Indoor and outdoor organophosphorus pesticides in an agricultural area in Japan

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ABSTRACT
Organoposphorus pesticides are used for agricultural or public purposes in Japan. Air pollution by its areal application is of concern. In this research, we conducted a pilot study of air pollution in agricultural residential areas by sprayed pesticide. Indoor and outdoor air was sampled for 1 week in four households and a public office using a chlomosorb-102 tube sampler. The concentration of these compounds was determined by solvent extraction and gas chromatographic analysis. In the samples obtained before spraying and after spraying, dichlorvos (the principal breakdown compound of trichlorfon) and fenitrothion were detected at <1.0–6.3 and <1.0–21.1 ng/m^3, respectively. The concentration of dichlorvos and fenitrothion in the indoor and outdoor samples in the public office were nearly non-detectable (0.5 ng/m^3). A correlation was found between outdoor and indoor dichlorvos and fenitrothion concentrations, suggesting the contribution of areal pesticide application to indoor air contamination.

INDEX TERMS
Indoor air; Organophosphorus pesticide; Trichlorfon; Fenitrothion; Agricultural area

INTRODUCTION

Organophosphorus (OP) pesticides are commonly used for pest control for agricultural or garden crops or plants in a public place. In some agricultural areas, pesticide is often sprayed on a broad area such as irrigated rice paddy fields, vegetable fields or orchards. The spray drift may become the source of air pollution. Baker *et al.* (1996) and John *et al.* (1995) found OP pesticides in ambient air near a spraying site in an agricultural community. In Japan, high volatility OP pesticides such as trichlorfon or fenitrothion are used for agricultural purposes. These pesticides have the possibility of causing high contamination of air and increasing the exposure risk. Moreover, recent housing construction is moving towards farmland. People living near the pesticide application site began to have great concern regarding the pollution of their residential environment. This is especially of concern for children. Children are said to be vulnerable to chemicals. Chemicals have the possibility of influencing their growth or development. Since children spend most of their time inside the house, the assessment of exposure in the residential environment is very important. Chensheng *et al.* (2000) and Nancy *et al.* (1995) conducted a survey of children's exposure to OP pesticides in an agricultural community, and they detected higher levels of pesticide in house dust in houses near pesticide treated orchards than those distant from the orchard. They also found the possibility of higher exposure to children living in those areas.

In Japan, few data are available about air pollution by agriculturally used pesticide. In this research, we conducted a pilot study to measure OP pesticide in air in an agricultural area.

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objective is to know the concentration of OP pesticide and the contribution of areal sprayed pesticide to indoor air contamination.

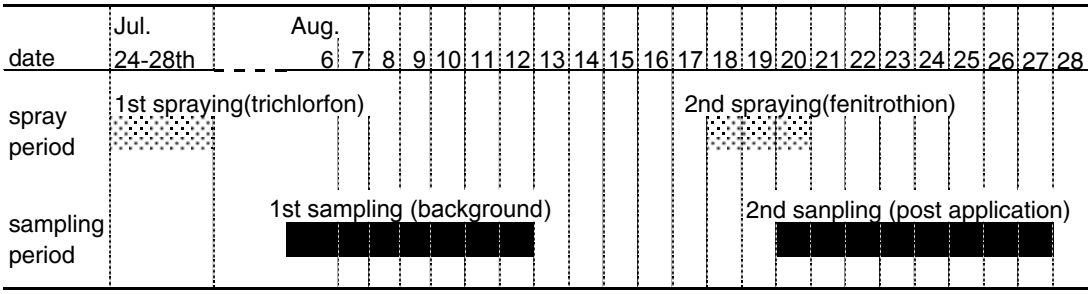
METHODS

Study Design

The study was conducted in some agricultural area in Japan where OP pesticides are used for pest control on irrigated paddy fields. In this area, water dissolved emulsion including trichlorfon (dimethyl-2,2,2-trichloro-1-hydroxyethyl phosphonate) was spayed on 24–28 July 2001 (first application) and fenitrothion (dimethyl-4-nitro-*m*-tolylphosphorothionate) on 18–20 August 2001 (second application) by a radio controlled helicopter. Trichlorfon is known to be hydrolysed to dichlorvos (2,2-dichlorovinyl dimethyl phosphate) in pH condition below 6.0 or a thermal condition and it has higher vapour pressure than the former. Four households were chosen and indoor and outdoor air was sampled for a week before and after the second application (6–13 August and 20–27 August 2001). A public office was also chosen and monitored because a public place is where people often visit and would be of great concern. All of the houses are wooden structures and the office building is a reinforced concrete structure and their locations are within 100 m from the rice field where pesticides were sprayed. A portable air pump (AirCheck2000, SKC Inc.) was set in the living room and around the entrance, and air was collected at approximately 0.5 m above the floor/ground at a flow rate of 2 l/min. A chromosorb-102 tube (8 mm i.d., length 110 mm, adsorbent 50/100 mg, SKC Inc.) was used to collect OP pesticides and the filter (PF100 Advantec) was attached to the inlet of the tube to remove particles in air. Duplicates were sampled outside of Office E. After sampling, samples were stored at 0°C in a cold box, and brought to the laboratory.

In House C air was sampled each 24hrs for 1week to know the decrease of concentration after application.

Table 1 Schedule of spraying period and sampling period



Analytical Method

Adsorbents were extracted with 2 ml acetone using an ultrasonic bath for 40 min and the sample extracts were analysed by gas chromatography with a frame photometric detector (GC-FPD Hewlett Packard GC6890). The chromatographic condition is as follows: column HP-1 Cross-linked methyl siloxane (0.32 mm i.d., 30 m length, 0.25 μm film thickness), injection volume 3 μl, inlet temperature 200°C, oven temperature programme 60°C (2 min hold) at 8°C/min to 250°C (3 min hold), temperature in detector 240°C. The extraction efficiency was 87.1%

Table 2 Extraction efficiency(%) and limit of quantitation of target compounds (ng/m^3)

Compound	Extraction efficiency±SD (%)	LOQ (ng/m3)	
		1week	24hour
Trichlorfon	87.7±2.1	1.0	7.0
Dichlorvos	91.6±11.5	1.0	7.0
Fenitrothion	89.6±3.5	1.0	7.0

for trichlorfon, 91.6% for dichlorvos and 89.6% for fenitrothion. The concentrations of target compounds were not adjusted by these extraction efficiencies. The limit of quantitation (LOQ) is 1.0 ng/m^3 for a 1-week sampling period and 7.0 ng/m^3 for a 24-h sampling period for each target compound.

For the determination of the extraction efficiency of trichlorfon, acetic anhydride was added to prevent the degradation of trchlorfon to dichlorvos in a thermal condition but not in sample analysis. So the figures described for trichlorfon are their sum.

RESULTS

OP pesticide concentration in outdoor and indoor air samples for a week in each house and the office is presented in Table 2. Dichlorvos and fenitrothion were detected in outdoor and indoor air samples at all of the sites. The range of concentration of fenitrothion in air sampled for a week after pesticide application was 2.1–14.6 ng/m^3 for outdoor air and ND to 6.3 ng/m^3 for indoor air. Dichlorvos was detected within the range 1.8–32.86 ng/m^3 for outdoor air and ND to 21.1 ng/m^3 for indoor air in this sampling period. In the public office, concentrations of the sprayed pesticides were nearly at the non-detectable level (0.5 ng/m^3).

As a result of the 24-h sampling in House C, the peak level of fenitrothion was 17 ng/m^3 in outdoor air and 9 ng/m^3 in indoor air on the first day of application, and the concentration in indoor air dropped to non-detectable level after 3 days with that of outdoor air (Figure 1).

Table 3 Concentration of organophosphorus pesticides in air sampled for a week in houses in agricultural area (ng/m^3)

Compound	Location	6th-13th Aug.		20th-26th Aug.	
		Indoor	Outdoor	Indoor	Outdoor
Fenitrothion	House A	1.0	-[a]	2.2	2.1
	House B	2.8	2.2	1.7	3.0
	House C	2.1	1.7	3.7	5.8
	House D	N.D.[b]	1.5	6.3	14.6
	Office E	N.D.	2.1	N.D.	5.0
Dichlorvos	House A	7.2	-	2.4	1.8
	House B	7.0	5.6	1.9	3.4
	House C	6.8	7.5	9.9	8.1
	House D	21.1	32.8	6.7	4.9
	Office E	1.8	2.4	N.D.	2.4

[a] sample lost in processing
[b] not detected

DISCUSSION

There was a positive correlation between the concentration of detected compounds in outdoor and indoor air samples (Spearman $r = 0.70$, $p < 0.03$ ($n = 9$) for fenitrothion and $r = 0.859$, $p < 0.002$ ($n = 9$) for dichlorvos). This indicates the contribution of outside pesticide application

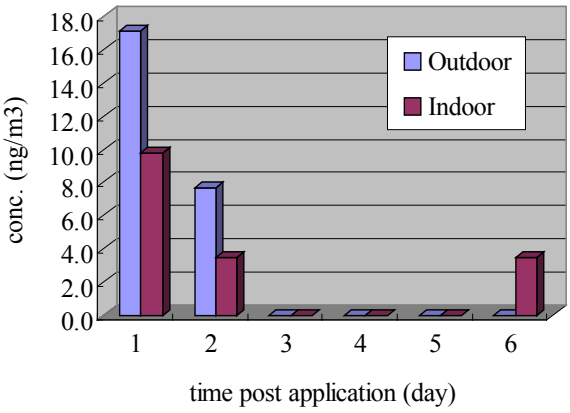


Fig.1 Result of 24 hour sampling in House C after areal pesticide application

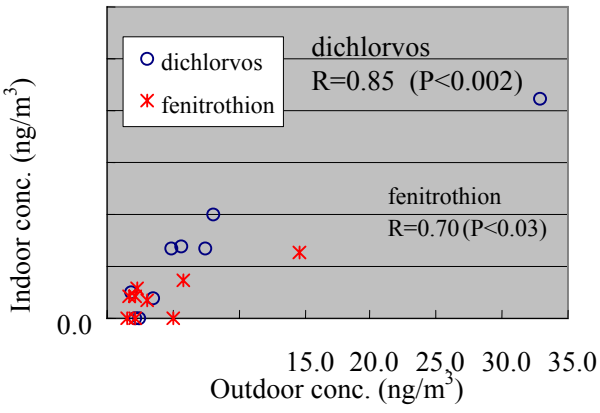


Fig.2 Scatter plot of outdoor and indoor OP pesticide concentration (ng/m^3)

application to indoor air pollution (Figure 2). In the period of this study, the windows were opened in most of the houses, this maybe the reason of the contamination of indoor air.

Dichlorvos was detected in all of the houses and the concentration is slightly higher than fenitrothion. This contamination may be caused by its higher usage or higher volatility besides the areal application. In this area, trichlorfon is often used for garden vegetable or tree. Since trichlorfon is known to change to dichlorvos under natural conditions (Murphy *et al*., 1996) the concentration of dichlorvos described here may include both of them. Contamination by this compound is of concern because it has higher toxicity than the parent (chronic inhalation NOAEL; 50 μg/m^3 for the former and 12.7 mg/m^3 for the latter). Investigation for understanding its production or duration in air is needed.

CONCLUSION

OP pesticides were detected in indoor air samples of houses in an agricultural area. The compound sprayed in the agricultural field was detected in indoor air and contamination lasted until 2 days after spraying. The indoor contamination was found to be related to the outdoor. Further study is expected to provide knowledge on indoor air contamination, distribution or frequency of exposure and it will help us to assess health risk among the population.

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