

ESTIMATION OF ATMOSPHERIC CONCENTRATIONS OF METHYL BROMIDE ALTERNATIVES AFTER A BAN ON METHYL BROMIDE

Y. Kobara*, S. Ishihara, A. Yokoyama and K. Ohtsu
Environmental Pesticide Assessment Unit,
National Institute for Agro-Environmental Sciences (NIAES),
3-1-3, Kannondai, Tsukuba, Ibaraki 305-8604, Japan

Methyl bromide (CH_3Br) was a major fumigant used in Japan to control soil-borne diseases in crops such as cucumbers, gingers, tomatoes, melons, green peppers, etc. The use of CH_3Br as a soil fumigant was phased out by 2005, but no new chemical or non-chemical alternative has yet been commonly used as its substitute. For now, chloropicrin, 1,3-dichloropropene and methyl isothiocyanate (MITC) and its generators (dazomet *etc.*) are seen as the best alternatives to CH_3Br . It is already difficult to satisfy demand for CH_3Br as a soil fumigant adequately, whereas there are not remarkable changes in the amount of 1,3-dichloropropene, chloropicrin and dazomet that have been used in major CH_3Br use areas (Fig. 1).

Our monitoring results of fumigants in the atmosphere during several months in horticultural areas showed that even under the current situation, fumigants such as 1,3-dichloropropene, chloropicrin, MITC and CH_3Br of high concentration (over several hundred $\mu\text{g}/\text{m}^3$) were detected frequently, but these high concentrations were temporary, and most air samples contained several $\mu\text{g}/\text{m}^3$ of fumigants. Restrictions on CH_3Br usage required an intensive search for improved technologies to reduce both dosage and emission of alternative chemicals from fumigated plots into the atmosphere, while maintaining its effectiveness for disease and weed control and providing adequate safety for people who live and work in areas where soil fumigations occur in multiple fields.

The purpose of this study is to estimate atmospheric concentrations of methyl bromide alternatives in the Kanto area under current soil fumigation methods in the near future. Sales tendencies of soil fumigants in major methyl bromide usage areas (prefecture) are very different by local conditions (Fig. 1). These sales predictions are very important to estimate atmospheric concentrations of alternative chemicals in the near future, were done from tendencies of past sales by the regression analysis (Table 2). Atmospheric concentrations of soil fumigants in the Kanto area were estimated by AIST-ADMER (National Institute of Advanced Industrial Science and Technology - Atmospheric Dispersion Model for Exposure and Risk Assessment). AIST-ADMER is a computer software program that enables estimation of average atmospheric concentrations of chemical substances by distributing chemical emissions in 5-km meshes, if the unit emission rate data or the information on chemical emissions at a source point is available.

In soil fumigation with chloropicrin, soil surfaces are covered with conventional polyethylene films (PE, 0.05 mm thickness), but in the case of 1,3-dichloropropene, are not usually covered in Japan. Emission losses of 1,3-dichloropropene and

chloropicrin were evaluated in field experiments from 11 June in 2001 on a volcanic ash soil (Hydric Hapludand) at the National Institute for Agro-Environmental Sciences, Tsukuba, Japan. "Manual injection method" was used for applying the "Dorochlor (80% chloropicrin, 20.2 g/m²)"- "D-D (92 % 1,3-dichloropropene, 17.2 g/m²)" mixture (in a volume ratio 1:1) into soil depth of ca. 17cm. Treated areas were 15 m² (2.5 m x 6 m), which were immediately covered with a conventional PE film (0.05mm thickness), then removed after 7 days (11 June). An automated gas chromatography system, equipped with flame ionization detectors (GC-FID) and four 7.5 L chambers (diam. 24.5 cm) was used to determine emission flux. The chambers were placed directly on the film or soil surface.

Our experiments showed that emission losses reached, respectively, about 42.7 % and 49.3 % of the applied amounts for chloropicrin and 1,3-dichloropropene with a PE film. However, Japanese growers generally don't use covering materials in the case of 1,3-dichloropropene, it is predicted that actual loss of 1,3-dichloropropene to the atmosphere is bigger than this estimate. The emission loss of 1,3-dichloropropene resulted 97 % of the applied amount with Fugacity Model (Level II).

We present average atmospheric concentrations of 1,3-dichloropropene and chloropicrin in 1995 and 2006 calculated based on experimental emission loss values with AIST-ADMER in Fig. 2. In the case of chloropicrin, the computed results indicate the air concentration levels in 2006 are approximately equal to levels in 1995. However, the results predict atmospheric concentrations of 1,3-dichloropropene in the west region of Kanto in 2006 will be lower than those in 1996, but in the east region of Kanto will be higher than those in 1995. It will not be allowed to increase 1,3-dichloropropene under conventional no covering method in these region.

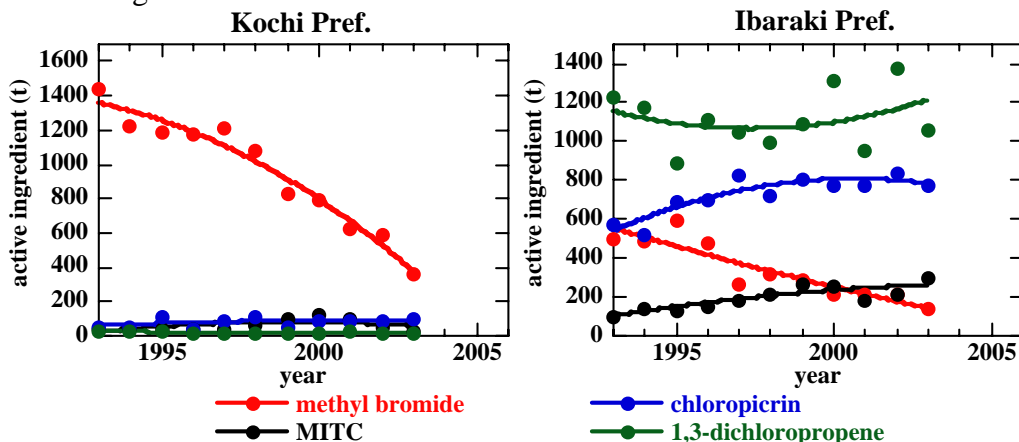
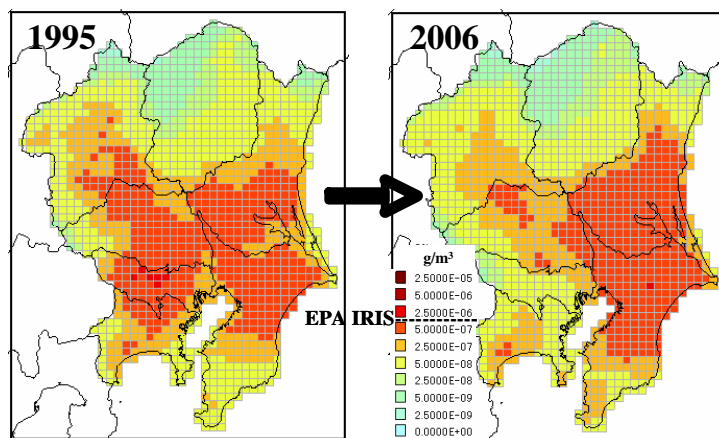


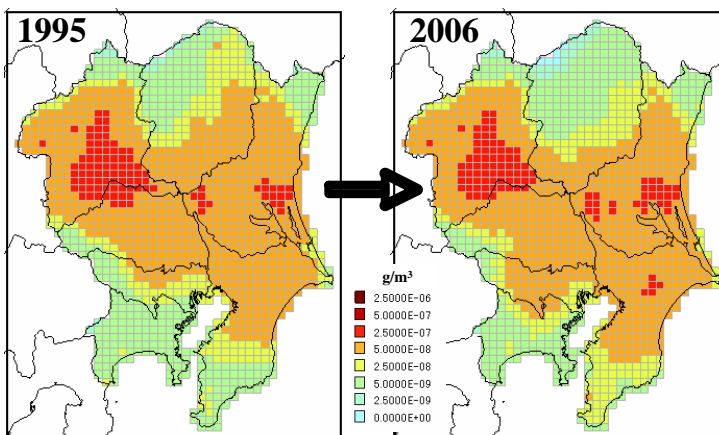
Fig. 1 Historical Soil Fumigant Sales in Major Methyl Bromide Use Areas (Active Ingredient, t)

Table 1 Sales of Soil Fumigants in 1996 and Prediction of Methyl Bromide Alternatives Sales in 2006 in the Kanto Area (Active Ingerdients, t)

Prefecture	1995				2006		
	methyl bromide	MITC	chloropicrin	1,3-dichloropropene	MITC	chloropicrin	1,3-dichloropropene
Fukushima	92	64	30	59	195	112	2
Ibaraki	591	130	686	891	260	750	1412
Tochigi	263	56	204	118	71	82	91
Gunma	367	90	1057	630	341	1054	453
Saitama	261	129	312	594	162	170	457
Chiba	489	76	380	1432	255	604	2336
Tokyo	109	10	21	1264	13	147	0
Kanagawa	129	27	24	273	30	24	310
Niigata	112	138	96	31	131	115	151
Yamanashi	118	8	11	7	11	11	5
Nagano	68	170	298	41	113	203	39
Shizuoka	279	80	297	128	141	216	349



1,3-dichloropropene



chloropicrin

Fig.2 Estimation of Atmospheric Concentrations of 1,3-dichloropropene and chloropicrin in the Kanto Area (Japan) by AIST-ADMER calculation (Daily Mean Concentration in 1995 and 2006)