OCCUPATIONAL EXPOSURE TO PERMETHRIN DURING ITS USE AS A PUBLIC HYGIENE INSECTICIDE

D. M. Llewellyn,* A. Brazier, R. Brown, J. Cocker, M. L. Evans, J. Hampton, B. P. Nutley and J. White
Health and Safety Executive, Magdalen House, Stanley Precinct, Bootle, Merseyside L20 3QZ, U.K.

(Received in final form 27 October 1995)

Abstract—Permethrin is an active ingredient found in many public hygiene insecticide products and exposure to it was assessed in a survey of 45 professional users. The exposures measured were over a wide range, with more than a 100-fold difference between average levels and the highest levels. Dermal contamination was evident on 93% of the operators, the highest contamination resulting from the use of leaking application equipment, demonstrating that proper maintenance of equipment is vital. Where the insecticide was applied at ground level most contamination was on the legs, indicating the importance of appropriate footwear. Contamination of the hands occurred despite the use of protective gloves, higher levels of contamination occurring when liquids were used. Dermal contamination was not always the principle route of exposure, and high airborne concentrations were linked with use in confined areas. Airborne concentrations were also associated with the physical form of the product used and the treatment method. To help in assessing the effectiveness of protective clothing and control measures, biological monitoring was carried out. Monitoring of metabolites in urine showed that systemic uptake occurred but evidence from toxicological studies indicates that the levels found were well below those considered to cause harm.

INTRODUCTION AND BACKGROUND
It is well recognized that the use of pesticides may potentially cause harm, and in an effort to minimize the dangers, many countries use regulatory schemes to control the marketing of pesticide products. Risk assessment is central to most of these schemes and common to all risk assessments, is the need to determine the hazard of the pesticide and predict the extent of exposure.

Procedures for identifying the hazards of pesticides involve toxicity studies. While there are broadly acceptable international protocols for toxicity testing (for example, OECD, 1993), at present no comparable protocols exist for assessing exposure. The nearest equivalent is a standard method published by the World Health Organization (WHO, 1993), which focuses on measuring dermal exposure to pesticides by a choice of two sampling methods.

Exposure may be estimated by monitoring during field trials or on the results of laboratory experiments, but these are rarely undertaken. It would be difficult and costly to monitor every pesticide for each of its intended uses, and so it is more usual for exposure predictions to be modelled on products for which data are already available. In practice, patterns of exposure can vary significantly owing to a number of factors including the physical form of the pesticide, the method of application and

*To whom correspondence should be addressed.
the circumstances under which the pesticide is applied. Strictly controlled field trials, laboratory experiments or models based on either of these, may not reflect the wide variation of exposures observed under normal working conditions.

To improve knowledge of non-agricultural pesticides and the way in which potential exposure occurs, a series of field surveys has been set up. Its purpose is to generate good quality, realistic exposure data which can be used to develop suitable exposure models. This paper describes the methodology, results and observations from the first study in the series, assessing exposure to permethrin in public hygiene insecticides.

Initially a pilot survey was carried out to confirm the adequacy of the analytical methods and refine the sampling strategy. This was followed by a larger main survey, the objectives of which were: to measure inhalation and dermal exposure to permethrin during application as a public hygiene insecticide; to determine if systemic uptake occurs and, if so, to what extent; to observe the effects of a wide range of factors which might influence exposure; and to obtain these data from a cross-section of industry and working situations.

SELECTION OF THE PESTICIDE

Permethrin was selected as a representative of the widely used synthetic pyrethroid group of insecticides as sensitive sampling and analytical methods were available, and because it is found in over 40 public hygiene insecticide products in the U.K.

Permethrin is a contact insecticide, which causes very rapid knockdown of flying and crawling insects. In this study it was used to control fleas, mites, flies, wasps, cockroaches and tobacco beetle, and was applied in a range of premises (public buildings, industrial premises and private dwellings).

Insecticide products usually contain the active ingredient (a.i.) with other components such as emulsifiers and solvents. The types of product in use during this survey included the following.

- Emulsifiable concentrates—an oil liquid diluted with water to give an emulsion.
- Wettable powders—a fine powder impregnated with the a.i., applied as a suspension in water.
- Dustable powders—the a.i. is mixed with, or impregnated on, an inert powder.
- Smoke generators—the a.i. is mixed with chemicals which, when ignited, produce smoke particles.
- Aerosols—the a.i. is released as minute droplets with a propellant gas.
- Soluble concentrates—concentrated forms of the a.i. are diluted with water to use.
- Ready-to-use dilute water-based forms.

No attempt was made to limit the choice of product used in the survey.

SELECTION OF SURVEY GROUP

Organizations taking part in the survey included both large and small private pest control companies as well as local government run pest control services. Pest
control staff carried out work reactively (in response to clients telephoning with specific problems) and routinely where clients premises were inspected (and treated if necessary) on a contract basis. Tasks sampled during the survey covered typical insecticide uses.

APPLICATION EQUIPMENT AND TREATMENT METHODS

Various types of application equipment were used to apply the insecticide in the most appropriate manner to control the pest.

1. Harbourages and nests were treated with either:
   (a) a coarse jet of liquid from a compression sprayer;
   (b) dusting, using piston activated or bellows pumps; or
   (c) spraying, using an aerosol can.

2. Banding involved laying a continuous band of insecticide around areas such as wall–floor junctions, across which insects would have to crawl to reach food supplies. Banding was applied by:
   (a) spraying medium to coarse droplets of liquid from a compression sprayer;
   (b) by dusting; or
   (c) by painting on a lacquer by brush.

3. Blanket coverage (applying slightly overlapping wide bands to large surface areas such as carpets and upholstery) was achieved by:
   (a) spraying liquids using a compression sprayer (medium/fine droplets); or
   (b) by dusting.

4. Smoke generators were positioned in rooms and ignited, the operator then leaving the building and allowing the smoke to carry the insecticide onto horizontal surfaces.

SAMPLING METHODS

Inhalation exposure was measured by monitoring atmospheric concentrations in the breathing zone of the operator. Personal samples were taken by drawing air through a GF/A filter mounted in a modified UKAEA sampling head. A tenax tube (standard NIOSH type) was incorporated downstream of the filter to capture any of the pesticide in its vapour phase. An aspiration rate of 0.5 l min⁻¹ was used in most cases. The samples were subsequently analysed using gas chromatography with electron capture detection.

Potential dermal exposure was determined using a modified version of a World Health Organization protocol (WHO, 1982). The method used absorbent targets which were 10 × 10 cm square gauze pads pinned or taped to the operators' clothing. This method was selected in preference to the WHO whole disposable overall method since the latter is extremely time consuming to analyse. The WHO overall method requires a great deal of solvent for extraction, followed by a concentration stage prior to analysis and would have limited the number of samples which could have been taken. Estimation of exposure from pads involves extrapolation and may be less precise but it allowed sampling of a greater range of conditions. To try to ensure that the results were representative, a large sample population was monitored.
To determine hand contamination, thin cotton gloves were worn under normal protective gloves. Both pads and gloves were analysed for permethrin using gas chromatography.

The time taken to complete insecticide treatments ranged from a few minutes to eradicate a wasps nest, to 2–3 h for a large spraying operation. The sensitivity of the analytical methods indicated that a minimum sample time of about 20 min was required so that a sensible limit of detection could be quoted for samples where permethrin was not found. Where treatments lasted 20 min or longer, sampling time corresponded with the time taken to carry out that treatment. However, in some instances (the treatment of nests, for example) where treatment times were short, sampling represents the composite of several treatments in a working day. At most sites operators worked alone, but for large jobs two, and in one instance three, men worked together. In all cases each individual was monitored.

Biological monitoring was carried out to see if systemic uptake was occurring. Following written consent, monitoring was carried out by the analysis of metabolites of permethrin in urine. Three or four samples were collected from each volunteer at the start of the day before contact with permethrin, mid-shift (where possible), at the end of the working day, and before starting work the next day.

Published data (for example, Woolen et al., 1992) suggested that these sampling times would be appropriate for the purpose. The samples were analysed using gas chromatography and mass spectrometry (Evans and Nutley, 1993).

RESULTS

Results were obtained for 45 operators from a total of 35 sites. To help interpret the results, details such as the product used, equipment used, treatment method, location and operator work method were recorded during each site visit.

Inhalation exposure

The results for airborne concentration are shown in Table 1. Figures quoted are for total (aerosol and vapour) airborne concentration. In 47% of the samples, permethrin in air concentrations were below the analytical detection limits (<5 µg on a filter, <2.5 µg on a tube). In the remaining samples, concentrations ranged from 0.007 to 237 µg m⁻³. As might be anticipated, inhalation exposure occurred more often where solid forms of the insecticide were used. Detectable airborne concentrations of permethrin were found at 79% of the sites where dustable powders were used compared with 43% where liquids were in use. Concentrations during the use of dustable powders ranged from below the limits of detection to 200 µg m⁻³.

At all but one of the sites, liquids were applied as a spray. The exception was a lacquer applied by brush and no airborne concentration was detected during its use. Spraying of liquids at ground level generally gave rise to little or no airborne aerosol. 87% of the values obtained under these conditions were <1 µg m⁻³. However, airborne concentration increased where spraying was carried out overhead when values ranged from none detected to 237 µg m⁻³, with only 33% being <10 µg m⁻³.

Where dustable powders were used, airborne concentrations varied with the method of treatment and the height at which the pesticide was applied. The lowest concentrations were recorded for nest treatment and ground level banding.
Table 1. Summary of airborne concentration data

<table>
<thead>
<tr>
<th>Formulation</th>
<th>Type of use</th>
<th>No. of samples*</th>
<th>Range (µg m(^{-3}))</th>
<th>Geometric mean (µg m(^{-3}))</th>
<th>% &lt;x µg m(^{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquids</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wettable powders</td>
<td>Blanket, ground</td>
<td>8</td>
<td>n.d.-0.07</td>
<td>0.01</td>
<td>100</td>
</tr>
<tr>
<td>Blanket, overhead</td>
<td>3</td>
<td>n.d.-52</td>
<td>0.04</td>
<td>67</td>
<td>100</td>
</tr>
<tr>
<td>Band, ground</td>
<td>4</td>
<td>n.d.-77</td>
<td>1.8</td>
<td>25</td>
<td>100</td>
</tr>
<tr>
<td>Band, overhead</td>
<td>4</td>
<td>n.d.-237</td>
<td>93</td>
<td>33</td>
<td>67</td>
</tr>
<tr>
<td>Dilute liquids</td>
<td>Blanket, ground</td>
<td>4</td>
<td>n.d.</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Soluble liquids</td>
<td>Blanket, ground</td>
<td>4</td>
<td>n.d.</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Aerosol</td>
<td>Nest</td>
<td>1</td>
<td>n.d.</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Lacquers</td>
<td>Band, ground</td>
<td>3</td>
<td>n.d.</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Solids</td>
<td>Dustable powders</td>
<td>Band, ground</td>
<td>3</td>
<td>0.03</td>
<td>67</td>
</tr>
<tr>
<td>Blanket, overhead</td>
<td>3</td>
<td>37-200</td>
<td>114</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Smoke</td>
<td>2</td>
<td>n.d.-9</td>
<td>0.48</td>
<td>50</td>
<td>100</td>
</tr>
<tr>
<td>Both</td>
<td>Dustable powders +</td>
<td>Band, ground</td>
<td>2</td>
<td>14-37</td>
<td>0</td>
</tr>
<tr>
<td>wettable powders</td>
<td>3</td>
<td>9.6-28</td>
<td>17</td>
<td>0</td>
<td>33</td>
</tr>
<tr>
<td>All samples</td>
<td></td>
<td>44</td>
<td>n.d.-237</td>
<td>0.11</td>
<td>59</td>
</tr>
</tbody>
</table>

*One operator using a range of formulations has been excluded.
Concentrations increased if banding was applied overhead, increasing further when the insecticide was blanket sprayed overhead. While the height at which the pesticide was applied affected airborne liquid concentrations, there did not appear to be any trends associated with treatment method.

The highest airborne concentrations recorded during the survey (237, 125, 198 and 200 \( \mu g \, m^{-3} \)) occurred where operators worked together in relatively confined areas (basements and corridors). These exposures appeared higher than might be predicted from the results of individuals working alone in similar situations.

**Dermal exposure**

The results of dermal sampling are summarized in Table 2. Estimation of potential dermal exposure was based on the WHO protocol (WHO, 1982) in which the mass of permethrin on the target pads is related to the exposed area of the body. The body area was divided into rectangles, based on a typical boiler suit worn by operatives. The head was treated as a cylinder 17.5 cm dia. and 25 cm high, with a percentage of the area subtracted to allow for curvature of the skull. To allow for unevenness of deposition in the areas where direct exposure was less likely, such as the insides of legs and under the arms, the final total was halved. The calculation of the whole body estimate is the sum of the amount of insecticide found on each pad multiplied by a factor relating pad area to body area, divided by two and added to the amount found on the gloves. The estimates of contamination ranged from <0.001 to 79 mg.

Contamination of the head pads ranged from <0.001 to 0.58 mg. On average this was the lowest component of total contamination, occurring mainly where overhead work was being carried out. Two operators with levels of contamination at the high end of the range (0.05 and 0.078 mg) carried out parts of their work either crouching or lying down thus potentially exposing their heads to contamination. Three other operatives carried out ground level work from a standing position but also had considerable contamination (0.58, 0.017 and 0.264 mg) of the head pad. The reason for this is not clear. They all wore disposable overalls with hoods, and observations made during sampling suggest that the pesticide may have been transferred from the outside of protective gloves to the head as the operators persistently pushed the hoods back away from their foreheads.

Contamination on the chest pads ranged from <0.001 to 1.8 mg. The operator with the highest result was observed lying on his chest at one point during the treatment. The operatives with the highest contamination levels were working in confined areas.

While the range of results for the back pad was lower than those for the chest pad (<0.001–0.919 mg), the factors influencing contamination were similar. The results from the arm pads ranged from <0.001 to 0.716 mg. The contamination on the arms of operators 6 (0.28 mg), 10 (0.65 mg) and 14 (0.17 mg) probably arose indirectly as the result of their position when applying the pesticide. It is difficult to explain the result of the operator with the highest contamination in the range (0.716 mg) since he was wearing elbow length gauntlets which would have covered the sample pad, but contamination from either the outside arm of his overall or the inside of his gauntlets may have occurred. The
<table>
<thead>
<tr>
<th>Formulation</th>
<th>Type of use</th>
<th>No. of samples*</th>
<th>Range (mg)</th>
<th>Mean (geometric) (mg)</th>
<th>% &lt; x mg</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>x = 0.1</td>
<td>x = 1</td>
</tr>
<tr>
<td>Liquids</td>
<td>Wettable powders</td>
<td>Blanket, ground</td>
<td>8</td>
<td>&lt;0.001-0.89</td>
<td>0.06</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Blanket, overhead</td>
<td>3</td>
<td>0.085-1.417</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Band, ground</td>
<td>4</td>
<td>0.014-0.212</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Band, overhead</td>
<td>4</td>
<td>0.158-11.644</td>
<td>0.82</td>
</tr>
<tr>
<td>Dilute liquids</td>
<td></td>
<td>Blanket, ground</td>
<td>4</td>
<td>0.017-79</td>
<td>1.3</td>
</tr>
<tr>
<td>Soluble liquids</td>
<td></td>
<td>Blanket, ground</td>
<td>4</td>
<td>0.101-0.772</td>
<td>0.3</td>
</tr>
<tr>
<td>Aerosol</td>
<td>Nests</td>
<td>1</td>
<td>0.05</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Laquers</td>
<td>Band, ground</td>
<td>3</td>
<td>&lt;0.001-3.29</td>
<td>0.1</td>
<td>—</td>
</tr>
<tr>
<td>Solids</td>
<td>Dustable powders</td>
<td>Band, ground</td>
<td>3</td>
<td>0.137-1.294</td>
<td>0.52</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Blanket, overhead</td>
<td>3</td>
<td>0.031-1.39</td>
<td>0.37</td>
</tr>
<tr>
<td></td>
<td>Smokes</td>
<td>2</td>
<td>0.1-0.6</td>
<td>0.24</td>
<td>—</td>
</tr>
<tr>
<td>Both</td>
<td>Dustable powders + wettable powders</td>
<td>Band, ground</td>
<td>2</td>
<td>&lt;0.001-0.08</td>
<td>0.01</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Band, overhead</td>
<td>3</td>
<td>0.182-1.343</td>
<td>0.63</td>
</tr>
<tr>
<td>All samples</td>
<td></td>
<td>44</td>
<td>&lt;0.001-79</td>
<td>0.23</td>
<td>—</td>
</tr>
</tbody>
</table>

*One operator using a range of formulations has been excluded.
higher levels of arm pad contamination corresponded with working in confined areas.

As only 26% of the jobs involved any overhead work it is not surprising that the greater part of contamination occurred on the lower body. Contamination of the thigh and ankle pads, when present, was considerably greater than that found on other regions of the body. Values for the thigh pad ranged from <0.001 to 15.9 mg while those for the ankle pad ranged from <0.001 to 149 mg. The high results for one operator (8.01 mg on the thigh pad and 149.0 mg on the ankle pad) were a result of gross contamination from a leaking compression sprayer.

Two operatives did not wear protective gloves and the levels of permethrin found on their cotton gloves were 11.331 and 0.212 mg. Excluding their results, the results from the gloves ranged from <0.001 to 1.283 mg. All these represent contamination that might have reached the skin. Most of the higher levels of hand contamination occurred when liquids were used. No correlation was noted between high levels of contamination and handling concentrated forms of the active ingredient.

**Biological monitoring**

The analysis of metabolites in urine gives an indication of total uptake but does not distinguish between routes of exposure. Samples were analysed for the following metabolites, *cis*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid (*cis*-DVCA), *trans*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid (*trans*-DVCA) and 3-phenoxybenzoic acid (3PBA). All the samples tended to contain increased metabolite levels after permethrin use. The results obtained cover a range from non-detected to 10.2 (*cis*-DVCA), 19.1 (*trans*-DVCA) and 46.2 (3PBA) mg.

![Fig. 1. Distribution of potential dermal exposure.](https://academic.oup.com/annweh/article-abstract/40/5/499/158818)
Occupational exposure to permethrin

nmoles of metabolite per mmol of creatinine. However, more than 90% of the values for any of the three metabolites measured were less than 10 nmol per mmol creatinine.

DISCUSSION

This survey shows that operator exposure almost always occurs during the normal use of permethrin, and that higher levels of exposure can be orders of magnitude greater than average levels.

Many of the studies which Crome (1985) reviewed have shown that inhalation exposure has been extremely low. This study has, however, demonstrated that during the use of permethrin in the field, inhalation may be a significant route of exposure. Based on the work of Hotchkiss et al. (1990) the relative quantities of pesticide taken up via the lungs or skin were estimated. For 56% of the operators, airborne concentrations were below the levels of detection and in these circumstances dermal contamination was more likely to occur than intake via the lungs. However, where detectable concentrations in air were found, it was estimated that inhalation was the main route of exposure.

The results, together with careful observation of the methods of handling the insecticide, have shown that a number of factors may influence the degree of exposure.

Form of the pesticide product

Crome (1985) reviewed 68 papers, mostly crop protection studies, and concluded that inhalation exposure was greater when dusts or powders were used than when liquids were used. The present study supports these findings. Detectable concentrations of permethrin in the atmosphere only occurred during treatments which involved the generation of an aerosol and atmospheric levels were clearly linked with the particle size of the aerosol generated. Droplets formed when liquids are sprayed are generally coarse and tend to settle out quickly. It was shown here that when liquids were sprayed near ground level, atmospheric concentrations in the breathing zone of the operators were below the limit of detection. Only when the insecticide was sprayed overhead were detectable levels of permethrin noted in the operator's breathing zone. Dusts and fumes have a smaller particle size so they take longer to settle out and are more likely to be susceptible to local air currents. The study confirms that the opportunity for inhalation is therefore increased. Surprisingly, dermal contamination did not appear to correlate with the physical form of product used, although contamination of the hands was higher when liquids were used.

The equipment used

Abbot et al. (1987), showed that the length of the lance on knapsack sprayers affected the distribution of dermal contamination. In the remedial timber treatment study by Tilt et al. (1992), dermal contamination was related to spray pressure. However, in this study, no trends could be demonstrated between potential exposure levels and the equipment used. This may be because they were masked by other variables.
The treatment method

Where dustable powders were used, atmospheric concentration was linked with the treatment method. Ranked in order of increasing levels of concentration were nest treatment, banding and then blanket spraying. This ranking also represents an increase in the surface area to which the pesticide was applied.

Not unexpectedly, dermal contamination to the upper half of the body usually occurred when the pesticide was applied overhead. Application near the ground resulted in contamination predominantly of the thighs and ankles. Similar effects were noted in a study by Abbot et al. (1983) for knapsack sprayers in forestry.

The location

Working in confined areas gave rise to higher levels of both airborne concentrations and dermal contamination. This effect seemed to be further compounded when two or more operatives were working together. However, with only a small number of operators working in confined areas, no firm conclusions can be drawn.

Operator work method

With the sampling methods used during the survey it was not possible to differentiate between direct (contact with airborne pesticide) and indirect (contact with treated areas) dermal contamination. However, careful observation of the work activities suggests that while direct dermal contamination is affected by the treatment location and the height at which the pesticide is applied, indirect contamination was mainly influenced by operator technique. Examples of this include the operator working lying down near recently treated areas or transfer of contamination from gloves to the head.

Other factors

Other factors which might have influenced exposure, such as treatment time, quantity of active ingredient used and application rate, were assessed, but no obvious links were found, probably owing to the varied patterns of work.

Relationship between biological monitoring results and operator exposure

The lack of correlation between the results of biological monitoring and operator exposure in this study probably best demonstrates the variation in the use and effectiveness of personal protective equipment (PPE). Observations made during the survey include the following; PPE was not always used but, when it was, it was not always of an appropriate type, was incorrectly worn, was incorrectly stored when not in use so liable to become contaminated and was not always properly maintained.

Hand contamination

The cotton gloves worn to monitor hand contamination were worn under protective gloves. If the results for the gloves were included in the calculation of regional distribution, contamination of this region would account for 7% of the total. This contamination may have arisen from penetration of the protective gloves, contamination of the insides of gloves or contamination of the hands either before
the gloves were put on or as they were removed. The study showed that contamination of the hands was more likely to occur when liquid forms of the pesticide were used.

Acknowledgements—Special thanks go to the pest control staff and the organizations for whom they work, for volunteering to take part in this survey. Thanks also to HSE’s Field Consultant Group at Manchester who carried out the pilot survey as well as inspectors, doctors, nurses and scientists who carried out the main survey.

REFERENCES
