov.

### Retention and release of iodine in soil

Since the information on the behavior of iodine in soil was limited, soil-incubation experiments using 5 soils and $^{131}$I were conducted in order to examine the time course of iodine retention by soil under both upland and submerged conditions.[4] As shown in Fig. 2, under upland condition the added carrier-free $^{131}$I in iodide form was retained largely by sandy soil. The rapid rate of retention was observed in the initial incubation periods, generally within 5 days. Then, the decrease of water-extractable iodine seemed to continue at a slow rate of exponential pattern in at least four soils.

Meanwhile, under submerged condition the retention was highly inactive as compared with that under upland condition. For example, after 2 weeks the ratios of submerged to upland treatments in the water-extractable fraction were from the lowest of 2 in Fujimi soil to the highest of about 300 in Yatsugatake soil.

Furthermore it seemed to be noteworthy that in some soils, especially in Yatsugatake soil, the retained $^{131}$I was released gradually into the aqueous phase as the incubation proceeded. This release of the retained iodine due to soil submergence was demonstrated more clearly in an experiment using Nishinasu soil as shown in Fig. 3.[7]

Almost all of the added $^{131}$I was retained by

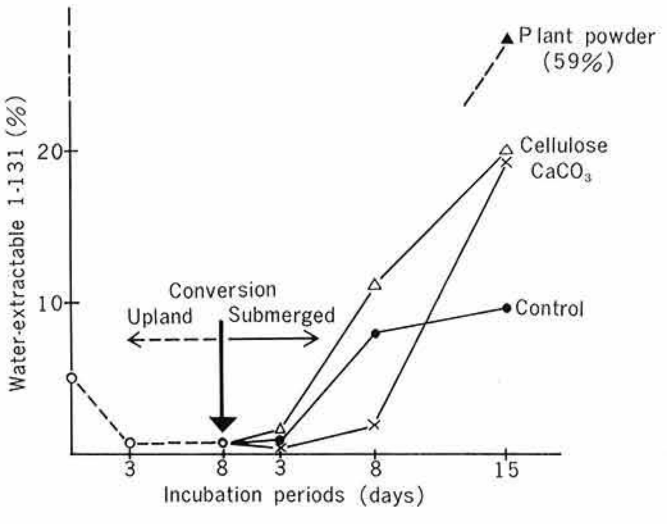


Fig. 3. Retention and release of I-131 in Nishinasu soil.

soils as the time gradually elapsed.

The degree of retention was variable among the soils, resulting in a range, in terms of the percentage of water-extractable fraction after 2 weeks, from 0.034% for Tanashi soil of volcanic ash origin to 12.1% for Fujimi the soil during the initial incubation periods under upland condition, but a part of the retained $^{131}$I was released due to the subsequent treatment of submergence. Moreover, it was found that the incubation with such materials as calcium carbonate and cellulose or plant powders for stimulating the reductive soil condition enhanced its release from the soil.

In the case of plant powder, about 60% of soil iodine was released by the submerged incubation treatment for 15 days, resulting in 4 ppm of iodine in the aqueous phase, which is well comparable to the concentration showing the iodine toxicity under the water culture condition, i.e., approximately more than 1 ppm.[4),7)]

In addition to this finding, Watanabe and the author detected similar concentrations in the soil solution derived from "Reclamation-Akagare" soils and identified its chemical form of iodide by UV-spectroscopy.[9)] An additional evidence for the reductive release of soil iodine as well as its association with soil organic matter is given in Table 3.[4)]

In contrast to iodine, the degree of retention and release of bromine in the soil was remarkably low. In both Tanashi and Nishinasu soils, less than 30% of the $^{82}Br$ in bromide form

**Table 3. Extraction of I-131 retained in soil with various agents***

| Treatment (0.1 M) | Extractable I-131 (%) | |
|---|---|---|
| | Yatsugatake | Tanashi |
| Water | 0.1 | 0.05 |
| HCl | 0.3 | 0.14 |
| NaOH | 21.7 | 3.0 |
| KI | 29.1 | 15.1 |
| NaF | 9.3 | 0.92 |
| $Na_4P_2O_7$ | 45.0 | 12.2 |
| $Na_2S_2O_3$ | 32.8 | 19.8 |
| $SnCl_2$ | 30.1 | — |
| $(NH_4)_2S$ | 31.7 | — |

* The soil with I-131 was incubated under upland condition for 10 days prior to extraction.

**Table 4. Bromine content in plants**

| Plant species | Associated soil | Growth condition | Plant portion | Br ppm in dry basis |
|---|---|---|---|---|
| Wheat | Tanashi | Pot | Whole-Y** | 170 |
| 〃 | 〃 | 〃 | Straw | 100 |
| 〃 | 〃 | 〃 | Ear | 25 |
| Upland rice | 〃 | 〃 | Whole-Y | 190 |
| 〃 | 〃 | 〃 | Straw | 134 |
| 〃 | 〃 | 〃 | Ear | 35 |
| Lowland rice | 〃 | 〃 -Cl* | Whole-Y | 180 |
| 〃 | 〃 | 〃 | 〃 -Y | 380 |
| 〃 | 〃 | 〃 | Straw | 348 |
| 〃 | 〃 | 〃 | Ear | 97 |
| 〃 | Nishinasu | Field | Whole-Y | 115 |
| 〃 | Mito | 〃 | 〃 -Y | 238 |
| 〃 | 〃 | 〃 | Straw | 172 |
| 〃 | 〃 | 〃 | Ear | 15 |
| Kidney bean | Tanashi | Pot | Whole-Y | 145 |
| Leaf vegetable | 〃 | 〃 | 〃 -Y | 130 |
| 〃 | 〃 | 〃 | 〃 | 60 |
| 〃 | 〃 | Field | 〃 | 40 |
| Pasture | Chiba | 〃 | 〃 | 91 |
| Weed | 〃 | 〃 | 〃 | 35 |

* Cl denotes chloride-fertilizer, i.e., $NH_4Cl$. Unless otherwise specified in pot samples, $(NH_4)_2SO_4$ was applied.

** Aerial whole plant was sampled for the analysis, and Y denotes the sampling at younger growth stage.

added at a dose of 1.5 ppm to the soil was retained by the incubation treatment, and no significant difference was found between the upland and submerged conditions.[5] The percentage release of bromine from Nishinasu soil by the submerged incubation treatment was 10% contrary to about 60% obtained for iodine.

## Iodine and bromine in plant

As shown in Table 4, bromine content in 20 plant samples originated from some volcanic ash soils in the Kanto province varies in the range of 15 to 380 ppm and more than 50 ppm are detectable in most of the samples.[11] Referring to other data on bromine content in land plant grown under natural condition without the treatment of bromide fumigation, these values of more than 50 ppm are extraordinarily high.

In addition, the straws of rice and wheat plants are rich in bromine as compared with their ear, and its content is likely to diminish due to the aging of the plants and the accompanied chloride in soil.

The effects concerning some of these soil conditions on the uptake of iodine and bromine were demonstrated clearly in an experiment shown in Table 5.

Using two volcanic ash soils, rice plants were grown for 6 weeks under both upland and submerged conditions in combination with two types of fertilizers, i.e., sulfate and chloride series. The availabilities of iodine and bromine to rice plant were shown to be highly variable due to the soil condition. The contents in the dried shoots ranged from 0.4 to 112 ppm of iodine and from 46 to 727 ppm of bromine, showing the competing effect of chloride especially on bromine uptake and the stimulating effect of soil submergence. Particulary, it was noteworthy that this preferential uptake due to soil submergence is predominant in the case of iodine. This resulted in a fact that the relative abundance of iodine to bromine in the plant is remarkably low under the upland condition in comparison with the value found under the submerged condition, which is close to that in the soil. Furthermore, the iodine contents obtained under submerged condition are extraordinarily high as compared with the reported value of less than 5 ppm in the normal land plant.

Judging from all the facts mentioned above and other related experimental evidences mainly on the toxicities of iodine and bromine in relation to rice variety,[5),6)] it can be concluded that the iodine toxicity is responsible primarily for the "Reclamation-Akagare disease" of

**Table 5. Root uptake of iodine and bromine from soil by rice plant in relation to soil conditions**

| Soil condition | Fertilizers applied* | Content (ppm) I | Content (ppm) Br | I/Br |
|---|---|---|---|---|
| a) Nishinasu soil | | | | |
| Upland | Cl | 1.3 | 46 | 0.03 |
| | $SO_4$ | 2.2 | 280 | 0.01 |
| Submerged | Cl | 17.1 | 90 | 0.19 |
| | $SO_4$ | 112 | 727 | 0.15 |
| b) Tanashi soil | | | | |
| Upland | Cl | 0.7 | 77 | 0.009 |
| | $SO_4$ | 0.4 | 108 | 0.004 |
| Submarged | Cl | 7.3 | 48 | 0.15 |
| | $SO_4$ | 17.4 | 260 | 0.07 |

* Cl and $SO_4$ denote chloride and sulfate-fertilizers composed of $NH_4Cl$, KCl and fused magnesium phosphate and of $(NH_4)_2SO_4$, $K_2SO_4$ and superphosphate respectively.

lowland rice.

## References


1) Tensho, K. and Mitsui, S.: The uptake of strontium and cesium by lowland rice. FAO expert committee on radioactive materials food and agriculture. Suppl., 158–181 (1960).

2) ———, Yeh, K. L. and Mitsui, S.: The uptake of strontium and cesium by plants from soil with special reference to the unusual cesium uptake by lowland rice and its mechanism. *Soil and Plant Food, Tokyo* **6**, 176–183 (1961).

3) ———, ——— and Nishimura, Y.: Unusual root uptake of I-131 by lowland rice. *Proc Soc. Sci. Soil and Manure, Japan* **9**, 17 (1963). [In Japanese. Abstract of oral presentation.]

4) ——— and ———: Radio-iodine uptake by plant from soil with special reference to lowland rice. *Soil Sci. Plant Nutri. Tokyo*, **16**, 30–37 (1970).

5) ———, ——— and Otohata, K.: Behaviour of bromine in soil-plant system. *Proc Soc. Sci. Soil and Manure, Japan* **16**, Part 1, 63 (1970). [In Japanese. Abstract of oral presentation.]

6) ———, ——— and Oshima, H.: Oodine toxicity in relation to "Reclamation-Akagare disease" of lowland rice. *Proc Soc. Sci. Soil and Manure, Japan* **16**, Part 2, 34 (1970). [In Japanese. Abstract of oral presentation].

7) ——— and ———: Study on iodine and bromine in soil-plant system in relation to the "Reclamation-Akagare disease" of lowland rice by means of RI-techniques. *Radioisotopes*, **19**, 574–579 (1970).

8) Vinogradov, A. P.: The geochemistry of rare and dispersed chemical elements in soils, 2nd Ed. (Translated from Russian) 45–64, Consultant Bureau, New York, (1959).

9) Watanabe, I. and Tensho, K.: Further study on iodine toxicity in relation to "Reclamation-Akagare disease" of lowland rice. *Soil Sci. Plant Nutr. Tokyo*, **16**, 199–201 (1970). plants and soil. *Talanta* **15**, 1135–1141

10) Yamada, Y.: Occurrence of bromine in plants and soil. Talanta **15**, 1135–1141 (1968).

11) Yeh, K. L. and Tensho, K.: Variable bromine content in soil, plant and milk. *Soil Sci. Plant Nutr. Tokyo*, in press.