EXPOSURE OF GREENHOUSE WORKERS TO ENCAPSULATED METHYL PARATHION (PENNCAP-M) BEING USED ON CHRYSANTHEMUMS IN SANTA CLARA AND SAN MATEO COUNTIES IN 1981

By

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SUMMARY

Potential dermal and inhalation exposure of greenhouse workers to encapsulated methyl parathion (Penncap-M) was measured in Santa Clara and San Mateo Counties in 1981. Air, soil, foliar dislodgeable and worker handwash samples were collected to determine potential exposure, and were correlated with blood cholinesterase levels from workers reentering greenhouses after application. Foliar dislodgeable residues ranged from 1.45 to 2.51 ug/cm^2 1 day after application, and degraded to nearly non-detectable levels 6 to 13 days after application. Workers removing buds from plants 5 to 6 days following application accumulated 31.8 to 184.0 ug on their hands during a 3-1/2 hour exposure period. Ambient air levels in greenhouses after application did not exceed 1 part per billion (ppb). Workers reentering greenhouses 24 hours after application showed no clinically significant depression of whole blood, plasma, or red blood cell cholinesterase levels. Data from this study was used to develop a low-hazard set of instructions that included: (1) a 5-day worker reentry interval; (2) a requirement that applicators must wear clean coveralls daily, rubber boots, rubber gloves, and a rubber or plastic apron, and that they must shower at the end of the work shift; (3) a requirement that if workers will have substantial body contact with the foliage (watering, fertilizing) before the end of the reentry interval, they must wear the same protective clothing as the applicator; (4) a prohibition of activities such as disbudding and flower picking during the safety interval.

INTRODUCTION

Penncap-M is a toxicity category 2 organophosphate pesticide registered for use on a wide variety of field and orchard crops. It is a microencapsulated, controlled release formulation of methyl parathion designed to prolong the effective lifespan of the product, with the potential for reducing exposure hazards to applicators and field workers. In 1981, the California Department of Food and Agriculture granted a Section 24 registration for the use of Penncap-M on greenhouse-grown chrysanthemums for the control of Liriomyza leafminer larvae. It was then determined that before permits were to be issued to all qualified applicants, the use would be studied in two greenhouses to determine if wording on the 24C instructions on safe use should be modified.

Two greenhouse operations were monitored with Penncap-M being applied at 2 pounds a.i./acre at each site. Air, leafpunch, and soil samples were collected at predetermined intervals after application, blood samples were collected as the opportunities presented themselves, and handwash samples were collected during disbudding operations in which workers would be expected to have maximal contact with treated foliage.

Some of the exposure data cannot be correlated with biological indicators due to a lack of cooperation on the part of the workers in submitting to repeated venipunctures for blood sampling.

APPLICATIONS

Penncap-M was applied once a week for 3 weeks to achieve control of the larval stage of the pest, which has a 19-day life cycle. The greenhouse operation in Santa Clara County represented the "worst case" with low ceilings (12 ft.) and no forced air ventilation. 10,000 sq. ft. were treated at a rate of 1 pint/50 gallons of water (Penncap-M is formulated as 2 pounds/gallon). One application was monitored for 6 days. The greenhouse operation in San Mateo County had high, peaked ceilings (25 ft.) and forced air ventilation fans operating at 16,000 cu. ft. per min. (cfm). 20,000 sq. ft. were treated at a rate of 1 pint/50 gallons water. Monitoring was conducted during 3 applications occurring on March 31, April 15, and April 22, 1981.

SAMPLING METHODS

Air samples were collected from different sites in the greenhouses and from the applicators' breathing zone. Sampling duration ranged from 1 to 6 hours. Sampling intervals are specified in Table 2. Methyl parathion was trapped on XAD-4 porous polymer resin adsorbent tubes (SKC #226-30-11-04) drawn with MSA Monitaire S pumps operating at 1 liter/minute. Between 60 to 360 liters of air were sampled at any 1 site. Pumps were calibrated before and after sampling with a Kurz 540S Flow calibrater.

Leaf-punch samples were collected along a diagonal of the field according to the method published by Gunther, et al., (1973). Two samples were collected at each interval and analyzed for dislodgeable residues. Selected samples were analyzed for penetrated residues.

Soil samples were collected along a diagonal from the top 1/2-inch of the soil. Due to the high moisture contents of the soil, dust samples were not collected. One sample was collected at each interval.

Handwash samples were collected during bud removal operations which occurred approximately 1 month before harvest. One pre-exposure and 1 post-exposure handwash were collected from each worker. Workers washed their hands with approximately 250 ml of distilled water delivered by a separatory funnel.

Blood samples were collected by venipuncture from inside the elbow and stored in 7 ml, heparinized Vacutainer tubes. Pre-application samples were taken and post-application samples were taken at intervals ranging from 24 hours to 8 days.

All samples were chilled with ice and shipped to CDFA's Chemistry Laboratory Services for analysis by gas-liquid chromatography.

RESULTS

All samples, except for blood, were analyzed for methyl parathion. Blood samples were analyzed for red cell, plasma, and whole blood cholinesterase activity.

Air samples indicated that ambient levels of methyl parathion did not exceed 1 ppb at any time after application. Methyl parathion levels in the operator's breathing zone (outside of the respirator) were very close to 1 ppb.

Soil residues of methyl parathion did not exceed 8 ppm at any time after application.

Foliar dislodgeable residues reached their highest levels 1 day after application--approximately 2.5 ug/cm^2 of leaf surface. These data indicate that the levels degraded to nearly nondetectable levels (minimum detectable level = 0.0025 ug/cm^2) between 6 to 13 days after application. Reapplications of Penncap-M do not appear to cause a build-up of residues on leaf surfaces.

Levels of residues penetrating inside the leaves reached a high of 60 ppm 4 days after application, and degraded to 6-14 ppm 13 days after application.

Whole blood, plasma, and red blood cell (RBC) cholinesterase values did not show any clinically significant depression in any exposed individuals.

Handwash data, collected from workers performing disbudding operations on the plants, indicate that workers could accumulate from 31.8 to 184 ug of methyl parathion on their hands in 3-1/2 hours of work.

Laboratory methodology is reported in Appendix I.

DISCUSSION

One objective of this study was to obtain foliar residue data and worker blood cholinesterase data to establish a reentry interval for this

particular use of encapsulated methyl parathion. California Department of Food and Agriculture regulations specify that any application of methyl parathion containing 2 pounds or greater a.i./acre has a reentry period of 14 days. The foliar dislodgable data gathered in this study indicate that a shorter reentry period may be acceptable and still insure worker protection for greenhouse use of encapsulated methyl parathion.

Correlations of foliar residues and blood cholinesterase levels exist for only 24 hours after exposure for all the blood donors involved. Twenty-four hours after application, highest foliar residue values were 2.46 and 2.51 ug/cm^2 (see Table 5). Two individuals who were monitored for changes in cholinesterase levels and entered a greenhouse after application (individuals 2 and 3; see Table 3) showed no whole blood, plasma, or red blood cell depression greater than 15 percent of baseline 24 hours after application. Further correlation between foliar dislodgeable residues and biological effects in workers (changes in blood cholinesterase) are unavailable due to a reluctance of workers in submitting to repeated venipunctures.

Clinical depression of blood cholinesterase did not occur in the individual performing the application (individual 1, Table 3). A second applicator was monitored, but 24-hour post-application blood data is unavailable due to improper collection of the sample by a local contract laboratory. Ambient levels of methyl parathion in the operator's breathing zone were 0.96 and 1.30 ppb during the 2 applications monitored. An application typically required 1 hour to complete. Handwash and dermal exposure data were not collected for applicators; however, applicators were wearing all protective clothing required for the application of category 1 pesticides: rain coat, rain pants, rubberized hat, rubber gloves, rubber boots, goggles, and approved half-faced respirator with organic vapor cartridges.

The workers who reentered the greenhouse for disbudding operations typically wore pants and long-sleeved shirts. They wore cotton gloves with the finger tips cut out, and had their fingers taped with masking tape. Tape and gloves were removed by the worker before his or her hands were washed. The data indicate that dermal exposure to methyl parathion can occur with substantial contact to the foliage. Highest levels of exposure are associated with 1 individual and could be related to that individual's work habits. Workers typically spend only a portion of their work week disbudding plants. Disbudding operations occur approximately 1 month prior to harvest.

Ambient levels of methyl parathion collected in the greenhouses were uniformly low (all below 1 ppb), and correlated well with levels from field applications reported in the literature (Jackson and Lewis, 1978, 1979). Encapsulated methyl parathion in the greenhouse was a little more persistent than in the field, possibly due to a lack of wind and direct sunlight in the greenhouse.

Methyl parathion levels in the soil agreed closely with values in the literature (All, et al, 1978).

Reapplications of Penncap-M on a weekly basis do not appear to cause an accumulation of methyl parathion residues on the foliage or in the air.

Reapplication appears to cause some slight accumulation of residue in the soil. This could be due to the low mobility of methyl parathion in the soil (All, et al, 1978).

There is very little information present in the literature on the mammalian toxicity of encapsulated methyl parathion. A study in which encapsulated methyl parathion was painted on newborn calves and 2- to 3-year old cattle reported that the minimum acute toxic doses were 4 liters of a 0.5 % concentration for calves (2000 mg/animal) and 10 liters of a 0.25 % concentration for mature cattle (2500 mg/animal), Palmer, J. S. (1978). Unpublished data submitted by the Pennwalt Corporation (manufacturer of Penncap-M) reports an acute oral LD_{50} in mice of >120 mg/kg a.i. and an acute dermal LD_{50} in rabbits of >1200 mg/kg a.i. Although the toxicity data for the different routes exposure was generated in different species, it could be construed that the rate of dermal transport of encapsulated methyl parathion is low.

The Pennwalt Corporation maintains the position that present foliar dislodgeable residue analysis methods (Gunther, et al, 1973) are not accurate because the method removes and extracts methyl parathion still bound in capsules that would presumably not be available for exposure to workers. Present monitoring techniques, therefore, would apparently magnify potential exposure; however, diffusion forces between skin and capsules are poorly defined. It is possible that worker exposure can be enhanced by capsules adhering to the skin (i.e., underneath fingernails) providing a continuous source of methyl parathion for continuous absorption. Also, the exposure monitoring techniques were designed to determine the "worst case" exposure possible. This controversy over the validity of current monitoring techniques for determining exposure of encapsulated pesticide formulations has not yet been satisfactorily resolved by studies correlating magnitude of biological effect (i.e., cholinesterase depression) with the different methods of analyzing exposure samples.

Samples were not analyzed for the oxon form due to (1) interferences present in the different sample materials, especially in dislodgable and penetrated foliage samples, and (2) interferences presented by methyl parathion due to incomplete gas chromatographic resolution and the greater detector sensitivity of the parent compound in relation to the oxon. Liquid chromatographic confirmation for oxon levels (the analytical method of choice) was unavailable at the time the study was conducted.

Information developed from this study was used to develop a low-hazard set of restrictions of using this product in greenhouses on chrysanthamums (see Appendix II for the revised 24C registration that was issued).

These restrictions included:

1. A 5-day worker reentry interval.

2. A requirement that applicators must wear clean coveralls daily, rubber boots, rubber gloves, and a rubber or plastic apron, and that they must shower at the end of the work shift.

3. A requirement that if workers will have substantial body contact with the foliage (watering, fertilizing) before the end of the reentry interval, they must wear the same protective clothing as the applicators.

4. A prohibition on activity such as disbudding and flower picking during the safety interval.

Table 1

Levels of methyl parathion found outside a respirator in the applicator's breathing zone during application of micro-encapsulated methyl parathion (trade name-Penncap M).

Application Site #	Exposure Time	Levels of Exposure (ppb)
1	80 min.[a]	0.96
2	40 min.	1.3

[a] Includes 20 minutes spent outside the greenhouse mixing a second load.

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Table 2

Levels of methyl parathion in the ambient air.

Application Site #	Time After Application[b]	Number of Samples Taken	Range (ppb) Low	High	Mean (ppb)
1	Pre-application	1	--	--	ND[c]
	immediate post-application	7	0.17	0.47	0.28
	6 hours. after	8	0.26	0.71	0.45
	24 hours	8	0.16	0.94	0.47
	30 hours	8	0.14	0.54	0.28
	2 days	4	0.02	0.38	0.19
	3 days	5	0.06	0.46	0.20
	4 days	5	0.15	0.65	0.36
	5 days	5	0.02	0.16	0.08
	6 days	5	0.04	0.16	0.09
2	24 hours	3	0.33	0.36	0.35
	2 days	3	0.08	0.24	0.16
	3 days	3	0.04	0.18	0.13
	4 days	3	0.11	0.12	0.11
	5 days	3	0.07	0.12	0.10
	6 days	3	0.06	0.10	0.08
	7 days	3	0.06	0.08	0.07
	13 days	1	--	--	0.06
Second application occurred 15 days after the first application, 5 days after the second application		1	--	--	0.02
Third application occurred 22 days after the first application, 5 days after the second application		1	--	--	0.31

[b] Except the pre-application samples taken before the application

[c] ND means none detected at the minmum detectable limit

Table 3

Blood cholinesterase values before and after the application of micro-encapsulated methyl parathion measure in units of micromoles of -SH/ml./minute.

Application Site #	Individual	Time After Application[d]	Whole Blood	Plasma	RBC
1	1	Baseline	17.7	9.3	27.9
		24 hours	17.2	10.2	25.4
		5 days	17.2	9.5	27.1
		8 days	18.5	10.2	28.2
	2	Baseline	18.8	9.95	31.3
		24 hours	17.7	10.7	27.8
		48 hours	18.7	10.6	29.5
	3	Baseline	16.6	8.75	26.6
		24 hours	15.5	9.2	22.8

(d) Except the baseline values which were taken prior to exposure

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Table 4

Soil Residues of methyl parathion following application of micro-encapsulated methyl parathion.

Application Site #	Time After Application	Results (PPM-Dry Weight basis)
1	Pre-application	ND
	1 day post	1.83
	3 days	0.48
		0.84
	4 days	1.29
		3.64
	5 days	0.78
		1.75
	6 days	0.98
		1.65
2	Pre-application	ND
	1 days post	7.85
	2 days post	5.65
		5.68
	4 days post	7.24
	6 days post	6.32
	13 days post	5.15
A second application occurred at 15 days.		
	5 days	5.47
A third application occurred at 22 days.		
	5 days	7.55
	13 days	2.78

Table 5

Levels of methyl parathion found as dislodgeable and penetrated residues in leaf samples.

Application Site #	Time After Application	Dislodgeable Residues (ug/cm^2)	Penetrated Residues (ppm)
1	Pre-application	ND[c]	--
	immediately after the application	1.59	--
	1 day	2.51	--
	2 days	1.45	--
		1.47	--
	3 days	0.71	--
		0.86	
	4 days	0.97	--
		0.41	--
	5 days	0.47	--
		0.68	--
	6 days	ND	--
		ND	--
2	Pre-application	0.24	--
	1 day	1.45	--
		2.46	--
	4 days	1.01	42.0
		0.80	60.1
	6 days	0.57	38.7
		0.61	37.1
	13 days	0.03	6.4
		0.03	5.5
Second application occurred 15 days after the first application			
	5 days after	0.55[e]	52.2[e]
	Second application	0.25[e]	166.2[e]
Third application occurred 22 days after the first application			
	5 days after	0.25	54.5
	third application	0.25	54.0
	13 days	0.04	13.7
		0.05	20.2

(e) These results may be inaccurate due to an error occurring during the analysis.

Table 6

Residues of methyl parathion washed off the hands of workers by water during debudding operations of mums where micro-encapsulated methyl parathion was applied.[f] Results are expressed in ug/sample.

Individual	Day 1 (5 Days After Application)		Day 2 (6 Days After Application)	
	Pre-Wash	Post-Wash	Pre-Wash	Post-Wash
1	ND[c]	38.2	1.54	33.3
2	1.86	88.6	0.92	52.7
3	ND	44.7	2.04	31.8
4	12.1	184.0	2.60	150.0
5	ND	37.6	ND	33.1

(f) The workers had done a small amount of debudding prior to the pre-exposure period handwash (both days). The exposure period was approximately 3-1/2 hours long.

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APPENDIX I

ANALYTICAL METHODS

Air Sample Preparation

1. Score each sample tube with a file in front of the first section of resin.

2. Break the tube open.

3. Remove and discard the wire and glass wool.

4. Transfer the front (larger) section of resin into a labeled desorption vial (SKC #226-09) and add 2 ml of ethyl acetate.

5. Discard the central foam plug and transfer the back portion of resin into a second labeled desorption vial and add 2 ml of ethyl acetate.

6. Place filled and labeled vials on a sample rotator and rotate for 1 hour at 30 rpm.

7. Determine the amount of methyl parathion present by gas chromatography.

Air Sample Desorption Efficiency

1. Remove the foam and back (small) portion of resin from an air sampling tube of the same lot number to be used for the air determinations.

2. Inject a known and reasonable quantity of methyl parathion standard (based upon levels expected in the field or the desired sensitivity) into the remaining section of resin in the tube with a microsyringe. Cap the tube, and store as the tube will be stored during sample shipment. The storage time should be the same as the time expected to elapse between collecting the sample and analyzing the sample.

3. Run at least 5 tubes (plus a blank tube handled like the others except injecting no methyl parathion) in this manner, and determine the mean by the analytical procedure.

4. Desorption efficiency = (area sample-area blank/area standard), where the standard is the same amount as injected into the tube.

Air Sample Calculations

1. Determine the weight of methyl parathion present on the tube sections in nanograms by gas chromatography.

2. Correct this total weight by subtracting any weight value present on the blank or control tube.

3. The corrected weight is divided by the desorption efficiency to obtain the final corrected weight of methyl parathion present.

4. The volume of air sampled is converted to standard conditions of 25°C and 760 mm Hg.

$$VS = V \times \frac{P \times 298}{760 \times (T + 273)}$$

Where VS = Volume of air at standard conditions.
V = Volume of air sampled.
P = Barometric pressure in mm Hg.
T = Temperature in degrees C.

5. Calculate ppb in air from the above data.

$$\text{ppb (v/v)} = \frac{ng \times 24.45}{VS \times 249.0} = \frac{ng}{VS} \times 0.09819$$

24.45 is the molar volume of methyl parathion at 25°C and 760mm Hg.

249.0 is the molecular weight of methyl parathion.

Dislodgeable Residue Preparation

1. Weigh the sample jar and punches.

2. Add 50ml of distilled water and approximately 4 drops of sur-ten solution (1:50) to the sample jars.

3. Cap the containers tightly with aluminum foil and a screw cap and rotate at 30 rpm for 20 minutes.

4. Decant the aqueous solution through a plug of glass wool into a 500ml separatory funnel.

5. Repeat steps 2-4 two times.

6. Extract the aqueous solution three times with 50ml chloroform.

7. Evaporate the chloroform extract to just dryness on a steam bath with flowing air.

8. Redissolve the residue in hexane, transfer quantitatively to a volumetric flask and bring to a final volume in hexane.

9. Analyze by gas chromatography. Report results as ug/cm^2 using the number of punches and punch diameter supplied by the inspector.

Penetrated Residues

1. After the last water rinse is drained for the dislodgeable residue, the punches are transferred to a blender jar. The empty sample jar is weighed and the net weight of the punches is recorded.

2. Add approximately 50 gms of anhydrous sodium sulfate and 100ml of ethyl acetate.

3. Blend the sample at high speed for three minutes, keeping the blender cup cool by immersing it in a container of cool water.

4. Filter the extract through a bed of glass wool. Make it up to a final volume with ethyl acetate and submit for GC analysis.

5. Calculate results as ppm using the sample weight determined in step 1.

Methyl Parathion In Water Handwashes

Reagents and Equipment:

1. Ethyl acetate, nanograde.
2. Sodium sulfate.
3. 500 ml graduated cylinder.
4. Assorted volumetric glassware and pipets as needed for samples and standards.
5. Hewlett Packard 5880 gas chromatograph with NPD detector

Analysis:

Measure and record the amount of solution. Place a 100 ml aliquot in a 250 ml separatory funnel, add 10 ml saturated NaCl solution, and extract with 50 ml ethyl acetate. Drain the water layer and place the ethyl acetate in a 100 ml glass-stoppered graduated cylinder. Reextract the water layer twice more with 20 ml ethyl acetate, combining the extracts in the graduated cylinder. Bring to volume, add sufficient sodium sulfate to dry the solvent, and analyze by GLC.

Calculations must reflect the fact that only an aliquot was analyzed. Results have been reported in micrograms/total sample, and the total amount of handwash solution should be noted.

Recoveries are in excess of 95%.

The Analysis of Methyl Parathion in Soil

Reagents and Equipment:

1. Methyl parathion, analytical grade standard

2. Ethyl acetate, nanograde

3. DuPont Sorval blender, or equivalent

4. Hewlett Packard 5880 Gas chromatograph equipped with nitrogen/phosphorus detector

Analysis:

Weigh 50 grams of the soil into a Sorval blender cup, add 100 ml ethyl acetate, about 50 g sodium sulfate, and blend two minutes. Pour off an aliquot of the solvent and analyze by NPD/GLC.

Calculation:

$$\text{ppm} = \frac{(\text{area of sample}) * (\text{ns std}) * \text{final volume in ml})}{(\text{area of std}) * (\text{microliters inj}) * (\text{grams sample})}$$

Results should be reported on a dry weight basis.
Peak heights may be used instead of areas.

Gas Chromatography of Methyl Parathion

Instrument: Hewlett Packard 5880 with NPD detector

Column: 6' x 2 mm glass column contains 10% SP-2100 coated on 100/120 mesh Chromosorb W-HP operations at 200 C and 35 ml/min. helium carrier gas.

Temperatures: Detector at 300 C
Injector at 220 C

Under these conditions, methyl parathion elutes in about 5.0 minutes.

STATE OF CALIFORNIA EDMUND G. BROWN JR., *Governor*

DEPARTMENT OF FOOD AND AGRICULTURE

1220 N Street
Sacramento 95814

May 5, 1981

REGISTRATION FOR SPECIAL LOCAL NEED
FOR DISTRIBUTION AND USE ONLY WITHIN CALIFORNIA

Trade Name: Penncap-M EPA Reg. No. 4581-292-ZA
Firm Name: Pennwalt Corporation

DIRECTIONS FOR USE
It is a violation of Federal law to use this product
in a manner inconsistent with its labeling.

Location: Chrysanthemum growing localities
Crop/Site/Commodity: Greenhouse-grown chrysanthemums
Target Pest/Problem: Lirimyza leafminer (larval stage)
Dosage: Apply 25 to 75 gallons of above spray solution per 10,000 square feet of greenhouse.
Dilution Rate: 1 pint of formulated product (or 0.25 lb. ai) in 100 gallons of water.
Method of Application: Ground equipment
Frequency/Timing of Application: Begin applications 7 days after transplanting is completed and continue on a 7-day schedule until flower buds show color.
Field Reentry Safety Interval: Five days
Preharvest Interval: Not applicable
Other Requirements: Applicators must wear clean coveralls daily, as well as rubber boots, rubber gloves, and a rubber or plastic apron, and they must wash under a shower at the end of the work shift.

Other workers in the greenhouse: No special protective requirements are needed for any worker after the expiration of the safety interval. If it is necessary for any worker to engage in any work activity that may involve substantial body contact with the plants during the safety interval, such as watering or fertilizing, that worker must be provided the same protection as the applicator, including clean coveralls daily and the availability of a shower at the end of the work shift. Activity such as disbudding and flower picking is prohibited during the safety interval.

Expiration Date: December 31, 1982

All applicable directions, restrictions, and precautions on the EPA-registered label are to be followed.

This labeling must be in the possession of the user at the time of pesticide application.

Restricted materials permit must be obtained from the county agricultural commissioner prior to this use. This does not constitute a recommendation of the Department of Food and Agriculture and will not prevent quarantine action if illegal residues are found on or in the crop. Neither the Department nor the county agricultural commissioner, the manufacturer or

formulator makes any warranty of merchantability, fitness of purpose, or otherwise, expressed or implied, concerning the use of a pesticide in accordance with these provisions. The user and/or grower acknowledges the preceding disclaimer and accepts liability for any possible damage resulting from this use. Do not use in mixture with other pesticides unless provided for in the labeling. Trial on a small area to check out unanticipated problems is suggested.

24(c) Registrant: San Mateo County Department of Agriculture
P.O. Box 999
Redwood City, California 94064

EPA SLN No. CA 810024

George A. Reese, Chief
Pesticide Registration and
Agricultural Productivity
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