Deposition of Iodine-131 on Plant Leaves (1)

Yoichiro OHMOMO and Masamichi SAIKI

Tokai Laboratory, Tokai Branch, National Institute
of Radiological Sciences, Tokai, Japan

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This experiment was carried out in order to know the deposition and the distribution of $\text{CH}_3^{131}\text{I}$, $^{131}\text{I}_2$ and Na^{131}I on plant leaves such as spinach, Chinese cabbage, Komatsuna and camellia quantitatively and autoradiographically. $\text{CH}_3^{131}\text{I}$ deposited on Chinese cabbage, spinach and camellia leaves on the similar level. On the contrary, $^{131}\text{I}_2$ deposited strongly on spinach and Chinese cabbage leaves and slightly deposited on camellia leaves. It was suggested that $^{131}\text{I}_2$ deposited more strongly on leafy vegetables than $\text{CH}_3^{131}\text{I}$. Both $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ were concentrated in the vein of the leaves. No radioactivity was detected on the leaves which were steeped in the Na^{131}I solution.

Effective removal of iodine-131 from the contaminated leaves was not clearly recognized by washing them with detergent, CCl_4 , ether, 0.5N HCl and 0.5N NaOH.

1. Introduction

Iodine-131, which is one of the most popular fission products, is an important radionuclide to human body, because it is concentrated in thyroid gland. If it was released in the air, it could be taken up through inhalation, drinking water and food such as milk and vegetables. Studies of chemical form, distribution and concentration of iodine-131 in different environmental substances or organisms are required for the estimation of radiation hazard due to radioactive iodine, which is supplied from nuclear explosion test or released in the air from nuclear power plant especially in an emergency. In a series of nuclear explosion tests sometimes significant amount of radioactive iodine was detected in cow's milk⁽¹⁾. And the maximum radiation dose to thyroid gland of infant was estimated⁽²⁾. Distribution of iodine-131 in milk and the deposition of $^{131}\text{I}_2$ on plant leaves have been studied. However, information on the influence of the chemical form of iodine upon the deposition of different kinds of plant leaves is quite limited. This experiment was carried out in order to know the deposition and the

distribution of organic and inorganic iodine-131 on plant leaves such as spinach, Chinese cabbage, Komatsuna, camellia and also filter paper and carbon filter paper. Removal of these iodine compounds from leaves was also examined by washing them with several kinds of chemicals.

2. Procedure

2.1 Apparatus

Outline of the experimental apparatus is shown in Fig. 1. Evaporated gases were introduced to exposure vessel through glass wool, which is connected to traps, by suction at the flow rate of 1,000 ml/hour.

2.2 Plant leaves

Spinach (*Spinacia oleracea* L.), Chinese cabbage (*B. rapa* var. *amplexicaulis* subvar. *Pe-tsai*), Camellia (*Camellia japonica* L.) and Komatsuna (*B. rapa* var. *Komatsuna*) leaves were used. Especially in case of a Chinese cabbage, a small whole was used.

2.3 Exposure

2.3.1 For the primary qualitative experiment

Seven μCi of $\text{CH}_3^{131}\text{I}$ containing 125 mg of

Fig. 1 Outline of the experimental apparatus.

stable CH_3I was evaporated by warming over 40°C and contacted with spinach leaves, camellia leaves, filter paper and carbon filter paper (carbon powder attached filter paper) in exposure vessel for 60 minutes. Three point of five μCi of $^{131}\text{I}_2$ containing 20 mg of stable I_2 was released by adding dropwise 1 or 2 ml of 30% H_2O_2 to the Na^{131}I solution which contained 0.5 g of $\text{Fe}_2(\text{SO}_4)_3$ and 0.5 ml of concentrated H_2SO_4 and contacted with the samples mentioned above.

2.3.2 For the quantitative experiment

Twenty μCi of $\text{CH}_3^{131}\text{I}$ containing 50 and 225 mg of stable CH_3I was evaporated and exposed to spinach, Chinese cabbage, filter paper and carbon filter paper for 60 minutes. Twenty μCi of $^{131}\text{I}_2$ containing 100 mg of stable I_2 and 52 μCi of $^{131}\text{I}_2$ containing 10 and 100 mg of stable I_2 were released and contacted with the samples mentioned above for 60 minutes. After that the leaves were washed with running water and patted softly with paper towel for removing water.

2.4 Measurement of radioactivity and autoradiography

The radioactivity was measured by means of GM counter having 6 cm^2 of window area after covering the contaminated leaves with very thin membrane. The results were shown as ^{131}I (cpm/ 6 cm^2) and the distribution was checked autoradiographically. Fuji X-ray film ($27.0\text{ cm} \times 35.6\text{ cm}$), which was made by Fuji Film Co., Ltd., was used for the autoradiography.

2.5 Removal

Removal of $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ was examined by washing the contaminated leaves with detergent, CCl_4 , ether, 0.5N HCl and 0.5N NaOH .

Experiment was carried out in a darkened

hood in order to avoid the decomposition of the evaporated iodine compounds by light.

3. Results and Discussions

It was rather difficult to identify the chemical form of the evaporated I_2 when it was released by the oxidation of the carrier free Na^{131}I and therefore in this experiment adequate amount of stable sodium iodide carrier was added to the radioactive solution in order to sustain the existence of I_2 from its proper violet color and also to know the effect of the carrier. It was conjectured that thus evaporated I_2 vapor probably contained the other chemical form of inorganic iodine such as iodine oxide or iodine peroxide. For convenience, however, iodine vapor generated by the oxidation of sodium iodide was expressed as I_2 vapor in this paper.

3.1 Qualitative experiment

Deposition of $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ on spinach, camellia, filter paper and carbon filter paper was shown in Fig. 2 and Fig. 3 respectively.

Filter paper Spinach leaf Carbon filter paper Camellia leaf

Exposure time to film: 160 hours
Development time: 10 minutes

Fig. 2 Deposition of $\text{CH}_3^{131}\text{I}$ on spinach leaf, camellia leaf, filter paper and carbon filter paper.

Filter paper Spinach leaf Carbon filter paper Camellia leaf

Exposure time to film: 20 hours
Development time: 3 minutes

Fig. 3 Deposition of $^{131}\text{I}_2$ on spinach leaf, camellia leaf, filter paper and carbon filter paper.

$\text{CH}_3^{131}\text{I}$ was deposited on spinach and camellia leaves on the similar level, but detectable amount of radioactivity was not observed on filter paper and carbon filter paper. On the contrary, $^{131}\text{I}_2$ was strongly attached on spinach leaves and carbon filter paper, and also slight deposition was found on camellia leaves and filter paper. This suggested that it would be possible to distinguish evaporated I_2 from CH_3I indirectly by measuring the radioactivity deposited on the carbon filter paper. In spite of the slight deposition of $^{131}\text{I}_2$ on the surface of the camellia leaves, remarkable sorption was observed at the end of the stalk. When it was sealed by wax, as shown in Fig. 4, no deposition was observed. This suggested that

Unsealed Sealed

Fig. 4 Deposition of $^{131}\text{I}_2$ on the camellia leaves, the ends of the stalks of which were unsealed and sealed by wax.

the wax-like material on the surface of the camellia leaves might inhibit the sorption of $^{131}\text{I}_2$. It was recognized that both $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ were concentrated in the vein of the leaves.

3.2 Quantitative experiment

Concentration of $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ on one or three fixed areas on spinach and Chinese cabbage leaves were measured, as shown in Table 1 and 2, in order to know the level of the deposition of both vapors quantitatively.

Table 1 Average concentration of $\text{CH}_3^{131}\text{I}$ on a Chinese cabbage* and spinach leaves (cpm/6 cm² of the leaves)

Amount of $\text{CH}_3^{131}\text{I}$ evaporated (Amount of CH_3I carrier)	Chinese cabbage*			Spinach
	Upper part	Central part	Lower part	Central part
20 μCi (45 mg)	207	68	59	431
20 μCi (200 mg)	240	202	53	413

* Outside leaf

Table 2 Average concentration of $^{131}\text{I}_2$ on a Chinese cabbage* and spinach leaves (cpm/6 cm² of the leaves)

Amount of $^{131}\text{I}_2$ evaporated (Amount of I_2 carrier)	Chinese cabbage*			Spinach
	Upper part	Central part	Lower part	Central part
20 μCi (100 mg)	2892	831	904	1939
52 μCi (100 mg)	10387	4928	4883	19712
52 μCi (10 mg)	10792	6222	7156	4807

* Outside leaf

It was recognized that $^{131}\text{I}_2$ deposited more than ten times as much as $\text{CH}_3^{131}\text{I}$ for the same level of the evaporation (20 μCi). That was also clearly recognized from darker autoradiograph of the $^{131}\text{I}_2$ -deposited leaves shown in Fig. 6 though the film was shorter exposed to the contaminated leaves and also to a developing solution than that of the autoradiograph of the $\text{CH}_3^{131}\text{I}$ -deposited leaves shown in Fig. 5. The difference of the amount of the carrier had no effect on the deposition

A Chinese cabbage leaf Spinach leaves

Exposure time to film: 160 hours

Development time: 10 minutes

Fig. 5 Deposition of $\text{CH}_3^{131}\text{I}$ on a Chinese cabbage and spinach leaves.

(The outside leaf) (The second leaf from outside)

A Chinese cabbage Spinach leaves

Exposure time to film: 20 hours

Development time: 3 minutes

Fig. 6 Deposition of $^{131}\text{I}_2$ on a Chinese cabbage and spinach leaves.

level for both $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$, presumably caused by the physical disorder of the leaves damaged by the corrosive iodine vapor coming from comparatively much quantity of the carrier used for this experiment.

No activity was recognized from the leaves which were steeped in Na^{131}I solution.

Removal of $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ from the contaminated leaves was examined by washing them with detergent, CCl_4 , ether, 0.5 N HCl and 0.5 N NaOH. As shown in Table 3 and 4, effective removal was not clearly recognized. Ohno, et al.⁽⁶⁾ showed that radioactive iodine on the leaves contaminated by radioactive fallout arising from Chinese Nuclear Explosion Test was effectively removed by washing with

Table 3 Removal of $\text{CH}_3^{131}\text{I}$ from spinach and Komatsuna leaves (cpm/6 cm^2 of the leaves*)

Washing reagents	Spinach		Komatsuna	
	Before washing	After washing	Before washing	After washing
Detergent	225	181	459	432
0.5 N HCl	164	155	513	476
0.5 N NaOH	308	278	596	515
CCl_4	199	187	256	254
Ether	305	287	406	387

* Activity was measured on the same place before and after washing.

Table 4 Removal of $^{131}\text{I}_2$ from spinach and Komatsuna leaves (cpm/6 cm^2 of the leaves*)

Washing reagents	Spinach		Komatsuna	
	Before washing	After washing	Before washing	After washing
Detergent	28330	25224	4562	3786
0.5 N HCl	7470	7262	6836	5750
0.5 N NaOH	14206	14590	7606	7492
CCl_4	5858	5398	1788	1526
Ether	14224	14182	5992	5978

* Activity was measured on the same place before and after washing.

water. It was conjectured that radioactive iodine on the leaves contaminated by fallout

might exist as particles or be attached to the dust and soil on the leaves, and therefore it might be easily washed away with water, while in our experiment the evaporated iodine might combine strongly with leaves which were previously cleaned out by running water and it would be difficult to remove it by washing.

4. Conclusion

$\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ evaporated from generator were exposed to Chinese cabbage, spinach, Komatsuna and camellia leaves, and also filter paper and carbon filter paper. And the deposition and the distribution were investigated quantitatively and autoradiographically.

$\text{CH}_3^{131}\text{I}$ deposited on Chinese cabbage, spinach and camellia leaves on the same level, but significant radioactivity was not observed on filter paper and carbon filter paper. On the contrary, $^{131}\text{I}_2$ deposited strongly on spinach, Chinese cabbage and carbon filter paper and slightly deposited on camellia and filter paper. It was conjectured that wax-like material of the surface of camellia leaves might inhibit the sorption of $^{131}\text{I}_2$. It was suggested that $^{131}\text{I}_2$ deposited more strongly on leafy vegetables than $\text{CH}_3^{131}\text{I}$. Both $\text{CH}_3^{131}\text{I}$ and $^{131}\text{I}_2$ were concentrated in the vein of the leaves. No radioactivity was detected from the leaves which were steeped in the Na^{131}I solution.

Effective removal of iodine-131 from the contaminated leaves was not clearly recognized

by washing them with detergent, CCl_4 , ether, 0.5N HCl and 0.5N NaOH.

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要 旨

放射性ヨウ素の葉面付着に関する研究 (1)

大桃洋一郎, 佐伯誠道

放射線医学総合研究所, 東海研究室

放射性ヨウ素が, 植物の葉面にどのように付着するかを知るために, $\text{CH}_3^{131}\text{I}$, $^{131}\text{I}_2$ および Na^{131}I による付着状態を, ホウレン草, キャベツ, 小松菜および椿の葉について観察し, さらに水洗, 中性洗剤などによる洗浄によって, どの程度除染しうるかについて検討した。 $^{131}\text{I}_2$ は $\text{CH}_3^{131}\text{I}$ よりもより多く付着するが, Na^{131}I 水溶液の葉面からのとり込みは認められなかった。この事実は, ヨウ素のとり込みが気孔を通して行なわれ単なる物理的付着だけではないことを示唆しているものと考えられる。葉面からとり込まれたヨウ素は葉脈に濃縮されることが明らかにされた。水, 中性洗剤, 酸, アルカリ, エーテルによる除染効果は, 顕著には認められなかった。