# Respiratory Route of Pesticide Exposure as a Potential Health Hazard

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Since 1965 the Michigan Department of Public Health has been conducting a Community Pesticide Study in the fruit growing area of southwestern Michigan in order to ascertain the impact of chronic exposure to a variety of pesticides and agricultural chemicals on human health. This project is one of fourteen such studies under contract to the Division of Pesticide Community Studies of the Environmental Protection Agency.

Our observation of many handling and application methods and procedures on the farm and in the orchard led us to question the role of the respiratory route of exposure as a direct hazard to the health of those exposed. Additionally, we were interested in which work activity might represent the greatest potential hazard.

The role of the respiratory route as a source of pesticide exposure has been studied in the past by both direct and indirect methods but little has been done in comparing exposures by different activities (DURHAM and WOLFE 1962, WOLFE et al. 1966, JEGIER 1964, SIMPSON 1965, BATCHELOR and WALKER 1954, WOLFE et al. 1972). A previous study of applications of pesticides to orchards utilizing low volume high concentrate (8x-12x) air blast spray machines resulted in a calculated respiratory exposure for workers operating these machines approximately 2.7 times as great as for those operating conventional high volume (1x) air blast sprayers (WOLFE 1966). SIMPSON's (1965) highest respiratory exposure measurement of guthion in apple orchards was equivalent to only 0.108% of a fatal dose per day. Respiratory exposure to carbaryl under similar conditions indicated little hazard (SIMPSON 1965). Comparisons by WOLFE, et al. (1966) of high concentrate and dilute air blast spraying indicated a calculated total dermal exposure of 27.9 mg/hr and a respiratory exposure of 0.055 mg/hr for the 8x-12x concentrations, while dilute 1x gave results of 19.4 mg/hr and 0.02 mg/hr respectively. BATCHELOR and WALKER (1954) found the majority of occupational accidents investigated involving parathion were predominantly dermal exposure, and many occupational exposures involved both the respiratory and dermal routes in such a way that it was difficult to distinguish their relative importance.

When organophosphates are used dermal exposure appears to be a greater potential hazard, but observations indicate that most persons are covered by clothing, except for their faces, necks and hands, during spray operations. As a result, it is felt the respiratory route of exposure should not be overlooked as a real source of short time high-concentration exposure. In order to better evaluate the actual hazards of pesticide exposure via the respiratory route an experimental study was designed with the following objectives:



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(1) to document and evaluate respiratory exposure to pesticides during several use activities to determine which activity presented the greatest hazard; and (2) to evaluate air levels of pesticides; (3) to estimate efficiency of simple respirators in protecting against respiratory exposure.

## METHODS AND MATERIALS

In order to study routine exposure operations on the farm the cooperation of several persons involved in a long term study of health related effects of pesticides was solicited. These persons were engaged in full time fruit farming in southwestern Michigan and utilized air blast speed sprayers for pesticide application. The speed sprayers were drawn by tractors through the orchards (BATCHELOR and WALKER 1954).

Direct methods of entrapment utilized two separate collection devices to simultaneously sample air: (a) a modified Willson single cone G-2 dust respirator (see Illustration No. 1) as described by DURHAM and WOLFE (1962), worn by the persons being studied to determine pesticides in inspired ambient air and (b) a Gelman midget impinger containing ethylene glycol to collect ambient air containing pesticides in the general area of the nostrils. The exposure operations of mixing and spraying were timed to the nearest minute.

The respirator filter pads used in these experiments were made by stapling 32 layers of surgical gauze to the alpha-cellulose filter normally used with this type respirator. All respiratory pads were pre-extracted with 200 ml of 1:1 acetone-hexane solvent in a 250 ml Soxhlet extractor prior to use in field operations. Six pads were extracted again as reagent blanks. Extracts were concentrated to several milliliters in Kuderna-Danish concentrators and then made up to 50 ml with hexane. Reagent blanks determined by electron capture gas chromotography were less than detectable limits. In use, these pads were placed in the masks, so the layers of gauze faced away from the wearer.

Midget impingers, containing ethylene glycol, were connected by means of a rubber hose to a portable vacuum source. The air sampling device was constructed in the instrument section of the Michigan Department of Public Health, Bureau of Occupational Health. A diaphragm vacuum pump, six volt motor and midget impingers were obtained from the Mine Safety Appliance Company. These are standard items used in the Monitaire Sampler. Six 1.2 volt wet type nickel-cadmium rechargeable batteries were combined into a 7.2 volt power pack. The pump, motor, and power pack were housed in an aluminum case which could be slung from the shoulder on a leather strap. A needle valve was incorporated to adjust airflow, and a vacuum gauge capable of measuring 0-30 inches of water was attached to allow for adjusting airflow at a standard rate. Calibration of the impingers was performed in the Michigan Department of Public Health, Bureau of Occupational Health, and no differences in airflow were found between impingers.

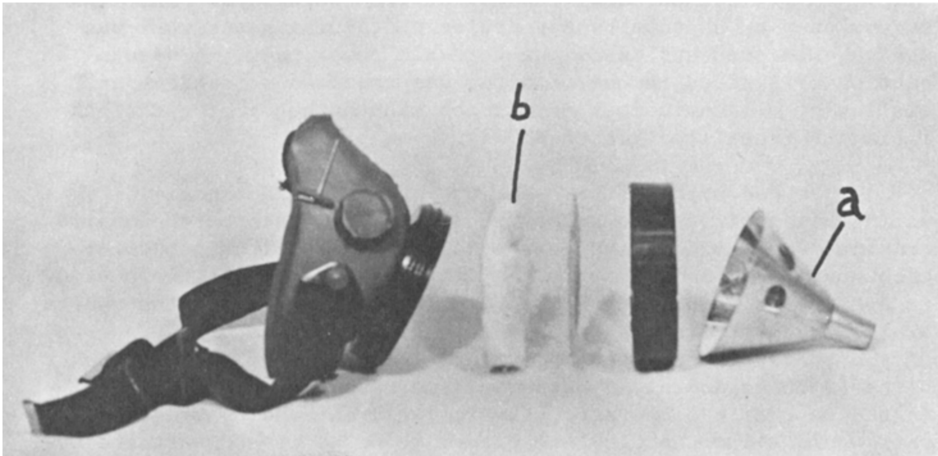


ILLUSTRATION NO. 1
WILLSON G-2 DUST RESPIRATOR
a. Special nose cone b. Special respirator filter pad

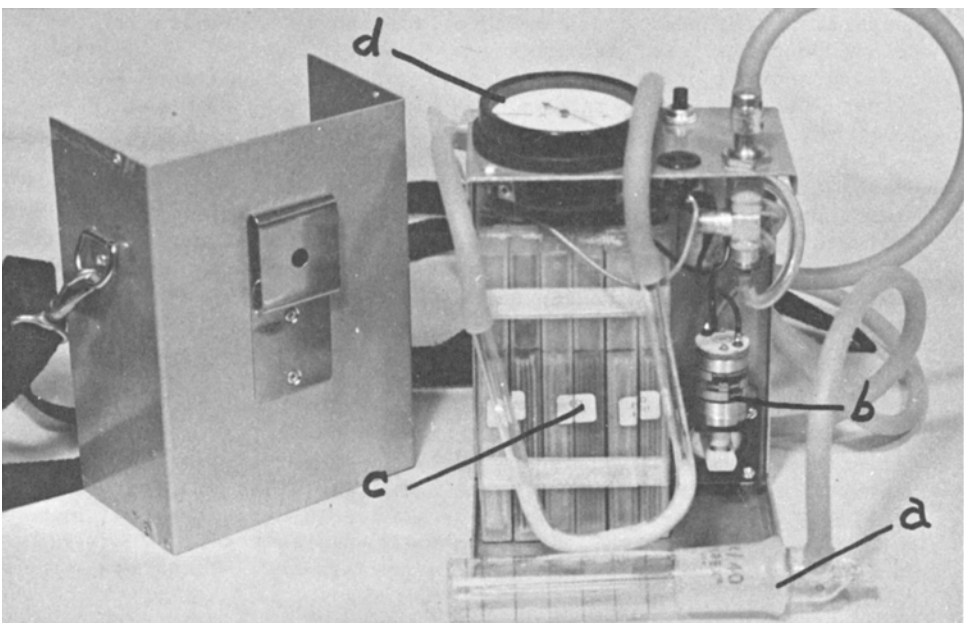


ILLUSTRATION NO. 2
MIDGET IMPINGER, PUMP AND POWER PACK
a. Midget impinger b. Six volt motor and diaphragm pump
c. Power-pack of six 1.2 volt nickel-cadmium batteries d. Vacuum gauge

To prevent the ethylene glycol from being drawn into the vacuum pump a "U" tube bypass device containing glass wool was attached to the outside of the portable power pack. A vacuum setting of sixteen inches of water was required to achieve an airflow of 0.1 cubic foot of air per minute through the charged impingers (see Illustration No. 2).

Matched pairs of samples were collected in the field during timed mixing and spraying operations. Each matched pair consisted of a midget impinger sample and a respirator pad for each operation, and separate respirators and impingers were used to avoid cross-contamination. In order to approximate the general area of the nostrils the impinger was pinned to the breast pocket of the operator and attached by a 24-inch piece of hose to the portable power source. There was some restriction of movement on the part of the operator, particularly in bending over, since this would allow the ethylene glycol to be aspirated past the bypass tube and into the vacuum pump. This happened on several occasions. Fortunately, the design of the system allowed us to readily disassemble the pump housing and clean it with distilled water. All the specimens from the mixing and spraying operations on the farm were collected either in the early morning or late evening when wind conditions were favorable for spraying. All measurements of pesticides on both pads and in impingers is in total nanograms. As each mixing and spraying operation was timed, calculations of nanograms per minute were made from these known elapsed times.

In all, a total of 22 paired midget impinger samples and respiratory pads were collected. These samples represent eleven mixing operations paired with eleven spray operations. They were analyzed by electron-capture gas chromatography and qualitatively by flame photometry (FPD) for confirmation of organophosphate pesticides (PERRINE PRIMATE LABORATORIES 1971). Organophosphates were not quantitated by FPD because the extracts had been stored too long by the time this detection system became operational in our analytical laboratory.

A Micro-Tek model 220 gas chromatograph (Tracor, Inc.) utilizing column packings supplied by the Perrine Primate Laboratory was used (PERRINE PRIMATE LABORATORIES 1971, THOMPSON et al. 1969a,1969b). Operating temperatures of columns, detectors, and inlet were 200° and 210° and 225°C respectively. Flow of nitrogen carrier gas was adjusted so that p,p'DDT eluted in 18-20 minutes.

## RESULTS AND DISCUSSION

Analysis of filter pads indicated that in eight out of eleven paired mixing and spraying operations there were greater total amounts of detectable pesticides found on pads used in the mixing activities regardless of time involved. In comparison of impinger substrates, five showed greater amounts during mixing, five during spraying, and one questionable. Based on these results it is apparent that the period of pesticide mixing and tank filling presents the greater potential hazard to respiratory exposure.

Table I shows amounts of compounds found by total nanograms and by rate per minute. In all cases the rate of potential exposure was greater during mixing operations for those compounds used and detected.

The type of formulation of the compound used has a bearing on the amount of recorded exposure in the mixing operation. When a formulation using 2.5 lbs. of 50% wettable powder guthion was dumped into a 500 gallon spray tank, we were able to detect 2,680 nanograms of guthion on the respirator pad and 354.6 nanograms guthion in the corresponding impinger. When six quarts of 22% emulsifiable concentrate guthion were added to a similar size tank, we were unable to recover any guthion either on the respiratory pad or in the substrate obtained from the midgetimpinger worn during the mixing operation. The total exposure times were five and three minutes, respectively. The wettable powder formulations are apt to "cloud" or "puff" back into the operator's face when the bag is opened and dumped either into another container for weighing or directly into the tank hatch opening. There appears to be more potential for dermal exposure when liquid formulations are used.

An interesting phenomenon observed is that in six paired mixing and spraying operations using captan we were able to detect captan on all six respirator pads but found none in the impinger samples (Tables I & II). This may be due to the breakdown of captan in the ethylene glycol in the impinger.

TABLE I
RESPIRATOR PAD ANALYSIS
COMPOUND USED AND DETECTED

CAPTAN

| Subject | Operation | Quantity Found (ng) | Exposure Time (min) | Quantity/ Minute (ng) |
|---|---|---|---|---|
| JS | *Mixing | 97,150 | 5 | 9430.0 |
| | *Spraying | 12,395 | 39 | 317.8 |
| | Mixing | 4,620 | 6 | 770.0 |
| | Spraying | 8,820 | 38 | 232.1 |
| GP | Mixing | 16,800 | 8 | 2100.0 |
| | Spraying | 9,800 | 24 | 408.3 |
| | Mixing | 4,788 | 3 | 1596.0 |
| | Spraying | 15,750 | 24 | 656.3 |
| | Mixing | 6,612 | 6 | 1102.0 |
| | Spraying | 4,698 | 24 | 195.8 |
| | Mixing | 4,000 | 3 | 1333.3 |
| | Spraying | 1,500 | 21 | 71.4 |

Table I (continued)

| Subject | Operation | Quantity Found (ng) | Exposure Time (min) | Quantity/ Minute (ng) |
|---|---|---|---|---|
| | | **GUTHION** | | |
| JS | Mixing | 2,680 | 5 | 536.0 |
| | Spraying | Trace | 39 | --- |
| | Mixing | Trace | 6 | --- |
| | Spraying | Trace | 38 | --- |
| | | ****ENDOSULFAN** | | |
| GP | Mixing | 182,800 | 5 | 36570.0 |
| | Spraying | 4,664 | 30 | 155.4 |
| | | **DDD** | | |
| RB | Mixing | 224,000 (p,p') | 7 | 32000.0 (p,p') |
| | | 37,440 (o,p') | | 5348.6 (o,p') |
| | Spraying | 280 (p,p') | 39 | 7.2 (p,p') |
| | | Trace (o,p') | | --- (o,p') |
| | Mixing | 5,605 (p,p') | 9 | 662.8 (p,p') |
| | | Interference (o,p') | | --- (o,p') |
| | Spraying | 500 (p,p') | 39 | 12.8 (p,p') |
| | | Interference (o,p') | | --- (o,p') |
| | | **DIAZINON** | | |
| RB | Mixing | Trace | 3 | --- |
| | Spraying | N.D. | 45 | --- |

Compounds Used But Not Detected on Respirator Pads

Copper Sulfate, Cyprex, Fundal, Lead Arsenate, Magnesium Sulfate, Manzate, Parathion, Phygon, Solubar, Sulfur, and Systox.

*The mixing and spraying operations in paired sequence represent a single spray tank filling and emptying.

**Endosulfan was identified as isomers I and II in the approximate ratio of 3:1. The commercial product was sold under the trade name of Thiodan.

N.D.=Not Detected.

TABLE II
MIDGET IMPINGER ANALYSIS
COMPOUNDS USED AND DETECTED

| Subject | Operation | Quantity Found (ng) | Exposure Time (min) | Quantity/ Minute (ng) |
|---|---|---|---|---|
| | | **GUTHION** | | |
| JS | Mixing | 355 | 5 | 70.9 |
| | Spraying | Trace | 39 | --- |
| | Mixing | 220 | 6 | 36.6 |
| | Spraying | N.D. | 38 | --- |
| GP | Mixing | N.D. | 3 | --- |
| | Spraying | 381 | 26 | 14.6 |
| | Mixing | N.D. | 5 | --- |
| | Spraying | N.D. | 30 | --- |
| | | **ENDOSULFAN** | | |
| GP | Mixing | 82,448 | 5 | 4,489.6 |
| | Spraying | 2,041 | 30 | 68.0 |
| | | **DDD** | | |
| RB | Mixing | 20,800 (p,p') | 7 | 2,971.4 (p,p') |
| | | 7,440 (o,p') | | 1,062.9 (o,p') |
| | Spraying | 123 (p,p') | 39 | 3.2 (p,p') |
| | | 37 (o,p') | | 1.0 (o,p') |
| | Mixing | 828 (p,p') | 9 | 92.0 (p,p') |
| | | 60 (o,p') | | 6.7 (o,p') |
| | Spraying | 49 (p,p') | 39 | 1.2 (p,p') |
| | | 10 (o,p') | | 0.3 (o,p') |
| | | **DIAZINON** | | |
| RB | Mixing | Trace | 3 | --- |
| | Spraying | Trace | 45 | --- |

Compounds Used But Not Detected

Captan, Copper Sulfate, Cyprex, Fundal, Lead Arsenate, Manzate D, Magnesium Sulfate, Parathion, Phygon, Solubar, Sulfur, Systox.

N.D.=Not Detected.

A limitation to our study was that in working with volunteers under actual pesticide use conditions we were restricted to evaluating those compounds used by the operator. In common practice, many pesticides are used in combinations of active ingredients. Therefore, a single spray application may contain multiple fungicides, insecticides and acaricides. Many of those actually used in this experiment were not detectable by our GLC electron capture methodology. For example, two of our paired samples used a combined formulation of DDD, copper sulfate, manzate D, parathion 8, solubar and magnesium sulfate. Of these, only DDD was detected. The parathion used was at the rate of one pint of 78% emulsifiable concentrate per 500 gallons of water.

In spite of preliminary testing in the laboratory, the midget impingers utilized proved to be inadequate for accurate measurements of pesticides in ambient air for comparison purposes in determining percentage inhaled. The flow rate appeared to be the primary limiting factor. As a result of this limitation we were unable to adequately measure ambient air levels of pesticides, and therefore were not able to determine efficacy of the modified respirator in protecting the user. One should recognize that the residues found on the respirator pads resulted from respiration in an actual work situation, whereas the impingers were set at a fixed air flow. Tables available for respiratory volume at different levels of physical activity leave much to guesswork, so no comparisons between impinger results and respirator pad results have been made on a cubic volume basis. It would have been ideal if impinger flow rate and breathing rate could have been matched. We believe another device, preferably larger in capacity, is necessary to accurately measure in ambient air for comparison purposes in a study of this type.

Even though we were able to detect greater numbers and quantities of pesticide on the modified respirator pads than we were through the impingers, the authors do not believe or intend to imply that this type respirator, even with modified pad arrangement, provides adequate protection against volatile pesticides.

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