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Bromine Uptake of Some Vegetable Crops Following Soil Fumigation with Methyl Bromide

Masao MASUI, Akira NUKAYA and Akira ISHIDA
College of Agriculture, Shizuoka University, Ohya, Shizuoka 422

Summary

Tomatoes, peppers, eggplants and strawberries were grown in soils fumigated with methyl bromide (MBr) to determine the effect of MBr on the growth and yield, bromine (Br) content in the plants and the content of water soluble Br in the soils. The growth and yield of 4 tesded crops were not affected by MBr fumigation. Br content in the plant parts of 4 crops tended to increase with increasing amounts of MBr. On a dry matter basis Br content in the fruit of 3 crops at 0, 100, 200 and 400 g/m^3 MBr treatments was as follows: in tomatoes 99, 113, 129 and 186 ppm; in peppers 77, 104, 181 and 282 ppm; in eggplants 72, 124, 213 and 364 ppm, respectively. Br content in the fruit of strawberries at 0, 200, 400 and 800 g/m^3 MBr treatments was 154, 179, 210 and 285 ppm, respectively. Water soluble Br in soil just after fumigation was significantly increased with increasing amounts of MBr. It was markedly decreased one and a half months after MBr fumigation at 200, 400 and 800 g/m^3, and thereafter gradually decreased with time at 400 and 800 g/m^3.

Introduction

Inorganic bromide (Br) is produced from the breakdown of methyl bromide (MBr) in soil, and can be taken up and accumulated in many crops (3, 4, 6, 10). This Br has recently been recognized as one of the substances of environmental pollution (10, 15, 16). As to the intake of Br ion, a joint FAO/WHO working party recommended in 1969 a maximum acceptable daily intake of 1 mg Br ion per kg body weight, with a tolerance level of 50 mg per kg in raw cereals or whole meal flour (2). Aside from this recommendation the Food and Drug Administration (FDA) of the Department of Health, Education and Welfare, and the Environment Protection Agency in the U.S.A. established a tolerance level of Br ion in many crops in 1972 (12, 13, 14). In our country little is known about the Br residues in vegetable crops grown in soil fumigated with Br containing compounds. In a previous paper authors (9) reported the growth and uptake of inorganic Br in muskmelon and cucumber plants grown in soil fumigated with MBr. This paper deals with the results of Br uptake of tomato, pepper, eggplant and strawberry plants.

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Materials and Methods

1. *Tomatoes.*

Some greenhouse growers sterilize soil beds with MBr using 0.5 to 2 times the amount recommended by the fumigant dealers. Therefore, 4 dosage levels of MBr were made as follows: 0 g/m^3 (not fumigated, 0 S for short), 100 g/m^3 (the half of standard, 0.5 S), 200 g/m^3 (standard, 1 S) and 400 g/m^3 (2 S). The corresponding amount of MBr of each dosage was released from a canister into the soil under a vinylfilm cover continuously for 4 days. The soil used was Iwata loam and originally had a pH of 5.8 and 2.2 ppm of water soluble Br. After removing the cover, fumigated soils were well tilled to evaporate the residual fumigant. Sixteen liters of fumigated solis were fertilized with 8 g N from $(NH_4)_2SO_4$, 10 g P_2O_5 from calcium superphosphate, 10 g K_2O from K_2SO_4, 30 g CaO from $Ca(OH)_2$, 2 g MgO from $MgSO_4 \cdot 7H_2O$ and 8 liters of well decomposed rice straw, and put into a 40×40×20 cm box. One 3 leafed seedling tomato cv. 'Kyoryoku-beiju' was planted in each box filled with fumigated soil on March 18, 1975, and grown under uniform conditions in the greenhouse. The treatments were replicated 7 times. The main stem was pinched at the 3rd node above the 5th cluster. Four fruits were set per cluster, and picked May 27 through June 26. The fruits in each cluster were dried at 70°C, and the leaves and roots were separated and dried. The dried fruit, leaves and roots were finely milled. Br content in these plant parts was determined by the same method as described in a previous paper (9). Soils were sampled just after MBr fumigation and at the end of the experiment, and water soluble Br was determined by the same method as described in a previous paper (9).

2. *Peppers.*

To determine the effects of soil fumigation with MBr on the growth and Br uptake of peppers, the treatment of 4 dosage levels of MBr, 0 g/m^3 (not fumigated, 0 S), 100 g/m^3 (0.5 S), 200 g/m^3 (standard, 1 S) and 400 g/m^3 (2 S) was made in 7 replications. The soil used, the fumigation method and fertilization amounts were the same as described in the tomato experiment. One pepper seedling with 8 leaves of cv. 'Suigyoku No. 2' was planted in each box filled with fumigated soil on April 3, 1975, and grown in the vinylhouse. Fruit sized 25 to 30 g was picked May 7 through June 18. The fruit was sorted into early (May 7 through May 21), mid (May 22 through June 4) and late (June 5 through June 18) pickings, further sectioned into flesh (socalled fruit) and seeds, dried and milled. The leaves, stems and roots were separated, dried and milled. Br content in the plants and soils was determined by the same method as described in the tomato experiment.

3. *Eggplants.*

To determine the effects of soil fumigation with MBr on the growth and Br uptake of eggplants, the treatment of 4 dosage levels of MBr (0, 0.5, 1 and 2 S) was made in 7 replications. The soil used, the fumigation method and fertilization amounts were the same as described in the tomato and pepper experiments. One seedling with 5 leaves of eggplant cv. 'Senryo' was planted in each box filled with fumigated soil on April 3, 1975, and grown in the vinylhouse. Fruit sized 60 to 80 g was picked May 9 through June 23. The fruit was sorted into early (May 9 through May 31) and late (June 1 through June 23) pickings, further sectioned into calyxes and the remaining part of fruit (so-called fruit), dried and milled. The leaves, stems and roots were also separated, dried and milled. Br content was determined on these plant parts and soils.

4. *Strawberries.*

To determine the effects of soil fumigation with MBr on the growth and Br uptake of strawberries, the treatment of 4 dosage levels of MBr, 0 g/m^3 (not fumigated, 0 S), 200 g/m^3 (standard, 1 S), 400 g/m^3 (2 S) and 800 g/m^3 (4 S) was made in 7 replications. The soil used was Takamatsu light clay with an original pH 5.7 and 6.0 ppm water soluble Br, and was fumigated with MBr by the method as described before. Twelve liters of fumigated soils were fertilized with 3.5 g N from $(NH_4)_2SO_4$, 5 g P_2O_5 from calcium superphosphate, 4.5 g K_2O from K_2SO_4 and 15 g CaO from $Ca(OH)_2$ and 6 liters of well decomposed rice straw, and put into a 40×40×12 cm box. Four nursery plants of strawberries cv. 'Hotta-Wonder' were planted in the box filled with fumigated soil on Oct. 1, 1975, and grown in the greenhouse at a minimum temperature of 12°C. Flowers were artificially pollinated with a hair pencil to assure the development of normally shaped fruit. Fruit was picked Nov. 27 through March 5. The fruit was sorted into early (Nov. 27 through Dec. 31), mid (Jan. 1 through Jan. 31) and late (Feb. 1 through March 5) pickings, further sectioned into calyxes and the remaining part of fruit (so-called fruit), dried and milled. The leaf blades, petioles and roots were also separated, dried and milled. To determine the changes of water soluble Br during the experiment, soils were sampled on Oct. 1 (just after fumigation), Nov. 15, Dec. 30 and March 5 (at the end of the experiment).

Results

1. *Tomatoes.*

The effects of soil fumigation with MBr on the growth and Br content in plant parts of tomatoes, and on the water soluble Br in the soil are shown in Table 1. The fresh weight of fruit per plant was not influenced by MBr levels. Light chlorosis caused by MBr treatment appeared at 1 and 2 S after picking of first cluster though the growth as expressed by the dry weights of leaves, stems and roots was not affected. Br content in the leaves was higher at 2 S, and that in the fruit of each cluster tended to increase with increasing amounts of MBr. Water soluble Br in the soil just after fumigation was significantly increased with increasing amounts of MBr. The difference in Br content between 0 and 2 S was less at the end of the experiment.

2. *Peppers.*

The effects of soil fumigation with MBr on the growth and Br content in plant parts of peppers, and on the water soluble Br in the soil are shown in Table 2. The fresh weight of fruit per plant was not influenced by MBr levels. No chlorosis or growth suppression was found even at 2 S. Br content in the leaves and fruit at each picking time tended to increase with increasing amounts of MBr, and that in the fruit tended to increase with time at all treatments. Br content in the seeds was higher at 2 S. Water soluble Br in the soil at 2 S was considerably decreased at the end of the experiment.

3. *Eggplants.*

The effects of soil fumigation with MBr on

Table 1. Effect of methyl bromide soil fumigation on growth and Br content of tomatoes and on Br soil content.

Treatment	FruitW fresh wt/ plant (g)	Br in plant dry matter and in air dried soil (ppm)									
		Leaves	Roots	Fruit in each flower cluster						Soil just after fumigation	Soil at the end of expt.
				1 st	2 nd	3 rd	4 th	5 th	Mean		
0 S	3001^{a}	72^{b}	58^{b}	94^{c}	126^{b}	110^{b}	79^{c}	87^{c}	99	2.5^{c}	1.3^{b}
0.5 S	3137^{a}	64^{b}	59^{b}	97^{c}	115^{b}	139^{b}	116^{b}	99^{c}	113	3.9^{bc}	1.6^{ab}
1 S	3034^{a}	67^{b}	80^{ab}	133^{b}	133^{b}	136^{b}	118^{b}	127^{b}	129	5.5^{b}	1.6^{ab}
2 S	3269^{a}	181^{a}	124^{a}	183^{a}	196^{a}	191^{b}	162^{a}	199^{a}	186	11.6^{a}	1.9^{a}
Mean				127	143	144	119	128			

W : Mean separation in columns by Duncan's multiple range test, 5% level.

Table 2. Effect of methyl bromide soil fumigation on growth and Br content of peppers and on Br soil content.

Treatment	FruitW fresh wt/ plant (g)	Br in plant dry matter and in air dried soil (ppm)										
		Leaves	Roots	Fruit				SeedsX			Soil just after fumigation	Soil at the end of expt.
				Early picking	Mid picking	Late picking	Mean	Early picking	Mid picking	Late picking		
0 S	1685^{a}	61^{c}	51^{b}	44^{c}	52^{c}	134^{b}	77	48	34	41	2.5^{c}	2.1^{c}
0.5 S	1761^{a}	101^{c}	65^{a}	47^{c}	72^{c}	192^{b}	104	Y	Y	Y	3.9^{bc}	3.6^{b}
1 S	1706^{a}	371^{b}	65^{a}	98^{b}	134^{b}	312^{a}	181	Y	Y	Y	5.5^{b}	4.0^{b}
2 S	1621^{a}	1017^{a}	66^{a}	214^{a}	248^{a}	383^{a}	282	160	168	228	11.6^{a}	5.6^{a}
Mean				101	127	255						

W : Mean separation in columus by Duncan's multiple range test, 5% level.
X : Not subjected to statistical analysis because composite samples were taken.
Y : Samples were not taken.

Table 3. Effect of methyl bromide soil fumigation on growth and Br content of eggplants and on Br soil content.

Treatment	No. ofW fruit/plant	Fruit fresh/wt plant (g)	Br in plant dry matter and in air dried soil (ppm)								
			Leaves	Roots	Fruit			Calyxes		Soil just after fumigation	Soil at the end of expt.
					Early picking	Late picking	Mean	Early picking	Late picking		
0 S	21.6^{a}	1683^{a}	61^{b}	6^{b}	50^{d}	93^{c}	72	109^{d}	98^{d}	2.5^{c}	2.8^{b}
0.5 S	23.1^{a}	1836^{a}	173^{b}	16^{ab}	122^{c}	126^{c}	124	291^{c}	298^{c}	3.9^{c}	3.1^{b}
1 S	22.0^{a}	1752^{a}	218^{b}	24^{ab}	229^{b}	197^{b}	213	519^{b}	563^{b}	5.5^{b}	3.9^{a}
2 S	22.7^{a}	1798^{a}	919^{a}	51^{a}	368^{a}	360^{a}	364	1117^{a}	1138^{a}	11.6^{a}	4.3^{a}
Mean					192	194					

W : Mean separation in columns by Duncan's multiple range test, 5% level.

Table 4. Effect of methyl bromide soil fumigation on growth and Br content of strawberries.

Treatment	Fruit[W] fresh wt/plant (g)	Br in plant dry matter (ppm)									
		Leaf blades	Petioles	Roots	Fruit				Calyxes[Y]		
					Early picking	Mid picking	Late picking	Mean	Early picking	Mid picking	Late picking
0 S	50.8[a]	116[c]	202[b]	322[c]	163[b]	140[b]	159[b]	154	133	141	148
1 S	48.4[a]	379[b]	204[b]	400[bc]	177[b]	164[b]	195[ab]	179	250	256	257
2 S	59.9[a]	553[b]	534[b]	614[b]	255[ab]	167[b]	207[ab]	210	285	257	230
4 S	55.1[a]	1341[a]	1876[a]	1408[a]	328[a]	299[a]	228[a]	285	650	531	460
Mean					231	193	197				

W : Mean separation in columns by Duncan's multiple range test, 5% level.
Y : Not subjected to statistical analysis because composite samples were taken.

Table 5. Changes of soil Br content during the experiment (ppm of dried soil).

Treatment	Oct. 1 (X)[W]	Nov. 15	Dec. 30	Mar. 5 (Y)	Mean
0 S	5.5[c]	5.3[c]	6.2[b]	5.2[b]	5.6
1 S	18.5[c]	7.8[c]	7.6[b]	9.2[b]	10.8
2 S	72.6[b]	16.6[b]	14.4[b]	10.8[b]	28.6
4 S	165.4[a]	52.8[a]	29.0[a]	28.3[a]	68.9
Mean	65.5	20.6	14.3	13.4	

W : Mean separation in columns by Duncan's multiple range test, 5% level.
X : Just after fumigation.
Y : The end of the experiment.

the growth and Br content in plant parts of eggplants, and on the water soluble Br in the soil are shown in Table 3. The number and fresh weight of fruit per plant were not affected by MBr levels. No chlorosis or growth suppression was found even at 2 S. Br content in the leaves and roots, and in the fruit and calyxes at each picking time tended to increase with increasing amounts of MBr. Generally Br was most concentrated in the calyxes, less concentrated in the leaves or fruit, and least concentrated in the roots. Water soluble Br in the soil at 2 S was considerably decreased at the end of the experiment.

4. *Strawberries.*

The effects of soil fumigation with MBr on the growth and Br content in plant parts of strawberries, and on the water soluble Br in the soil are shown in Table 4 and 5. The fresh weight of fruit per plant was not affected by MBr levels. No chlorosis and growth suppression was found even at 4 S. Br content in the plant parts tended to increase with increasing amounts of MBr. Generally Br was least in the fruit. Water soluble Br in the soil just after MBr fumigation was significantly increased with increasing amounts of MBr. It was markedly decreased one and a half months after MBr fumigation at 1, 2 and 4 S, and thereafter gradually decreased with time at 2 and 4 S.

Discussion

In general, favorable effects of MBr as a soil sterilant on the growth of crops can be attributed to the alleviation of injuries caused by soil-borne diseases and nematodes in continuous cropping. In the present experiments with tomatoes, peppers, eggplants and strawberries the growth and yield were not influenced by MBr fumigation. One of the reasons appears to be attributable to the absence of soil-borne diseases and nematodes due to the use of a newly incorporated paddy soil.

In spite of the evaporating of residual MBr in soils prior to planting, light chlorosis on the tomato leaves caused by MBr treatment appeared at 1 and 2 S after picking of first cluster though the growth and yield were not affected. In another experiment authors (8) found the germination of tomato and spinach seeds very sensitive to MBr. Thus tomatoes may be more sensitive to MBr than peppers, eggplants and strawberries.

Inorganic Br which is present as a breakdown product of MBr can be taken up and distributed in the tissues of the plants (3, 4, 7, 9, 11). Generally, Br content in the plant parts of the 4 crops increased with increasing amounts of MBr. However, the distributed pattern of Br taken up in the tissues varied somewhat within the 4 crops. In tomatoes Br content was considerably lower in the fruit, leaves and roots, and differences in Br content were not found among these tissues. In peppers and eggplants Br was most concentrated in the leaves and calyxes, least concentrated in the fruit, and least concentrated in the roots. In strawberries Br was most concentrated in the leaf blades, petioles and roots, less concentrated in the calyxes, and least concentrated in the fruit. The low content of Br found in the fruit of the 4 crops is similar to the muskmelon, cucumber and tomato results obtained by the authors (9) and Kempton et al (4).

The composition of soil plays a role in the penetration of MBr. Chisholm and Koblitsky (1) reported that the degree of sorption of MBr by the soil decreased in the sequence peat>clay>sand. In the experiments with tomatoes, peppers and egglants, the water soluble Br just after fumigation was 2.5, 3.9, 5.5 and 11.6 ppm at 0, 0.5, 1 and 2 S, respectively, while in the experiment with strawberries it was 5.5, 18.5, 72.6 and 165.4 ppm at 0, 1, 2 and 4 S, respectively. This was probably due to a potential difference in the retention of MBr between the two soils used in the experiments, as Iwata light clay in the experiments with tomatoes, peppers and eggplants had only 13.3% clay, while Takamatsu light clay in the experiment with strawberries had 35.8% clay.

Water soluble Br in the soil just after fumigation was significantly increased with increasing amounts of MBr as shown in Table 5. It was markedly decreased one and a half months after fumigation at 1, 2 and 4 S, and thereafter gradually decreased with time at 2 and 4 S. These reduction in the Br levels may be caused by (i) daily irrigation with water, (ii) taking up inorganic Br formed by the breakdown of MBr and (iii) evaporation of the residual MBr during the growing period of strawberries. However, it was difficult to determine the amounts of Br lost by the causes mentioned above.

The uptake extent of Br into plant tissue is closely related to the amount of inorganic Br in the soils (3, 4, 9). As shown in Table 1 to 5, Br content in the tomato, pepper, eggplant and strawberry plants reflected the water soluble Br content in the soils though the distributed pattern of Br in the tissues varied somewhat within the crops. Br content in the late picked pepper fruit was higher than that in the early picked one as shown in Table 2. This may be related to the greater root system at the late stage of pepper growth, and to the limited numbers and smaller size of the fruit. Similar results were reported on cucumber fruit by the authors (9).

Tomato, pepper, eggplant and strawberry, fruit on the plants grown in soils fumigated with 400 g/m^3 MBr, which is the highest dosage and one often being used by greenhouse growers, contained 186, 282, 364 and 210 ppm Br in dried tissue, respectively. These values are comparable to 9, 28, 22 and 27 ppm Br in fresh tissue, respectively. Although there are no statutory limits on the Br content of foodstuffs in Japan except for cereal grains, maximum levels for Br in a range of foodstuffs have been proposed by the FAO Codex Committee on Pesticide Residues (FAO/WHO), including suggested limits of 20 to 30 ppm for various fresh fruits and 30 to 250 ppm for dried fruits. Also, in the U. S. A. tolerances for Br residues in plants grown in soil treated with various nematocides have been established by the FDA (12). These values are 50, 30 and 50 ppm for tomatoes, peppers and eggplants, respectively, on a fresh weight basis (strawberries not listed). In a comparison of Br content in the tomato, pepper and eggplant fruits obtained by the authors with those by the FDA, Br in these fruits were within the tolerance limit in the soil fumigated with 400 g/m^3 MBr though Br in the pepper fruit was almost in the margin of the limit. When MBr is frequently used to sterilize soil beds, accumulated Br in a soil will produce fruit containing large quantities of Br which is beyond the tolerance limit. Thus more attention needs to be paid to obtaining the maximum effectiveness for eradicating soil-borne diseases with the minmum amount of MBr, and to taking some means for lowering Br in the fumigated soil before cultivation, such as leaching or evaporating the residual MBr as Maw and Malkomes (5, 10) pointed out.

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臭化メチルの土壌くん蒸消毒が数種野菜の臭素吸収に及ぼす影響

増井正夫・糠谷　明・石田　明

(静岡大学農学部)

摘　要

臭化メチル (MBr) の土壌くん蒸消毒が，トマト，ピーマン，ナス，イチゴの生育と収量，植物体と土壌の臭素 (Br) 含量に及ぼす影響を明らかにするため，これらの野菜を栽培した．供試4種の野菜の生育と収量は，MBr の消毒によって影響されなかった．4種の野菜の植物体各部の Br 含量は，MBr の使用量の増加とともに高まる傾向がみられた．果実の Br 含量を乾物当たりでみると，MBr 0, 100, 200, 400 g/m³ のトマトでは，それぞれ 99, 113, 129, 186 ppm であり，ピーマンではそれぞれ 77, 104, 181, 282 ppm であり，ナスではそれぞれ 72, 124, 213, 364 ppm であった．MBr 0, 200, 400, 800 g/m³ のイチゴの果実の Br 含量は，それぞれ 154, 179, 210, 285 ppm であった．MBr 消毒直後の土壌の Br 含量は，MBr の使用量の増加とともに高まったが，消毒1月半後には，200, 400, 800 g/m³ では著しく減少し，その後 400, 800 g/m³ では，徐々に減少した．