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## — Exposure to pesticides

Part IV. The harvesting of chrysanthemums in greenhouses

## — S 131-5

TNO Arbeid, Hoofddorp



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## Exposure to pesticides

Part IV. The harvesting of chrysanthemums in greenhouses

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## FOREWORD

The Department of Occupational Toxicology of TNO Nutrition and Food Research investigates health risks of exposure to chemical substances in the working environment on request of the Ministry of Social Affairs and Employment. The present study is the fourth part of the research project 'Pesticides in greenhouses for flower culture'. The first part was performed in greenhouses for carnation culture, the second part in greenhouses for rose culture and in the third part the exposure during application of pesticides to chrysanthemums in greenhouses was studied.

These three parts are also published in this series (Brouwer et al., 1991<sup>ab</sup>; De Vreede et al., 1994). In the present part, the determinants of exposure to pesticides after re-entry during harvesting of chrysanthemums in greenhouses are investigated.

We thank the management of the 'Nederlandse Tuinders Studieclubs' (NTS), section Flowers and Plants, for their cooperation, especially 'de landelijke chrysantencie' who has supported this study in a positive way towards the flower growers.

For their assistance in performing the field work, we want to thank Sjaak de Vreede and Ronald Hoevers. For the chemical analyses of the samples we extend special thanks to Erik Hoogduyn and Roel Engel.

Finally, the cooperating chrysanthemum growers, without whom it would not have been possible to carry out this investigation, are gratefully acknowledged.

## SUMMARY

In 12 greenhouses for chrysanthemum culture in the Netherlands, dermal exposure of hands and forearms, and respiratory exposure to chlorothalonil during harvesting activities have been measured. Two harvesting techniques were compared, during which respectively the highest and lowest levels of exposure were expected. The difference between both techniques was the way some actions (removing leaves and clods) were carried out: manually or automatically with a harvesting machine. Those greenhouses were selected that used chlorothalonil with a high-volume spraying technique and one of the harvesting techniques. In addition, in four greenhouses, the dislodgeable foliar residue (DFR) of chlorothalonil on chrysanthemum plants has been monitored throughout the growing period, as well as the percentage of the applied dose that resulted in foliar deposition.

It appears that dermal exposure differs for the different harvesting techniques. The exposure was on average 3.6 mg/h (geometric mean; active ingredient) for harvesters who pull the plants out of the ground and break the clod off the plants by hand (manual harvesting), 1.1 mg/h for harvesters who only pull out the plants (automatic harvesting) and 1.1 mg/h for workers who put the bunches of flowers into covers (wrapping). Re-entry time had a large influence on exposure. Working after a relatively short re-entry interval (11-17 days) resulted in an exposure of about a factor 6 to 9 higher than after a long re-entry interval (32-56 days). The respiratory exposure during all tasks was less than 1% of total potential exposure to chlorothalonil.

The intensity of skin contact during different tasks is expressed as a transfer factor which is calculated as ratio of dermal exposure and dislodgeable foliar residue. Transfer factors (geometric mean) established in this study were approximately 1300 cm<sup>2</sup>/h for manual harvesting, 1000 cm<sup>2</sup>/h for automatic harvesting and 800 cm<sup>2</sup>/h for wrapping. Variation in these task specific transfer factors was relatively high. The calculation of transfer factors for separate activities showed that clod breaking/removing leaves (usually combined with pulling out plants) results in the highest transfer (2000 cm<sup>2</sup>/h) and bundling in the lowest (800 cm<sup>2</sup>/h), as expected. Transfer during pulling was approximately 1200 cm<sup>2</sup>/h. Variation was still high, indicating other influences like differences in work practice and work rate

and probably re-entry time, besides inaccuracy due to low values of DFR.

The variance in dermal exposure could be explained for 60% by differences in dislodgeable foliar residue (DFR) that was shown to be a measure of source strength. A relation was found between the Leaf Area Index (LAI), as a measure of crop density, and the percentage of the applied dose that resulted in foliar deposition. With this relation, the DFR for the chrysanthemum crop can be predicted from the Leaf Area Index. In general, the level of foliar residues of chlorothalonil decreases between two applications. After an application most DFR is found in the top part of the plant. This implies that the time of application during crop growth affects the location in the plant where the pesticide residues will be found. This will affect exposure as it is related to manual crop activities.

## SAMENVATTING

In 12 chrysantenteelt bedrijven is de huidblootstelling van beide handen en onderarmen, en de inhalatoire blootstelling aan chloorthalonil gemeten tijdens oogstwerkzaamheden. Er zijn twee oogsttechnieken vergeleken, bij welke respectievelijk de hoogste en de laagste blootstellingsniveaus werden verwacht. Het verschil tussen beide technieken was de wijze waarop bepaalde handelingen (kluitbreken en ritsen) werden uitgevoerd: handmatig of automatisch met behulp van een oogstmachine. Bedrijven werden geselecteerd op grond van het gebruik van chloorthalonil gecombineerd met een hoogvolume spuittechniek en één van beide oogsttechnieken. Verder is bij vier bedrijven een vak chrysanten vanaf het planten tot de oogst gevolgd door steeds voor en na een toepassing met chloorthalonil het afveegbaar residu van deze stof op het blad te meten. Op deze manier is de verdeling van het middel over de tak bepaald en het percentage van de toegepaste hoeveelheid bestrijdingsmiddel die tijdens de toepassing door het gewas werd afgevangen.

Het is gebleken dat de huidblootstelling aan chloorthalonil verschilt voor beide oogsttechnieken. Deze blootstelling was gemiddeld 3,6 mg/uur (geometrisch gemiddelde; actieve stof) voor oogsters die met de hand de kluit van de plant afbreken, 1,1 mg/uur voor oogsters die niet kluitbreken en 1,1 mg/uur voor medewerkers die de bossen chrysanten inhoezen. De periode tot herbetreding, dit is het aantal dagen tussen de laatste toepassing en de oogst, had grote invloed op de blootstellingsniveaus. Na een relatief korte herbetredingsperiode (11 tot 17 dagen) werden zes tot negen maal hogere blootstellingsniveaus gevonden dan na een lange periode (32 tot 56 dagen). De inhalatoire blootstelling droeg tijdens alle taken minder dan 1 % bij aan de totale potentiële blootstelling aan chloorthalonil.

De intensiteit van het contact met het gewas tijdens de verschillende taken is uitgedrukt als een overdrachtsfactor, berekend als ratio van de huidblootstelling en het afveegbaar residu (AR). Voor kluitbrekend oogsten, automatisch oogsten en hoezen konden overdrachtsfactoren worden berekend van respectievelijk 1300, 1000 en 800 cm<sup>2</sup>/uur (geometrisch gemiddelde). De spreiding in deze overdrachtsfactoren was relatief groot. Berekening van overdrachtsfactoren voor de aparte handelingen liet zien, dat tijdens oogsten het kluitbreken/ritsen de hoogste overdracht geeft (2000 cm<sup>2</sup>/uur) en het bossen de laagste (800 cm<sup>2</sup>/uur), zoals verwacht. Overdracht tijdens het uit de grond trekken van



de planten was ongeveer 1200 cm<sup>2</sup>/uur. Het feit dat de spreiding nog steeds groot was, wijst erop dat er naast onnauwkeurigheid door lage waarden van het AR, andere invloeden op de overdracht zijn, zoals verschillen in werkwijze en werktempo en waarschijnlijk de herbetredingsperiode.

De variatie in huidblootstelling kon voor gemiddeld 60% verklaard worden door de variatie in de hoeveelheid afveegbaar residu, dat als een maat voor bronsterkte beschouwd kan worden. Er is een relatie gevonden tussen de Leaf Area Index (LAI), als maat voor de gewasdichtheid, en het percentage van de toegepaste hoeveelheid bestrijdingsmiddel dat door het gewas wordt afgevangen. Op grond van deze relatie kan het afveegbaar residu voor het chrysantengewas voorspeld worden uit de LAI. Over het algemeen is er tussen twee toepassingen sprake van een afname van het afveegbaar residu. Meestal vindt na een toepassing de sterkste toename van het afveegbaar residu plaats in de bovenste zone van de plant. Dit betekent dat het tijdstip in de teelt waarop wordt toegepast, invloed heeft op de plaats in het gewas waar het middel tijdens de oogst teruggevonden wordt. Dit kan de huidblootstelling beïnvloeden, aangezien de blootstelling gerelateerd is aan contact met het gewas.

## **1 INTRODUCTION**

### **1.1 Pesticides and exposure**

Pesticides are used frequently in many sectors in agri- and horticulture in the Netherlands. Especially in the culture of flowers in greenhouses, relatively large amounts of pesticides are applied. The largest part of the flower production is exported and there is an absolute requirement for absence of (traces of) plague or disease in the flowers to be exported. Exposure to pesticides mainly occurs during mixing, loading and application of pesticides and during manual activities in treated crops, usually referred to as re-entry. Only a limited number of persons is involved in the application of pesticides during relatively short periods of time, whereas nearly all workers in greenhouses are daily exposed to pesticides because of manual crop activities. In the Dutch flower culture, a population of about 25,000 persons is potentially exposed to pesticides during the production of cut-flowers and pot-plants in greenhouses.

Major routes of exposure to pesticides after re-entry are airways and skin. Respiratory exposure arises from residual pesticide vapour or from airborne particles contaminated with pesticides. Dermal exposure may originate from airborne particles as well as from direct contact with the crop carrying foliar residues of pesticides.

Exposure after re-entry has led to incidents of illness among fieldworkers in citrus and peach crops in the USA (Gunther et al., 1977; Maddy et al., 1990). From the observed levels of dermal exposure during cutting of carnations in greenhouses, Brouwer et al. (1992<sup>a</sup>) concluded that for crop activities a health risk may exist for pesticides with a low 'no-effect level' and good skin-penetrating properties.

To prevent adverse health effects in workers caused by pesticides, a health risk evaluation is required for pesticide approval procedures. Only those pesticides of which the health risk based on a proper health risk evaluation is acceptable and controllable, may be considered for approval (Bestrijdingsmiddelenwet 1962).

For a health risk evaluation of a pesticide, data on toxicology and on occupational exposure are required. Toxicity is an intrinsic property of a particular pesticide. Exposure however, is largely dependent on the job being done, how it is done and under what conditions, rather than on the chemical nature of the product. In case of re-entry, exposure is highly determined by (Van Hemmen, 1992):

- the type of activity (bud removal, cutting, sorting, bundling);
- agricultural conditions (crop, level of mechanization, application technique, application rate, formulation type, re-entry interval);
- climatic conditions (temperature, relative humidity, wind conditions);
- work aspects (protective equipment, work practices, personal hygiene).

In order to be able to extrapolate exposure data on one pesticide during re-entry, to other pesticides, the influence of these factors on the level of exposure must be investigated. Such an investigation may also yield information for adequate control measures.

Previous studies in this series on exposure to pesticides have been conducted in the cultivation of carnations and roses (Brouwer et al., 1991<sup>ab</sup>). Together with chrysanthemums, carnations and roses account for approximately 50% of the total acreage for flower culture in greenhouses in the Netherlands (IKC, 1993<sup>a</sup>).

## 1.2 Aim of the study

The aim of the present study was to gain insight into the determinants and the extent of exposure to pesticides after re-entry for different harvesting techniques in the cultivation of chrysanthemums in greenhouses.

For various reasons (described in § 3.1) the pesticide chlorothalonil was selected for the estimation of the level of exposure.

The following questions are to be answered:

- What are the levels of dermal exposure to chlorothalonil of the left and the right-hand during the various harvesting tasks for different harvesting techniques?
- What are the levels of respiratory exposure to chlorothalonil during the various harvesting tasks for different harvesting techniques?
- What is the relation between dermal exposure during the various harvesting tasks for different harvesting techniques, and the amount of chlorothalonil on the foliage (dislodgeable foliar residue)?
- What is the relation between the amount of chlorothalonil that results in foliar deposition after application and the growing phase of the crop?
- What is the course in time of dislodgeable foliar residue and how is it spread over the plant?
- What are the possibilities to extrapolate the results of chlorothalonil to other pesticides used in the cultivation of chrysanthemums?

## **2 STUDY DESIGN**

### **2.1 The cultivation of chrysanthemums**

The total acreage for chrysanthemum culture in the Netherlands in 1992 was 766 ha, which amounts to 19% of total acreage for cut-flower culture in greenhouses, spread over about 750 greenhouses. In total about 2,700 workers are involved in the production of chrysanthemums (PTG, 1994). In the culture of chrysanthemums, a distinction can be made between the production of chrysanthemum cuttings and the production of flowers. Previously, a study has been conducted concerning the first (Peelen, 1992<sup>a</sup>). The present study relates to the latter.

Chrysanthemums are grown by using cuttings from mother plants. It takes about 13 weeks before cuttings have grown into harvestable crops. This means that about four chrysanthemum cultures per year are feasible. Chrysanthemums are harvested just once, unlike roses and carnations which are cultured respectively for at least 5 years and 1-2 years. This difference largely affects the common re-entry interval before harvesting. In the cultivation of roses and carnations, re-entry occurs after 1 or 2 days, whereas in the cultivation of chrysanthemums, it may be a matter of weeks.

Chrysanthemums may be affected by various diseases and plagues, e.g. leafminer, thrips and Japanese rust. Since no damage is tolerated in flowers for the export, the amount of pesticides used may be relatively large. Often several pesticides are applied simultaneously in a mix. The amount of pesticides used is on average 223 kg active ingredient per ha per year (estimate from about 1988), i.e. about 56 kg per culture. This value is decreasing rapidly. With the exception of roses, 15 to 20 kg active ingredient per ha per year is used for other flower cultures (LNV, 1990).

### **2.2 Harvesting techniques**

During pilot studies an inventory was made of harvesting techniques often used in practice. Consecutive actions during harvesting of chrysanthemums are (in brackets the short description):

- a) pulling the plants out of the ground (pulling)
- b) removing the lower leaves in one movement (removing leaves)

- c) breaking the clod off the plant (clod breaking)
- d) collecting the flowers into a bunch on the arm or on a cord or conveyor belt (bundling)
- e) binding bunches of flowers with elastic strings (binding)
- f) wrapping bunches of flowers into covers and putting them into boxes (wrapping)

Actions a, d and f are only done manually in practice, whereas b, c and e can be done either manually or automatically with a harvesting machine. Harvesting techniques are determined by different combinations of manual and automatic actions:

- 1) All actions are done manually by one worker.
- 2) All actions are done manually by at least two workers (actions a-d by one worker and e-f by another worker; the bunches are in some cases transported by means of a conveyor-belt).
- 3) Action a, d and f are done manually by two workers. Action b, c and e are done automatically by means of a harvesting machine.

Other combinations may occur. A survey on the three distinctive harvesting techniques mentioned, appeared to be not feasible on account of the occurrence of these techniques in greenhouses using the selected pesticide (§ 3.1). Therefore the approach in the present survey has been to study two harvesting techniques, during which respectively the highest and lowest levels of exposure were expected, assuming that levels of exposure depend on the intensity of contact with the treated crops. These techniques were defined as follows:

- A) pulling, removing leaves and clod breaking manually and bundling by one worker, followed by binding manually by another worker (method 2) or automatically and wrapping;
- B) pulling and bundling by one worker, automatically removing leaves, sawing off clod and binding; then wrapping by another worker (method 3).

The difference between both techniques that was of most interest in this survey, was the way action b and c, removing leaves and clods, were carried out: manually or automatically. Method A (manually) was expected to result in the highest exposures, whereas method B (automatically) probably would result in relatively low exposures, as the intensity of contact with the plants was minimal.

### 2.3 Monitoring strategy

Measurements have been carried out in 10 different greenhouses equally divided between the two harvesting techniques described above.

Major routes of exposure to pesticides are airways and skin, so personal respiratory and dermal exposure have been measured. Two harvesting tasks have been distinguished, namely harvesting (action a-d) and wrapping (action e-f). In all greenhouses visited, two harvesters and one wrapper were working together at the same time. Dermal exposure has been measured for all workers, whereas respiratory exposure has been measured for all wrappers but for only one harvester per greenhouse.

The transfer of pesticide residue from the surface of the leaves to the skin of the worker is most important for dermal exposure (Popendorf and Leffingwell, 1982; Popendorf, 1985; Brouwer et al., 1992<sup>b</sup>) and depends on two factors: the amount of pesticide available and the intensity of skin contact with the treated crops.

The amount of pesticide on the leaves which is available for transfer at harvest, depends on the initial deposit and the rate of decline of the pesticide after application (Bates, 1990). Some factors which affect the initial deposit are the dose applied per acreage, frequency of application, the type of formulation and the application technique. The characteristics of the crop itself like growth stage and the nature of the crop surface play also an important role, e.g. in the degree of interception of the pesticide by the crop. The rate of decline after application is determined by growth dilution and degradation of the pesticide due to various physical, chemical and biological processes, e.g. rain irrigation, evaporation, oxidation, hydrolysis, photodegradation or plant metabolism (Bates, 1990).

In this survey, the application technique was chosen to be the same for all cases and other variables were recorded if possible. The amount of pesticide available at harvest has been quantified as the dislodgeable foliar residue (DFR) of leaves taken from two contact zones, namely high and low in the plant. In four greenhouses the course in time of DFR in relation to crop growth has been followed by measuring DFR before and after each application. The ratio of total leaf surface and ground area covered by the crop is used as a measure for the growing stage of the crop. This ratio is called the Leaf Area Index.

The intensity of skin-crop contact is determined by the crop-related factors, e.g. density and height, by the type and frequency of manual activities and by worker-dependent

factors such as work practice and clothing. The intensity of contact related to exposure is integrated in a crop- and task-specific transfer factor. The intensity of contact per action could only be ranked in a qualitative manner. For instance, the intensity during removing leaves and clod breaking was considered to be higher than during pulling and bundling. Since some of these actions were done left-handed and others right-handed, it was meaningful to measure dermal exposure of both hands separately. Information on individual work practices enabled linking of exposure of left or right hand to the dislodgeable foliar residue of the zone in the plant where most skin contact with this hand was observed. In this way, transfer factors per hand could be calculated. Removing leaves and clod breaking was usually done with one hand, so manual harvesting may result in a relatively high level of exposure of one of the hands, so that differences between tasks and harvesting techniques may become more evident.



### **3 MATERIALS AND METHODS**

#### **3.1 Selection of greenhouses and pesticide**

Through the help of the NTS (Dutch horticulture study clubs) a list of addresses was obtained from the largest flower auction in the Netherlands of 411 chrysanthemum greenhouse owners that used more than 0.25 ha for chrysanthemums. A questionnaire was sent to these greenhouse owners to gather information on the use of pesticides, the seasonal variation in use, the application techniques, the techniques used in harvesting, and consent to participate in the survey. 239 greenhouse owners returned the questionnaire (58%) of which 10 returned it blank and 48 didn't grow chrysanthemums at all or not during the whole year. In the remaining 181 greenhouses the only culture was chrysanthemums. Of the 181 greenhouse owners 99 (55%) were willing to participate in the survey.

The information of the 181 questionnaires was used to make an inventory of the most frequently used (used in most greenhouses) pesticides at that moment (1991) and a top 10 list was drawn up of these pesticides (Table 3.1). Criteria for the selection of a suitable pesticide were (i) the availability of a validated analytical method, (ii) a widespread use of the pesticide in the cultivation of chrysanthemums in greenhouses, (iii) the number of greenhouse owners using the pesticide and willing to participate in the survey, in combination with another important criterion, namely (iv) the concentrations of pesticide that could be measured under field conditions. Whether concentrations are measurable in practice depends on the application rate and the rate of decline of foliar residues as a result of growth dilution, degradation of the pesticide and disappearance of pesticide on account of contact and rain irrigation. Indications on the rate of decline of several pesticides were derived from pilot studies (Peelen, 1992<sup>b</sup>). Information on stability was also obtained from the literature (Hartley and Kidds, 1983). In view of the rate of decline and the low (near the detection limit) levels of dislodgeable foliar residues found in pilot studies, abamectin, dichlorvos, heptenofos and methomyl could not be considered for selection. Methiocarb was not admitted for the cultivation of chrysanthemums. Bitertanol was used in less than 30% of the greenhouses which was not enough to study the differences in exposure between the harvesting techniques. The same reason applied to pyrazofos, oxamyl, pirimicarb and deltamethrin. Leaving triforin as the best alternative.

However, by that time (fall 1992) a new inventory of the use of pesticides had to be made, due to supposed resistency of Japanese Rust to triforin and an expected drop in its use. Possible substitutes of triforin were chlorothalonil, mancozeb and tolylfluanid (Euparene); chlorothalonil and mancozeb being applied early in the growing-period due to undesirable visible residues and tolylfluanid late in the growing-period due to possible damage to young plants. Further information indicated that Euparene was not used on a large scale.

A second inquiry into the use of chlorothalonil, mancozeb, triforin and methomyl was held in a subpopulation of greenhouse owners willing to participate and using a spray pistol (one nozzle), by far the most widespread application technique (98%) (De Vreede et al., 1993). This inquiry resulted in about 17 greenhouses using chlorothalonil, and being measurable in practice this was the pesticide finally chosen for the survey.

Table 3.2 shows the occurrence of three different harvesting techniques in the cultivation of chrysanthemums in greenhouses. Because of the use of these techniques in the greenhouses willing to participate and using chlorothalonil, the survey was confined to comparing only two techniques, namely harvesting manually (two workers) and using a harvesting machine (as described in § 2.2). Selected greenhouses using chlorothalonil and a high-volume application technique were equally divided between these two harvesting techniques. Measurements were carried out in 10 different greenhouses in two of which was measured twice.

In four of the selected greenhouses, two for each harvesting technique, a supplementary study has been conducted to study the course in time of dislodgeable foliar residue after application. The owners of these greenhouses were asked to do an application of chlorothalonil about 14 days before the harvest.

Table 3.1 The use of pesticides in the cultivation of chrysanthemums (1991)

| Active ingredient | Pesticide  | Number of greenhouses (percentage) | Number of greenhouses willing to participate (percentage) |
|-------------------|------------|------------------------------------|-----------------------------------------------------------|
| abamectin         | Vertimec   | 177 (98%)                          | 99 (56%)                                                  |
| triforin          | Funginex   | 124 (69%)                          | 70 (56%)                                                  |
| methomyl          | Lannate    | 75 (41%)                           | 38 (51%)                                                  |
| dichlorvos        | various    | 62 (34%)                           | 40 (65%)                                                  |
| heptenofos        | Hostaquick | 61 (34%)                           | 42 (69%)                                                  |
| methiocarb        | Mesuroi    | 54 (30%)                           | 38 (70%)                                                  |
| bitertanol        | Baycor     | 53 (29%)                           | 26 (49%)                                                  |
| pyrazofos         | Curamil    | 51 (28%)                           | 29 (57%)                                                  |
| oxamyl            | Vydate     | 49 (27%)                           | 26 (53%)                                                  |
| deltamethrin      | Decis      | 38 (21%)                           | 23 (61%)                                                  |
| pirimicarb        | Pirimor    | 38 (21%)                           | 22 (58%)                                                  |

Table 3.2 Harvesting techniques in the cultivation of chrysanthemums (1991)

| Harvesting technique <sup>a</sup> | Number of greenhouses (percentage) | Number of greenhouses willing to participate (percentage) |
|-----------------------------------|------------------------------------|-----------------------------------------------------------|
| 1) manually by one worker         | 54 (30%)                           | 27 (50%)                                                  |
| 2) manually by two workers        | 14 (8%)                            | 6 (43%)                                                   |
| 3) harvesting machine             | 138 (76%)                          | 88 (64%)                                                  |
| total <sup>b</sup>                | 206                                | 121                                                       |

<sup>a</sup> as described in § 2.2

<sup>b</sup> more than one technique per greenhouse may be used

### 3.2 Assessment of dermal exposure

Dermal exposure of all workers in the selected greenhouses was measured by means of (prewashed) cotton glove monitors covering hand and forearm (stretch-cotton: 275 g/m<sup>2</sup>, surface area (one-sided) 370 cm<sup>2</sup>; J. van der Wee B.V., Riel, The Netherlands). Measurements were performed in the treated crop during the period of harvesting. A pair of gloves was used for a maximum period of one hour, in order to prevent breakthrough of the gloves followed by transfer of pesticide from glove to hand. In a pilot study the breakthrough was found to be less than 5% for chlorothalonil after one hour. The breakthrough was determined by means of a hand-washing procedure that used an isopropanol/water mixture (60/40 v/v). This procedure was validated for chlorothalonil in laboratory experiments with five volunteers. In these experiments, the effectiveness of washing was determined as the amount of pesticide washed off the hands divided by the net amount put on the hands (expressed as a percentage). An average effectiveness of 74% (SD = 11%) was found, which was sufficient to use the hand-washing procedure for studying the breakthrough of the gloves (annex 1).

After a break in the work period or when the work period took more than one hour, the workers were provided with a new set of gloves. The gloves of the left and right hand were collected separately. Both left or right hand gloves used by a worker during the total work period were treated as one sample. Samples were stored in polyethylene bottles in the dark at 4°C awaiting chemical analysis for the amount of pesticide.

### **3.3 Assessment of respiratory exposure**

Respiratory exposure during harvesting was measured in each greenhouse of one of the harvesters and of the wrapper. Measurements were carried out using an IOM personal air sampler (IOM, Negretti Automation, Aylesbury, England) with a Mixed Cellulose Ester filter (25 mm, pore diameter 8  $\mu$ m, Millipore Corporation, USA) attached to a constant-flow air-sampling pump operating at 2 l/min (S2500A, Dupont, USA), estimating the inhalable fraction (ACGIH, 1985; Mark and Vincent, 1986; CEN, 1992; ISO, 1992). Flows were checked before and after the sampling period by using a precalibrated rotameter tube (ROTA, Dr Hennig GmbH, Germany) and weren't allowed to differ more than 10%. Filters were stored in polypropylene tubes (Greiner und Söhne GmbH, Germany) in the dark at 4-7°C awaiting chemical analysis for the amount of pesticide.

### **3.4 The sampling of leaves**

After re-entry (during harvesting) several samples of 12 leaves were taken from randomly selected flowers in the sector of the greenhouse that was being harvested, at the height the flowers were handled. They were taken from the high and low contact zone in the plants. If a contact zone differed more than about 10 cm between both harvesters, samples were taken in duplicate form for each harvester at the specific height where contact occurred. If a contact zone was at similar height, samples were taken in triplicate form for this contact zone at the average height. All relevant heights were recorded. Samples were stored in polyethylene bottles in the dark at 4-7°C awaiting further processing.

In four greenhouses additional leaf samples were taken before and after each application with chlorothalonil, preferably less than 24 hours before and after the application. However, before the application it occurred twice that the interval was longer than 24 hours, namely respectively 2 and 4 days. After the application a longer interval occurred four times, namely 2 days (twice) and 3 days (twice).

The sampling procedure was as follows. Twelve plants were taken randomly from the sector of the greenhouse that was being studied. The height was measured and each plant was divided into three equal parts (the zones high, middle and low). From each zone, one

leaf was taken at random for the assessment of dislodgeable foliar residue (DFR). These 12 leaves per zone were stored in a polyethylene bottle in the dark at 4-7 °C awaiting further processing. The rest of the leaves were cut from the plants too and collected per zone.

For each zone, total leaf surface area (one-sided) was measured with an area meter (LI-COR, 3100, Lincoln, Nebraska, USA). The Leaf Area Index (LAI) was calculated for each zone and application and used as a measure of crop density.

### **3.5 Assessment of dislodgeable foliar residue**

After the collection of leaves, dislodgeable residues were obtained through a procedure based on the method described by Iwata et al. (1977). Briefly, approximately 400 cm<sup>2</sup> leaf area of chrysanthemum leaves were extracted twice by shaking for 30 minutes with about 400 ml distilled water containing 16 drops of a Triton X-100 solution (Triton X-100-water 1:50 v/v). Then the bottle containing the leaves was rinsed with another 100 ml water and after removal of the leaves it was rinsed once with 400 ml methanol. From other leaf areas the mentioned amounts were adjusted proportionally. All extract liquids were combined and the solution was shaken for 30 minutes. This solution containing the dislodgeable residue was analysed for chlorothalonil. Projected leaf surface area (one-sided) was measured with an area meter (LI-COR, 3100, Lincoln, Nebraska, USA).

To assess the measure of representiveness of a sample of 12 leaves for a particular zone in the whole crop, a total of 10 test samples was collected by two researchers during harvesting in one greenhouse. The precision of the assessment of dislodgeable foliar residue depends not only on the representiveness of the samples, but also on the variation of sampling, the variation between persons taking samples and the variation of chemical analysis. Resulting coefficients of variation based on the test samples were 20% and 35% for the respective sample takers, the difference being significant (annex 2). The coefficient of variation ( $CV_{total}$ ) based on the series of the regular duplicate samples using the 'percentage of difference' method (Boleij et al., 1987) was 2.4% ( $n = 26$  duplicates). This value is lower, because duplicates were taken from plants close to one another.

### **3.6 Chemical analysis**

Methods of chemical analyses were developed by the present laboratory. Details have been described elsewhere (Jongen et al., 1991).

In solutions of dislodgeable foliar residue (DFR) or methanol extracts of gloves or filters, chlorothalonil was quantified by means of reversed-phase liquid chromatography with methanol-water (3:1 v/v) containing 5 g/l ammoniumacetate as eluent on a C<sub>18</sub> column (Biosil C18HL 150 4.6) with UV detection at 232 nm. The analytical limit of detection (LOD) was fixed on 40 µg/l for filters and 100 µg/l for DFR samples and gloves, which means that a set of gloves, a sample of 400 cm<sup>2</sup> leaf area and a filter must contain an amount of chlorothalonil of respectively 50, 120 and 0.2 µg to be detected. In practice sometimes larger or smaller amounts could be detected depending on the degree of interference caused by pollution of the samples. Stored at 4°C in the dark, no loss of chlorothalonil on filters and DFR solutions was observed within 22 days and on gloves within 70 days. Analytical recovery from filters, gloves and DFR solutions was more than 95%. The between-day analytical coefficient of variation was less than 5% for all matrices.

### **3.7 Additional observations**

The individual work practices were described giving information on which hand (left or right) was used for which action (pulling, bundling, removing leaves, clod breaking, wrapping) and for each action the height of the contact zone with the plant. The greenhouse owners were asked to keep a log-book of all applications to the flower-bed under study (pesticides used, concentration of pesticide in spray liquid, volume sprayed per area). Application rates were calculated based on the log-book information.

The work rate, expressed as number of flowers handled per hour, was estimated by dividing the total number of flowers harvested over the sampling period, by the number of harvesters.

### **3.8 Quality control**

For quality assurance blank and spiked samples were taken at the site to assess background levels in the greenhouse and stability of the samples during transport.

Blank samples of gloves were taken by putting one glove from the stock into a 1 litre polyethylene bottle. Blank filters were treated as normal samples except for sucking the air. These partial blanks served to assess contamination during the handling of filters.

Background levels in the greenhouses were determined by means of a stationary measurement of chlorothalonil in air using the method described in § 3.3.

Two gloves were spiked with 1 ml of a standard solution of chlorothalonil (1.5 g/l), which corresponded to the amount expected during the sampling period. One spiked glove was stored directly and the other was laid down during measurement somewhere in the greenhouse. No spiked filters were taken, because filters were damaged by spiking.

### **3.9 Statistical analyses**

The exposure data were statistically analysed using SOLO Statistical System (BMDP Statistical Software Inc., Los Angeles, USA, 1988) for personal computer. The dermal as well as the respiratory exposure data were log-transformed to meet the normal distribution. Results are expressed as geometric mean (GM) and geometric standard deviation (GSD). For studying differences in dermal exposure, exposures of the left and right hand were added for each person, since these were significantly (positively) correlated ( $r = 0.82$ , Spearman's rank correlation test). Dermal exposure data obtained in the two pilot studies were included, since dermal exposure had been measured using the same strategy as in the survey. Log-transformed dermal exposure data were approximately normally distributed (Kolmogorov-Smirnov normality test  $p > 0.05$ ). Differences between groups were tested by the non-parametric Mann-Whitney test (Snedecor and Cochran, 1982). A probability of  $p < 0.05$  was considered to be significant.

The association between dermal exposure (left and right hand separately) and dislodgeable foliar residue (DFR) and other variables was studied by (multiple) linear regression using least square estimates. For dislodgeable foliar residue data below the limit of detection the half of this limit was used, being an acceptable method for handling nondetectable values, if GSD is high and the percentage nondetectable values is low (Hornung and Reed,

1990). Data from the two pilots were excluded from this analysis, because the leaves for the assessment of DFR were sampled in a different way. Log-transformed personal dermal exposure data of both hands separately and DFR data were not normally distributed (Kolmogorov-Smirnov test  $p < 0.05$ ), but log-transformation of the data did improve the analysis so the log-transformed data were used. Log-transformed personal and background respiratory exposure data were normally distributed (Kolmogorov-Smirnov test  $p > 0.10$ ).



## 4 RESULTS

### 4.1 Dermal exposure after re-entry

Dermal exposure of 36 workers to chlorothalonil (both hands separately) was measured during harvesting of chrysanthemums in 12 greenhouses. They worked in crops that were treated 11 to 56 days before, with an average of 27 days. Application rates varied from 44 to 279 gram active ingredient per 1000 m<sup>2</sup> cultivated area, the average (119) being below the recommended 150 g/1000 m<sup>2</sup> (IKC, 1993<sup>b</sup>). Liquid formulations were used. In one greenhouse a powder was used instead. Glove monitors were worn for an average of 94 minutes. There were no significant differences in re-entry time, application rate and sampling period between greenhouses using the different harvesting techniques. In greenhouses where clods were removed automatically, about 20% more flowers were harvested per hour compared to greenhouses where clods were broken by the workers. This difference was not significant. More details are presented in Table 4.1.

Table 4.1 General data on dermal exposure measurements during the harvest of chrysanthemums.

| Harvesting technique:                                          | Manual<br>clod breaking |       | Automatic<br>clod removal |       |
|----------------------------------------------------------------|-------------------------|-------|---------------------------|-------|
| Number of measurements                                         | 36                      |       | 36                        |       |
| during harvesting                                              | 24                      |       | 24                        |       |
| during wrapping                                                | 12                      |       | 12                        |       |
| Work rate <sup>a</sup><br>(number of flowers handled per hour) |                         |       |                           |       |
| during harvesting                                              | 1072                    | (306) | 1266                      | (310) |
| during wrapping                                                | 2021                    | (649) | 2468                      | (582) |
| Sampling period (min) <sup>a</sup>                             | 96                      | (19)  | 92                        | (12)  |
| Re-entry time (days) <sup>a</sup>                              | 24                      | (18)  | 29                        | (13)  |
| Application rate (g a.i./1000 m <sup>2</sup> ) <sup>a</sup>    | 130                     | (90)  | 109                       | (87)  |

<sup>a</sup> expressed as arithmetic mean and standard deviation (in parentheses); a.i. = active ingredient

The results on total dermal exposure of both hands are presented in Table 4.2. It appeared, as expected since the activities are not different, that exposure during wrapping did not differ between both harvesting techniques (annex 3), so three tasks were distinguished, namely harvesting using the different harvesting techniques and wrapping. Further, two clusters could be distinguished in the large range of re-entry times, namely short and long re-entry time, respectively from 11 to 17 days and from 32 to 56 days with no

observations in between. Dermal exposure in these two clusters differed significantly ( $p < 0.0001$  Mann-Whitney test). Therefore dermal exposure is stratified for re-entry time, although this resulted in unbalanced groups with few observations.

Table 4.2 Dermal exposure to chlorothalonil of both hands and forearms.

|                                      | Dermal exposure (mg/h) <sup>a</sup> |                          |   |                         |    |                         |
|--------------------------------------|-------------------------------------|--------------------------|---|-------------------------|----|-------------------------|
|                                      | n                                   | short re-entry           | n | long re-entry           | n  | total                   |
| harvesting<br>manual clod breaking   | 8                                   | 7.2 (1.4) <sup>bde</sup> | 4 | 0.90 (1.6) <sup>b</sup> | 12 | 3.6 (3.0) <sup>fg</sup> |
| harvesting<br>automatic clod removal | 4                                   | 3.6 (1.7) <sup>d</sup>   | 8 | 0.59 (4.9)              | 12 | 1.1 (4.8) <sup>f</sup>  |
| wrapping                             | 6                                   | 3.3 (2.1) <sup>ce</sup>  | 6 | 0.35 (5.5) <sup>c</sup> | 12 | 1.1 (5.5) <sup>g</sup>  |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b c d e f g</sup>  $p < 0.05$  (Mann-Whitney test for two groups)

From Table 4.2 it is easy to see, that re-entry time had a large influence on dermal exposure. Working after a short re-entry interval resulted in an exposure that is about a factor 6 to 9 higher than after a long re-entry interval (significant for two groups of workers). Also the variation in exposure level was much less after short re-entry. Differences in level of exposure among tasks are more evident after short re-entry. Dermal exposure during manual harvesting was significantly higher than during both automatic harvesting and wrapping. Dermal exposure during automatic harvesting was of the same level as during wrapping. Differences were also found in exposure of each hand. The right hand was usually more contaminated with pesticide than the left hand. Most workers used their right hand for pulling out plants and their left hand for collecting the flowers into a bundle. In case of manual harvesting, clod breaking and removing leaves was usually done with the hand that was also used for pulling out plants.

The relation between dermal exposure and dislodgeable foliar residue (DFR) was studied by combining dermal exposure level of each hand with the (hand-specific) DFR of the contact zone where the most intensive contact was observed. Resulting combinations are shown in Figure 4.1. A distinction is made between hands used for clod-breaking and/or removing leaves and hands used for other actions. Comparing these two groups of hands, it appears that the hand used for both actions mentioned is higher exposed than the other

hand. However, it should be noted that there are no observations for manual harvesting after long re-entry, since these observations obtained in both pilots with different leaf sampling strategy, were excluded.

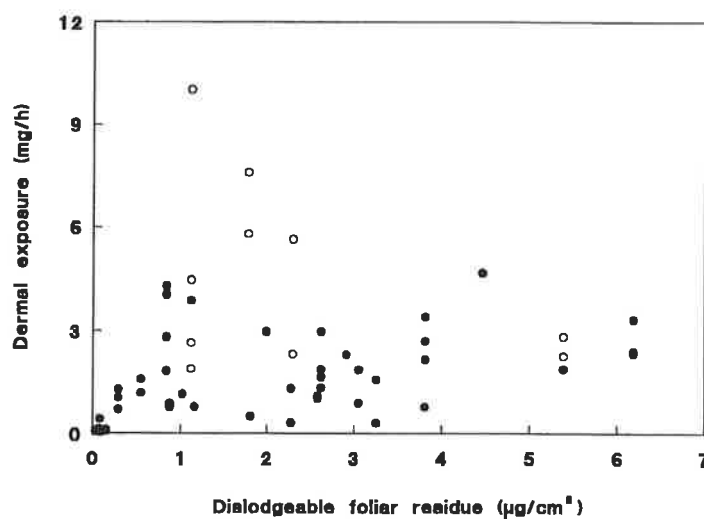


Figure 4.1 Association of dermal exposure during harvesting and dislodgeable foliar residue.  
 ○ observations for clod-breaking and/or removing leaves,  
 ● observations for other actions.

A constant fraction of dislodgeable foliar residue is supposed to be transferred to the skin or clothing of workers during harvesting activities, in which case, a transfer factor can be calculated as the ratio of dermal exposure and dislodgeable foliar residue (Popendorf and Leffingwell, 1982; Nigg et al., 1984; Zweig et al., 1985). Transfer factors established in this way were approximately 800 cm<sup>2</sup>/h for wrapping, 1000 cm<sup>2</sup>/h for automatic harvesting and 1300 cm<sup>2</sup>/h for manual harvesting (Table 4.3).

Table 4.3 Transfer factors per task.

| Task                                 | n  | Transfer factor <sup>a</sup><br>(cm <sup>2</sup> /h) | 95% Confidence limits |                  |
|--------------------------------------|----|------------------------------------------------------|-----------------------|------------------|
|                                      |    |                                                      | LCL <sup>b</sup>      | UCL <sup>b</sup> |
| harvesting<br>manual clod breaking   | 16 | 1335 (2.6)                                           | 801                   | 2226             |
| harvesting<br>automatic clod removal | 24 | 1032 (2.3)                                           | 729                   | 1462             |
| wrapping                             | 20 | 832 (3.2)                                            | 483                   | 1433             |
| Total                                | 60 | 1029 (2.7)                                           | 798                   | 1327             |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> LCL = lower confidence limit; UCL = upper confidence limit

In order to investigate to what extent dermal exposure is related to dislodgeable foliar residue (DFR), regression analysis was carried out. Linear regression analysis with dermal exposure as dependent (y) and DFR as independent variable (x), describing a simple linear model ( $y = a \cdot x$ ), provides a regression coefficient ( $a$ ) that can also be used directly as a measure of the transfer factor (Brouwer et al., 1991<sup>ab</sup>). In our case, log-log regression analysis was carried out, since using log-transformed data resulted in a far better regression model than when using non-transformed data. However, in this kind of model ( $\ln(y) = \beta \cdot \ln(x)$  or  $y = x^\beta$ ), the regression coefficient ( $\beta$ ) has a totally different meaning and cannot be used as a measure of the transfer factor! This log-log regression model may imply, that the transfer factor is dependent on dislodgeable foliar residue. So the association depicted in Figure 4.1 may in fact not entirely, i.e. over the total range of DFR, be linear.

In Figure 4.2 the transfer factor (calculated as ratio, see above) is plotted against DFR. Again, a distinction is made between observations for clod-breaking and/or removing leaves and observations for other activities. Comparing these two groups, it appears that for similar values of DFR, transfer during clod-breaking and/or removing leaves is higher than during other activities. Further, it appears that transfer decreases with increasing DFR and becomes relatively constant for  $DFR > 2 \mu\text{g}/\text{cm}^2$ . For low values of DFR, the transfer factor has a very large variation.

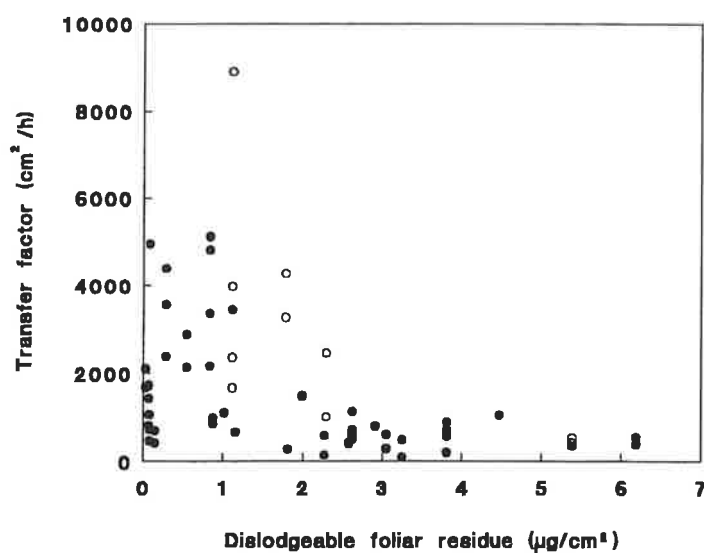


Figure 4.2 Transfer of chlorothalonil during harvesting in relation to dislodgeable foliar residue.  
 ○ observations for clod-breaking and/or removing leaves,  
 ● observations for other actions.

Results from the log-log regression analysis are presented in Table 4.4. The regression equation with DFR as independent variable explains about 60% of the variation found in dermal exposure during harvesting activities. Results of separate regression analyses for different tasks and re-entry intervals are also presented in Table 4.4. For manual harvesting and for short re-entry no significant association was found.

Table 4.4 Results of log-log regression of dermal exposure (mg/h) of hands and forearms during harvesting activities and dislodgeable foliar residue ( $\mu\text{g}/\text{cm}^2$ ).

| Task / re-entry                                 | n  | Regression<br>coefficient <sup>a</sup> | 95% Confidence limits<br>LCL <sup>b</sup> UCL <sup>b</sup> |      | R <sup>2</sup> <sub>adj</sub> <sup>c</sup> |
|-------------------------------------------------|----|----------------------------------------|------------------------------------------------------------|------|--------------------------------------------|
| <b>Task:</b>                                    |    |                                        |                                                            |      |                                            |
| harvesting <sup>d</sup><br>manual clod breaking | 16 | - 0.33 (0.19)                          | - 0.74                                                     | 0.08 | 0.12                                       |
| harvesting<br>automatic clod removal            | 24 | 0.86 (0.11)                            | 0.63                                                       | 1.08 | 0.72                                       |
| wrapping                                        | 20 | 0.56 (0.14)                            | 0.26                                                       | 0.85 | 0.44                                       |
| <b>Re-entry time:</b>                           |    |                                        |                                                            |      |                                            |
| short re-entry <sup>d</sup>                     | 36 | -0.09 (0.19)                           | - 0.48                                                     | 0.30 | 0.00                                       |
| long re-entry                                   | 24 | 0.81 (0.09)                            | 0.62                                                       | 1.00 | 0.78                                       |

<sup>a</sup> regression coefficient and standard error of estimate in parentheses

<sup>b</sup> LCL = lower confidence limit; UCL = upper confidence limit

<sup>c</sup> the amount of variation in the dependent variable explained by the fitted regression equation and adjusted for sample size (n)

<sup>d</sup> regression relation not significant

If the pilots were included in the regression analysis, neglecting the difference in sampling strategy, a relation would be found between dermal exposure and DFR for manual harvesting. The variance explained by the equation would only be 22%. The model for wrapping would improve to 58% explained variance with a regression coefficient of 0.78.

Work rate as determined had no significant effect on dermal exposure level.

## 4.2 Dislodgeable foliar residue

In four greenhouses, the course in time of DFR throughout the growing period has been followed by measuring DFR before and after each application with chlorothalonil. Data on the applications are presented in Table 4.5. In general six applications with the same application rate for each greenhouse were done until harvest.

Table 4.5 Data on the applications in the four greenhouses.

| Greenhouse | Application                         | Formulation | Application rate<br>(g a.i./1000 m <sup>2</sup> ) <sup>a</sup> |
|------------|-------------------------------------|-------------|----------------------------------------------------------------|
| 1          | all                                 | liquid      | 66                                                             |
| 2          | all                                 | liquid      | 72                                                             |
| 3          | 1 <sup>st</sup> and 5 <sup>th</sup> | liquid      | 83                                                             |
|            | 2 <sup>d</sup> - 4 <sup>th</sup>    | powder      | 124                                                            |
| 4          | all                                 | liquid      | 50                                                             |

<sup>a</sup> a.i. = active ingredient

In Figure 4.3 the relation is illustrated between the amount of chlorothalonil that results in foliar deposition after application and the Leaf Area Index (LAI) as measure of crop density. For each application in all four greenhouses the percentage of applied chlorothalonil that was intercepted by the crop was calculated. This was done by extrapolation of the DFR from the sampled leaves to the measured leaf area of all zones. The increase in the total amount of chlorothalonil due to the application was calculated. This increase per total leaf surface of the 12 sampled plants was converted to an increase per ground area by correcting for the number of plants per ground area. Finally, this increase in DFR per ground area has been expressed as percentage of the applied dose.

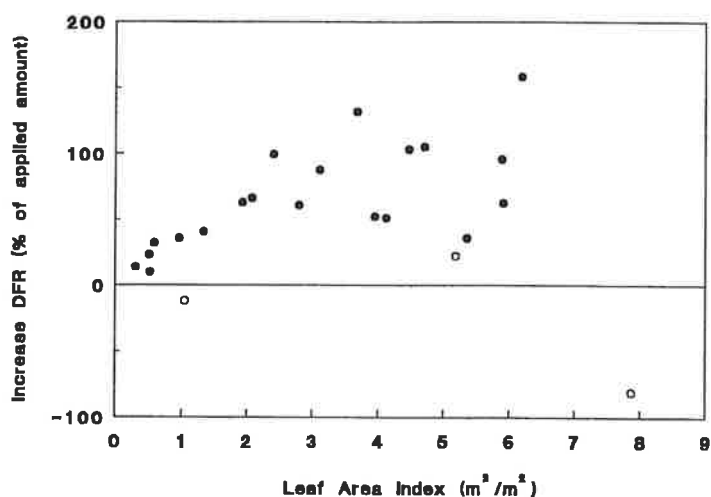


Figure 4.3 The percentage of applied chlorothalonil intercepted by the crop after application, in relation to the Leaf Area Index.

- observations included in the regression analysis (if LAI ≤ 5)
- observations excluded from the regression analysis (see Figure 4.4)

In order to investigate to what extent the percentage of applied pesticide intercepted by the crop is related to the Leaf Area Index of the crop at the time of application, linear regression was carried out. It is expected, that at the end of the growing period the percentage of applied pesticide intercepted by the crop will reach a plateau of (theoretically) 100% interception. Only in the beginning of the growing period, a linear relation will exist. From the data it can be concluded that a plateau is reached at a LAI of approximately 5. Some other points have been excluded from the analysis as well, because either rain irrigation had taken place between the application and the measurement of DFR after the application or because of the use of a pesticide formulation in which the amount of active ingredient was reduced sharply. The regression line is shown in Figure 4.4. The corresponding regression equation (for  $LAI \leq 5$ ) reads as follows:  
 increase in DFR as percentage of the applied dose =  $18 * LAI + 19$  ( $n = 16$ ;  $R^2 = 0.60$ ).

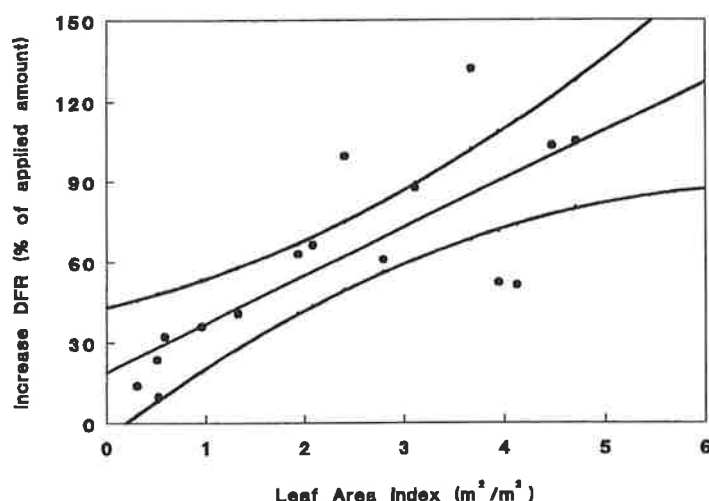


Figure 4.4 Association of the percentage of applied chlorothalonil intercepted by the crop after application and the Leaf Area Index, with the 95% confidence interval.

The course in time of DFR per zone is illustrated in Figure 4.5 for one greenhouse. In the course of time total absolute DFR on the crop increases. This increase is largest in the zones high and middle. However, not all four greenhouses gave exactly the same picture (annex 4). In general it is shown that after an application DFR is increased and it decreases again between applications. This is in agreement with the observed difference between dislodgeable foliar residue at harvest after short re-entry and long re-entry time (Table 4.6).



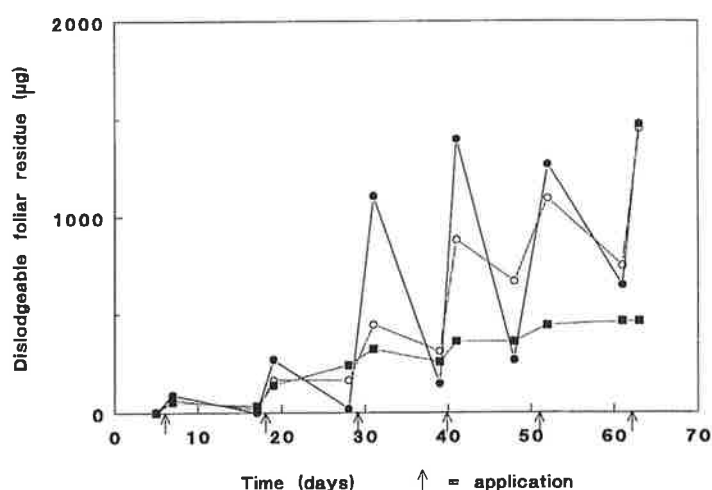


Figure 4.5 The course in time of dislodgeable foliar residue throughout the growing period in the zones high, middle and low as a result of six high-volume spraying applications with chlorothalonil. Data from one greenhouse.  
 — ● — High zone — ○ — Middle zone — ■ — Low zone

Table 4.6 Dislodgeable foliar residue (DFR) after short and long re-entry time and application rate.

| Re-entry       | n  | Hand-specific <sup>a</sup><br>DFR ( $\mu\text{g}/\text{cm}^2$ ) | n | Application rate <sup>b</sup><br>(g a.i./1000 m <sup>2</sup> ) |
|----------------|----|-----------------------------------------------------------------|---|----------------------------------------------------------------|
| short re-entry | 36 | 2.2 (2.1) <sup>c</sup>                                          | 6 | 112 (83)                                                       |
| long re-entry  | 24 | 0.30 (4.9) <sup>c</sup>                                         | 4 | 130 (104)                                                      |

<sup>a</sup> expressed as geometric mean (GM) of the DFR levels assigned to each hand and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> of last application before harvest, expressed as arithmetic mean and standard deviation in parentheses; a.i. = active ingredient

<sup>c</sup>  $p < 0.05$  (Mann-Whitney test for two groups)

### 4.3 Respiratory exposure after re-entry

The respiratory exposure of 26 workers to chlorothalonil was measured during harvesting of chrysanthemums in 12 greenhouses. It appeared that exposure during harvesting nor wrapping differed between both harvesting techniques (annex 3), so only two tasks were distinguished.

Table 4.7 shows that re-entry time had a significant influence on respiratory exposure of about a factor 3 to 8. Again, differences in level of exposure between tasks were more evident after a short re-entry interval. Respiratory exposure during harvesting was higher than during wrapping after short re-entry time.

Table 4.7 Respiratory exposure to chlorothalonil after re-entry.

| Task       | Respiratory exposure (mg/m <sup>3</sup> ) <sup>a</sup> |                           |    |                           |
|------------|--------------------------------------------------------|---------------------------|----|---------------------------|
|            | n                                                      | short re-entry            | n  | long re-entry             |
| harvesting | 6                                                      | 0.024 (1.6) <sup>b</sup>  | 80 | 0.0030 (2.9) <sup>b</sup> |
| wrapping   | 6                                                      | 0.0082 (2.1) <sup>c</sup> | 60 | 0.0024 (2.4) <sup>c</sup> |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b c</sup> p < 0.05 (Mann-Whitney test for two groups)

### 4.4 Quality control

All blank samples were below the detection limit.

The mean background level of chlorothalonil in air was 0.5 µg/m<sup>3</sup> (n = 9, GSD = 1.8), only just detectable. This was 9% of the mean personal respiratory exposure level found. In all but one greenhouse the difference between the two spiked gloves was 10% or less (not significant). In most cases the glove that was laid down contained less chlorothalonil as the glove that was stored directly. Important factors may be temperature, relative humidity and sampling spot. There is no information about these factors. The difference with a factor two between both spikes in one greenhouse remains unexplained.

## 5 DISCUSSION

### 5.1 Exposure after re-entry

The levels of dermal exposure during harvesting of chrysanthemums grown in greenhouses ranged from on average 1.1 mg/h for both automatic harvesting and wrapping to 3.6 mg/h for manual harvesting. Considering differences in crop, pesticides, application rates and techniques, and activities, these levels are in the range found by Brouwer et al. (1992<sup>ac</sup>) in harvesting of roses and in cutting of carnations grown in greenhouses, and by Davis et al. (1983) in apple thinning, all using the same method of assessment. Respiratory exposure (adjusted to mg/h) contributed less than 1% to total external exposure to chlorothalonil after re-entry. A similar proportion was found during harvesting of carnations (Brouwer et al., 1992<sup>a</sup>).

On average however, the levels of exposure during harvesting of chrysanthemums are somewhat lower than observed for cutting of carnations after spraying the same pesticide. This may be due to the difference in application rate of the last application before harvest and the resulting initial foliar residue. Average observed application rates in the chrysanthemum and carnation culture were respectively 119 g a.i./1000 m<sup>2</sup> ( $\pm$  85) and 261 g a.i./1000 m<sup>2</sup> ( $\pm$  124; n=9) (Brouwer, 1994). Further, the DFR in chrysanthemums was found to be lower than in carnations, which may be due to lower initial residues after application and to the decline of foliar residues. Where re-entry intervals in the carnation culture were 1-2 days, they were 2-8 weeks in the chrysanthemum culture. The importance of re-entry time is emphasized by the finding in this study that levels of both dermal and respiratory exposure after relatively short re-entry time (11-17 days) were considerably higher than after long re-entry time (32-56 days). Thirdly, there are differences in the harvesting method: carnations are cut using a knife while chrysanthemums are pulled out off the ground and clods are broken, the latter leading to more contact with the crop, on the basis of which one would expect higher exposure levels.

The variance in dermal exposure during harvesting of chrysanthemums can be explained for 60% by differences in dislodgeable foliar residue (DFR). This association has been found for automatic harvesting (72%) and wrapping (44%), but could not be established

for manual harvesting. This may have several reasons. Because of the decision not to use the data of the pilot studies in the regression analysis, there were no observations left for manual harvesting after long re-entry time. This resulted in a rather small variation, especially on a log scale, in both DFR and dermal exposure level, both variables having only about a factor 5 between the highest and lowest value. For wrapping and automatic harvesting, these factors were 15-20 times higher. Secondly, it is important to realize that the action 'clod breaking', in which the manual harvesting technique distinguishes itself from automatic harvesting, is an additional action, which is usually done with the hand that is also used for pulling out plants. To understand the meaning of this, levels of dermal exposure per hand used for a specific action, are summarized in Table 5.1. It is assumed that exposure during specific actions is independent of harvesting technique. Mean exposure is estimated of (i) a single action, i.e. of all hands that are solely used for the action mentioned, (ii) among others this action, i.e. of all hands that are used for this action and for any additional actions, and (iii) any but this action, i.e. of all hands that are not involved with the action mentioned.

Table 5.1 Hand-specific dermal exposure per action.

| Action                                   | Dermal exposure (mg/h) |            |                          |            |                     |            |
|------------------------------------------|------------------------|------------|--------------------------|------------|---------------------|------------|
|                                          | single action          |            | among others this action |            | any but this action |            |
|                                          | n                      | GM (GSD)   | n                        | GM (GSD)   | n                   | GM (GSD)   |
| <b>Harvesters:</b>                       |                        |            |                          |            |                     |            |
| pulling                                  | 12                     | 0.71 (5.9) | 25                       | 1.2 (4.6)  | 23                  | 0.53 (5.3) |
| bundling                                 | 20                     | 0.43 (5.3) | 25                       | 0.55 (4.9) | 23                  | 1.3 (4.9)  |
| clod breaking and/<br>or removing leaves | <sup>a</sup>           |            | 14                       | 2.4 (2.7)  | 34                  | 0.53 (5.2) |
| <b>Wrappers:</b>                         |                        |            |                          |            |                     |            |
| holding                                  | 12                     | 0.62 (6.2) | 14                       | 0.71 (5.7) | 10                  | 0.30 (5.2) |
| wrapping                                 | 10                     | 0.30 (5.2) | 12                       | 0.40 (5.3) | 12                  | 0.62 (6.2) |

<sup>a</sup> does not occur

From Table 5.1 it appears, that in general, for harvesters pulling results in higher exposure than bundling and for wrappers holding results in higher exposure than the actual wrapping. Clod breaking and/or removing leaves as one of the actions results in the highest exposure, as expected. For the regression analyses per task, combinations of dermal exposure and DFR are rather straightforward for automatic harvesting and wrapping, since generally each hand performs one action. In case of clod breaking/removing leaves however, one hand performs several actions and hence has contact with several zones in

the plant. A choice was made to take the DFR of the zone where most intensive contact was observed, being clod breaking/removing.

Transfer factors (expressed as GM) established in this study were approximately 800 cm<sup>2</sup>/h for wrapping, 1000 cm<sup>2</sup>/h for automatic harvesting and 1300 cm<sup>2</sup>/h for manual harvesting. These are of the same order of magnitude as those for harvesting peaches, citrus and grapes (Popendorf and Leffingwell, 1982) who calculated factors of 2000-5000 cm<sup>2</sup>/h (GM) on the basis of a whole body technique for measuring dermal exposure. Brouwer et al. (1992<sup>ac</sup>) found similar transfer factors for harvesting roses and carnations.

Since there is a substantial, though not significant, difference in number of flowers handled per hour between both harvesting techniques, it may be meaningful to express transfer factors per number of flowers instead of per hour. This results in a significant difference between transfer during harvesting and during wrapping, as wrappers handle twice as many flowers per hour as harvesters. Further, overlap in 95% confidence interval of both harvesting methods is a little smaller (annex 5).

The variation in the task specific transfer factors is relatively high, considering there are in principle no differences in crop, pesticide, application rate and technique, but only differences in activities. This variation may be explained partly by the fact that each task is comprised of several actions with different intensities of contact. Therefore it was attempted to estimate transfer factors for separate actions. Observations were divided into five groups of main actions based on the estimated level of exposure due to this action. In this classification, actions with higher exposure dominated over actions with lower exposure (Table 5.2).

Table 5.2 Transfer factors per action.

| Action                                   | n  | Transfer factor <sup>a</sup><br>(cm <sup>2</sup> /h) | 95% Confidence limits |                  |
|------------------------------------------|----|------------------------------------------------------|-----------------------|------------------|
|                                          |    |                                                      | LCL <sup>b</sup>      | UCL <sup>b</sup> |
| <b>Harvesters:</b>                       |    |                                                      |                       |                  |
| pulling                                  | 14 | 1165 (2.2)                                           | 732                   | 1854             |
| bundling                                 | 16 | 792 (2.1) <sup>c</sup>                               | 536                   | 1170             |
| clod breaking and/<br>or removing leaves | 10 | 2008 (2.6) <sup>c</sup>                              | 1011                  | 3987             |
| <b>Wrappers:</b>                         |    |                                                      |                       |                  |
| holding                                  | 12 | 1174 (2.4)                                           | 666                   | 2070             |
| wrapping                                 | 8  | 496 (4.0)                                            | 157                   | 1568             |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> LCL = lower confidence limit; UCL = upper confidence limit

<sup>c</sup> p < 0.05 (Mann-Whitney test for two groups)

From Table 5.2 it appears that clod breaking/removing leaves results in the highest transfer. The results from Table 5.2 are in agreement with the transfer factors per task from Table 4.4, assuming that comprising actions take an equal amount of time performing the task.

If transfer is expressed per number of flowers, overlap in 95% confidence intervals becomes a little smaller, resulting in a significant difference in transfer between clod breaking/removing leaves and pulling (annex 5).

Figure 4.2 showed that for low values of DFR, the transfer factor decreases with increasing dislodgeable foliar residue. This may, at least partly, be explained by the inaccuracy of the calculated transfer factor, resulting from low values of DFR of which some were below the limit of detection ( $n = 10$ ). Otherwise, transfer of chlorothalonil from chrysanthemum leaves to hands of workers may seem more efficient when the available amount of pesticide on the leaves is small. This apparent effect however, may be explained, if the proportion of available foliar residue that can be dislodged from the leaves by the current DFR procedure, is not constant but decreases when residue ages (McEwen et al., 1980). For equal dermal exposure levels, this would result in artificially extra high transfer factors in case of relatively old residues. In this case, DFR would not be a constant measure of available residue.

Furthermore, the transfer factor appears to be (negatively) affected by re-entry-time in this study, for when the regression model with dermal exposure as dependent and DFR as explaining variable is adjusted for re-entry time, a decrease is found in the regression coefficients (not shown). Research by McEwen and co-workers in which parathion was applied to mature apple trees (1980) provided indications to this latter effect on transfer, namely that the proportion of residue transferred from the foliage to the worker's hands is less as residue ages.

In general, variation in transfer factors is high, indicating other influences like differences in work practice and work rate and probably re-entry time, besides inaccuracy due to low values of DFR. So the transfer of pesticide from crop to worker may be an important determinant of dermal exposure, but is not easily captured in a single figure for complex situations.

Further research is recommended for the variables DFR and the transfer of pesticide from leaves to hands, e.g. concerning the effect of re-entry time on these variables.

## **5.2 The relation between the increase in DFR after application and the growing phase of the crop**

The amount of pesticide residue on the leaves is a determinant of exposure. The initial residue is, among others, determined by application rate and degree of interception of the pesticide by the crop. The relation that is found between Leaf Area Index and the percentage of the applied dose that is intercepted by the crop, for a high volume spraying application of chlorothalonil in the cultivation of chrysanthemums, is as follows:

$$\text{increase in DFR as percentage of the applied dose} = 18 * \text{LAI} + 19 \text{ (for LAI} \leq 5\text{)}.$$

This means that Leaf Area Index affects the degree of interception which can be quantified. Willis and McDowell (1987) found that an average of 62% ( $\pm 27\%$ ) of the applied dose is intercepted by the crop, taking together different crops, different growing phases and different pesticides. This value is in the range of the relation found in this study, but it is clear that the percentage may be considerably lower when LAI is small.

Performing the regression analysis that resulted in the relation mentioned above, three observations, which were obtained under different circumstances, were excluded to prevent a wrongly large influence on the relation. In case of two of these observations, rain irrigation had taken place between the application and the measurement of DFR after the application, respectively within 48 and 24 hours after application. From literature it appears that rain irrigation may affect pesticide residues. This effect is largest when rain occurs within 24 hours after application (Willis and McDowell, 1987). Other research shows that rain up to 48 hours after application may affect pesticide residues (Hartley and Graham-Bryce, 1980). In the two cases mentioned, a decrease or only a slight increase in DFR has been measured. The third excluded observation also showed a decrease in DFR, probably because of the use of a pesticide formulation in which the amount of active ingredient was reduced sharply.

It is to be expected, that the percentage of the applied dose intercepted by the crop will reach a plateau with increasing crop density. Therefore only observations with  $\text{LAI} \leq 5$  were included in the linear regression analysis.  $\text{LAI} \leq 5$  has been chosen on the basis of Figure 4.3. For some observations percentages over 100% were calculated. This may partly be due to the relatively large inaccuracy of the extrapolation of DFR of 12 leaves to

the leaf surface area of a total zone. Moreover between investigation variances of DFR sampling were observed (see Annex II) which may effect reliable extrapolations. The estimation of spray volume used per square metre is also liable to inaccuracy. Further, as crop density increases, pesticide application residues late in the growing-period will be less homogeneously spread over the crop compared to early in the growing-period, resulting in higher variation in interception late in the growing-period.

Using the Leaf Area Index, initial dislodgeable foliar residue, which affects exposure, can be predicted. This may be important for assessing health risks in pesticide approval procedures of other pesticides without additional exposure measurements. But the relation found between LAI and initial residue should first be validated and preferably extended to other crops, pesticides and application techniques. However, Leaf Area Index is rather cumbersome to measure and it requires the destruction of several plants. A parameter that may be used instead is plant height, which correlates very well with the total LAI for chrysanthemums. Linear regression showed that plant height together with the number of plants per ground area explain 94% of the variation in total LAI (annex 6).

### **5.3 Distribution of chlorothalonil in chrysanthemum plants.**

To predict pesticide exposure during crop activities, it is important to know the distribution of pesticide over the plant. The distribution appears to be different for each of four studied greenhouses. In general, DFR is increased after an application and it decreases again between applications. Mostly, the increase in DFR is largest in the highest zone, probably because of downward spraying. In the high zone, a sharp decrease in DFR between applications is observed. Assuming that plant growth is located mainly in the highest zone of the plant, the percentage of newly grown leaves (surface) in this zone between applications can be estimated from the increase in plant height. These percentages are shown in Table 5.3. It appears that just before a following application, leaves in the high zone are for the greater part new, especially early in the growing-period. So the decrease in DFR in the high zone may be due to the growing of new leaves. Further, DFR may decrease because of growth of existing leaves, degradation or evaporation of the pesticide, and/or rain irrigation.

In general, it can be concluded that the point of time of application affects the location



where the pesticide is deposited. At harvest, a pesticide that is applied early in the growing-period will mainly be found in the low zone of the plant, while a pesticide applied late in the growing-period will be found in the high zone of the plant. This may affect exposure during certain crop activities.

Table 5.3 Estimated percentages of newly grown leaves in the high zone of chrysanthemums.

| Between<br>application: | Percentage newly grown leaves (%) in four greenhouses |    |     |     |         |
|-------------------------|-------------------------------------------------------|----|-----|-----|---------|
|                         | 1                                                     | 2  | 3   | 4   | Average |
| 1 and 2                 | 100                                                   | 88 | 75  | 100 | 91      |
| 2 and 3                 | 100                                                   | 75 | 88  | 80  | 86      |
| 3 and 4                 | 75                                                    | 69 | 66  | 65  | 69      |
| 4 and 5                 | 24                                                    | 86 | 100 | 4   | 54      |
| 5 and 6                 | 30                                                    | 35 | -   | 7   | 18      |

## 6 CONCLUSIONS

- Re-entry time is an important determinant of exposure during crop activities. Levels of exposure during harvesting of chrysanthemums after a short re-entry interval are considerably higher than after a long re-entry interval.
- Levels of dermal exposure to chlorothalonil during harvesting of chrysanthemums are higher for harvesters than for wrappers. And dermal exposure during manual harvesting is higher than during automatic harvesting.
- Levels of respiratory exposure to chlorothalonil during harvesting of chrysanthemums are considerably lower than dermal exposure levels (less than 1%).
- Dermal exposure during harvesting depends on an average of about 60% on the amount of dislodgeable pesticide on the foliage (DFR), therefore it seems appropriate to use DFR for a first estimation of dermal exposure of workers after re-entry in greenhouses.
- A relation has been found between the Leaf Area Index and the percentage of the applied dose that is intercepted by the crop. For a high volume spraying application of chlorothalonil in the cultivation of chrysanthemums, this relation explains about 60% of the variation and reads as follows:  
increase in DFR as percentage of the applied dose =  $18 * LAI + 19$  (for  $LAI \leq 5$ ).  
On account of this relation, initial dislodgeable foliar residue can be predicted from application rate and Leaf Area Index for chrysanthemums.
- From studying the distribution of pesticide over three zones in the plant, it appeared that the point of time of application (related to the growing stage of the crop )affects the location where the pesticide is deposited. This may affect exposure as it is related to different crop activities such as harvesting.

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## **ANNEX 1 Hand-washing as method to estimate breakthrough of gloves**

In a pilot study in two greenhouses, the breakthrough of gloves was examined by means of a hand-washing procedure.

The hand-washing procedure was validated as follows. In a laboratory study with five volunteers 0.5 ml of a 1% homogeneous emulsion of Daconil (500 g chlorothalonil per litre) in water was added to the hands with a pipette. Another 0.5 ml of distilled water was added to the hands to rinse the pipette tip. After 15 minutes the volunteers washed both hands in their own way for 30 seconds in a polyethylene bag containing 500 ml of a 2-propanol/water mixture (60/40 v/v). The solubility of chlorothalonil residues in this mixture is approximately 100% (Jongen et al., 1991). This washing procedure was repeated with a second bag with a fresh mixture. After washing, the contents of each bag were homogenized by shaking, and approximately 10 ml from each bag was taken for analysis. These samples were extracted with n-hexane. Analytical recovery from the samples was 100%. Chlorothalonil was then quantified as described in § 3.6. The analytical limit of detection (LOD) of approximately 5 µg/l amounts to a limit of detection for the hand-washing procedure of 5 µg/l 2-propanol/water mixture. Stored at 4°C in the dark, chlorothalonil in the 2-propanol/water mixture was stable for at least 22 days and the between-day analytical coefficient of variation was less than 5%.

Control samples were taken for each volunteer by adding 0.5 ml 1% emulsion of Daconil (500 g chlorothalonil per litre) in water plus 0.5 ml of distilled water to 500 ml 2-propanol/water mixture. Results of these samples showed that on average 4.39 (± 0.19) mg was put on the hands.

The results for each person are illustrated in Figure 1. The effectiveness of washing, defined as the amount of pesticide washed off the hands divided by the net amount put on the hands and expressed as a percentage, ranged from 57% (person A) to 86% (person D). It should be noted that during adding pesticide to the hands of person A, some pesticide was spilt. The wash solution from the second washing contained 16 to 28% of the total amount recovered which was remarkably high. These percentages (of first and second washing) were similar to those found in the field.

On average the washing effectiveness of this hand-washing procedure was 74% (SD = 11%).

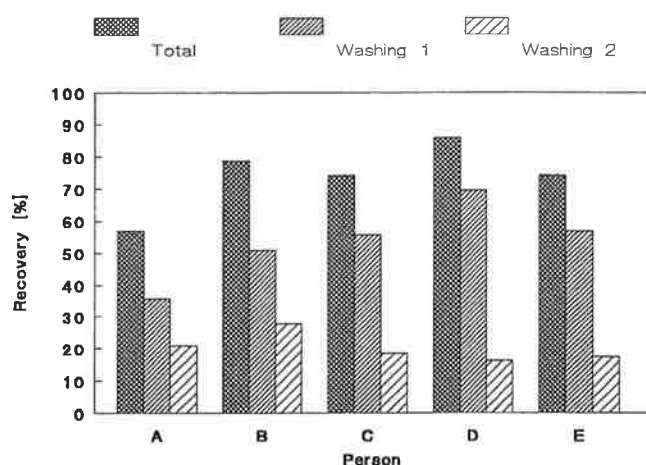


Figure 1 Effectiveness of washing for chlorothalonil

To estimate the breakthrough of gloves in the field, dermal exposure of all workers was measured as usual by means of gloves as described in § 3.2. However, before the workers put on the gloves, they washed their hands by the hand-washing procedure to start with clean hands. After the work period, they put off the gloves and washed their hands again. The latter washing contained the amount of pesticide broken through the gloves. The results are summarized in Table 1.

Table 1 Estimation of breakthrough of gloves

| Worker              | Hand washing (before) ( $\mu\text{g}$ ) | Perc. first washing (%) | Hand washing (after) ( $\mu\text{g}$ ) | Total <sup>a</sup> dermal exposure ( $\mu\text{g/h}$ ) | Break-through (%) <sup>c</sup> |
|---------------------|-----------------------------------------|-------------------------|----------------------------------------|--------------------------------------------------------|--------------------------------|
| <b>Greenhouse 1</b> |                                         |                         |                                        |                                                        |                                |
| harvester 1         | 172                                     | 85                      | 48                                     | 1800                                                   | 3.6                            |
| harvester 2         | 81                                      | 88                      | 19                                     | 610                                                    | 4.2                            |
| wrapper             | 25                                      | 88                      | 8 <sup>b</sup>                         | 41                                                     | <sup>d</sup>                   |
| <b>Greenhouse 2</b> |                                         |                         |                                        |                                                        |                                |
| harvester 1         | 28                                      | 79                      | 11 <sup>b</sup>                        | 707                                                    | 2.1                            |
| harvester 2         | 76                                      | 78                      | 23                                     | 864                                                    | 3.6                            |
| wrapper             | 67                                      | 79                      | 15 <sup>b</sup>                        | 100                                                    | <sup>d</sup>                   |

<sup>a</sup> of both hands together

<sup>b</sup> value near limit of detection

<sup>c</sup> assuming a washing effectiveness of 74%

<sup>d</sup> not relevant because of low values



In Table 2 the average breakthrough (n = 4) is calculated assuming a washing effectiveness ranging from 100 to 25%. An effectiveness of 74% results in a breakthrough that is less than 5%, which is acceptable. If the true washing effectiveness was only 50%, then average breakthrough would still be about 5%.

Table 2 Calculated breakthrough, assuming several values for washing effectiveness (n = 4)

| Washing effectiveness<br>(%) | Average breakthrough |     |
|------------------------------|----------------------|-----|
|                              | (%)                  | SD  |
| 100                          | 2.5                  | 0.7 |
| 74                           | 3.4                  | 0.9 |
| 50                           | 5.0                  | 1.3 |
| 40                           | 6.2                  | 1.7 |
| 25                           | 10                   | 3   |

## ANNEX 2 The precision of the assessment of dislodgeable foliar residue

To assess the precision of the assessment of dislodgeable foliar residue, a total of 10 test samples was taken by two researchers in one greenhouse. Each researcher took 5 samples of 12 leaves from the flowers in the sector that was going to be harvested that day. All samples were taken from the zone about 47 cm above the ground. Results are shown in Table 1. The precision is expressed as coefficient of variation ( $CV_{total}$ ) which is calculated as follows:  $CV_{total} = (SD/average\ DFR) * 100\%$ .

Table 1 Average dislodgeable foliar residue (DFR), standard deviation (SD) and coefficients of variation ( $CV_{total}$ ) of the test samples.

| Researcher | Sample number | DFR ( $\mu g/cm^2$ ) | Average DFR <sup>a</sup> | SD   | $CV_{total}$ (%) |
|------------|---------------|----------------------|--------------------------|------|------------------|
| 1          | 1             | 3.74                 | 2.36                     | 0.83 | 35               |
|            | 2             | 2.49                 |                          |      |                  |
|            | 3             | 1.83                 |                          |      |                  |
|            | 4             | 2.06                 |                          |      |                  |
|            | 5             | 1.67                 |                          |      |                  |
| 2          | 6             | 2.04                 | 1.64                     | 0.32 | 20               |
|            | 7             | 1.66                 |                          |      |                  |
|            | 8             | 1.69                 |                          |      |                  |
|            | 9             | 1.14                 |                          |      |                  |
|            | 10            | 1.68                 |                          |      |                  |

<sup>a</sup> difference between researchers is significant,  $p < 0.05$  (Mann-Whitney test)

### ANNEX 3 Classification of workers into groups

From Table 1 it can be seen that dermal exposure during wrapping does not differ between both harvesting techniques while dermal exposure during harvesting does. Therefore three groups of workers are distinguished in the analysis, namely harvesters using the different harvesting techniques and wrappers. The high variation is mainly due to the influence of re-entry time on exposure. Stratification for short and long re-entry time would in principle result in lower variation.

Table 1 Dermal exposure of both hands and forearms to chlorothalonil.

| Harvesting technique   | Dermal exposure (mg/h) <sup>a</sup> |                         |   |           |
|------------------------|-------------------------------------|-------------------------|---|-----------|
|                        | n                                   | harvesting <sup>b</sup> | n | wrapping  |
| manual clod breaking   | 12                                  | 3.6 (3.0)               | 6 | 1.0 (9.1) |
| automatic clod removal | 12                                  | 1.1 (4.8)               | 6 | 1.2 (3.4) |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> significant difference in this task between harvesting techniques (Mann-Whitney test  $p < 0.05$ )

In Table 2 it can be seen that a similar difference between harvesting techniques does not exist for respiratory exposure. So for respiratory data only two groups are distinguished, namely harvesters using any harvesting technique and wrappers.

Table 2 Respiratory exposure to chlorothalonil after re-entry.

| Harvesting technique   | Respiratory exposure ( $\mu\text{g}/\text{m}^3$ ) <sup>a</sup> |            |   |           |
|------------------------|----------------------------------------------------------------|------------|---|-----------|
|                        | n                                                              | harvesting | n | wrapping  |
| manual clod breaking   | 8                                                              | 7.0 (4.8)  | 6 | 4.6 (3.0) |
| automatic clod removal | 6                                                              | 7.7 (3.1)  | 6 | 4.4 (2.7) |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

It can be concluded, that differences between two harvesting techniques studied or differences in intensity of contact with the chrysanthemum crop do affect dermal exposure to chlorothalonil, but do not affect respiratory exposure levels.

#### ANNEX 4 Distribution of chlorothalonil in chrysanthemum plants

In § 4.2 an example is shown of the course in time of dislodgeable foliar residue throughout the growing period in the zones high, middle and low as a result of six high-volume spraying applications for chlorothalonil. However, not all four greenhouses gave the same picture (Figures 1,2,3). In the course of time total absolute DFR on the crop increases. For greenhouse 1 (Figure 4.5 text) this increase is largest in the zones high and middle. In greenhouse 2 (Figure 1) and 4 (Figure 3) the increase is at the end of the growing period largest in the high zone. However, in greenhouse 3 (Figure 2) the increase is largest in the low zone. In general it is shown that after an application DFR is increased and it decreases again between applications.

The points of time when rain irrigation took place are only given in Figure 1, because it is unknown when exactly rain irrigation took place in the other greenhouses. At any rate no rain irrigation took place between application and the measurement of DFR after application in these greenhouses.

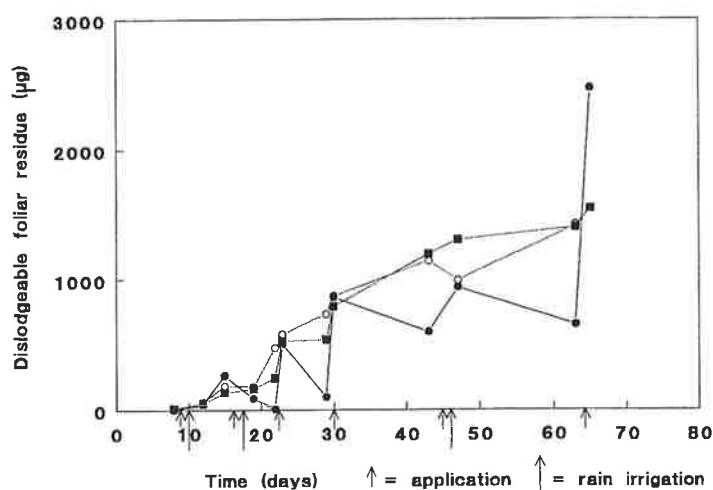


Figure 1 The course in time of dislodgeable foliar residue throughout the growing period in the zones high, middle and low in greenhouse 2.  
 — ● — High zone      — ○ — Middle zone      — ■ — Low zone

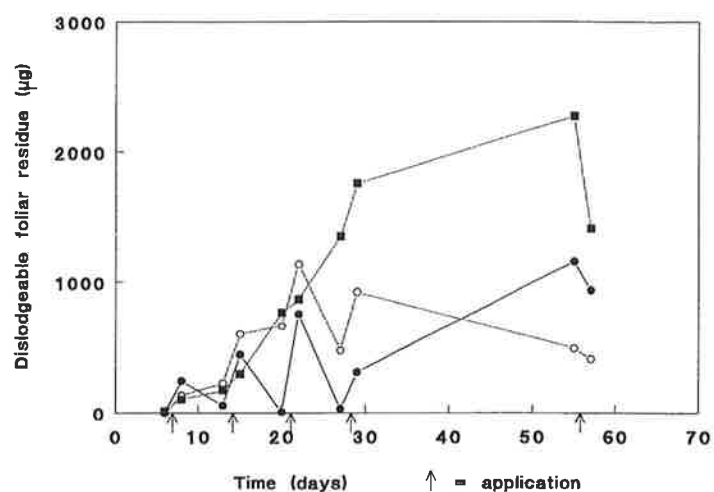


Figure 2 The course in time of dislodgeable foliar residue throughout the growing period in the zones high, middle and low in greenhouse 3.  
 — ● — High zone      — ○ — Middle zone      — ■ — Low zone

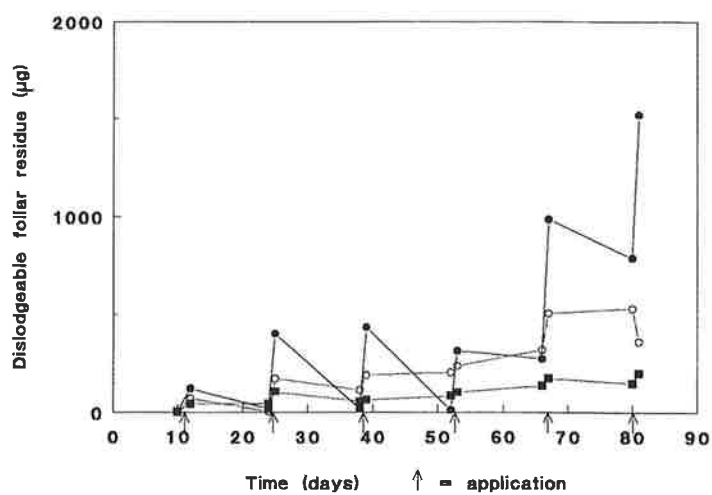


Figure 3 The course in time of dislodgeable foliar residue throughout the growing period in the zones high, middle and low in greenhouse 4.  
 — ● — High zone      — ○ — Middle zone      — ■ — Low zone

## ANNEX 5 How to express transfer factors

Transfer factors are usually determined in one of the following ways:

- 1) as ratio of dermal exposure and dislodgeable foliar residue (DFR) or
- 2) as regression coefficient in a linear model with dermal exposure as dependent and DFR as independent variable.

The dimensions used for dermal exposure and DFR are usually mg/h and  $\mu\text{g}/\text{cm}^2$  respectively, resulting in  $\text{cm}^2/\text{h}$  as dimension for the transfer factor.

In this study in the cultivation of chrysanthemums, two harvesting techniques are compared that differ in the way removing leaves and clods is carried out. In case of manual harvesting the worker removes the leaves and breaks the clods, whereas in case of automatic harvesting this is done by a machine. It appears that in greenhouses where clods were removed automatically, about 20% more flowers were harvested per hour compared to greenhouses where clods were broken by the workers. Further, wrappers generally handle twice as many flowers as harvesters do. Therefore it is meaningful to take this systematic difference in work rate into account in the transfer factor by dividing it by work rate. Because work rate was estimated on basis of the number of bunches of flowers harvested, transfer is expressed as  $\text{cm}^2/\text{bunch}$ , while each bunch contains five flowers. In Table 1, transfer factors per task are given and in Table 2 those per action (respectively comparable with Table 4.4 and 5.2).

Table 1 Transfer factors per task ( $\text{cm}^2/\text{bunch}$ ).

| Task                                 | n  | Transfer factor <sup>a</sup><br>( $\text{cm}^2/\text{bunch}$ ) | 95% Confidence limits |                  |
|--------------------------------------|----|----------------------------------------------------------------|-----------------------|------------------|
|                                      |    |                                                                | LCL <sup>b</sup>      | UCL <sup>b</sup> |
| harvesting<br>manual clod breaking   | 16 | 7.0 (2.4) <sup>c</sup>                                         | 4.4                   | 11.0             |
| harvesting<br>automatic clod removal | 24 | 4.2 (2.4) <sup>d</sup>                                         | 2.9                   | 6.1              |
| wrapping                             | 20 | 1.9 (3.5) <sup>cd</sup>                                        | 1.1                   | 3.4              |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> LCL = lower confidence limit; UCL = upper confidence limit

<sup>cd</sup>  $p < 0.05$  (Mann-Whitney test for two groups)

Table 2 Transfer factors per action (cm<sup>2</sup>/bunch).

| Action                                   | n  | Transfer factor <sup>a</sup><br>(cm <sup>2</sup> /bunch) | 95% Confidence limits<br>LCL <sup>b</sup> UCL <sup>b</sup> |      |
|------------------------------------------|----|----------------------------------------------------------|------------------------------------------------------------|------|
| <b>Harvesters:</b>                       |    |                                                          |                                                            |      |
| pulling                                  | 14 | 4.8 (2.2) <sup>c</sup>                                   | 3.0                                                        | 7.7  |
| bundling                                 | 16 | 3.4 (2.2) <sup>d</sup>                                   | 2.2                                                        | 5.3  |
| clod breaking and/<br>or removing leaves | 10 | 10.6 (2.2) <sup>cd</sup>                                 | 6.1                                                        | 18.5 |
| <b>Wrappers:</b>                         |    |                                                          |                                                            |      |
| holding                                  | 12 | 2.8 (2.6)                                                | 1.5                                                        | 5.0  |
| wrapping                                 | 8  | 1.1 (4.4)                                                | 0.31                                                       | 3.7  |

<sup>a</sup> expressed as geometric mean (GM) and geometric standard deviation (GSD, in parentheses)

<sup>b</sup> LCL = lower confidence limit; UCL = upper confidence limit

<sup>cd</sup> p < 0.05 (Mann-Whitney test for two groups)

In Figure 1 and 2 task and action specific transfer, expressed per hour and per bunch of flowers, are compared. Indicated error bars were obtained by dividing the difference of UCL and LCL by two. Expressing transfer per bunch of flowers results in a significant difference between transfer during harvesting and during wrapping, as wrappers handle twice as many flowers per hour as harvesters (Figure 1). Further, overlap in 95% confidence intervals is a little smaller, so differences between both harvesting methods and between separate actions become more clear or even significant (between clod breaking/removing leaves and pulling).

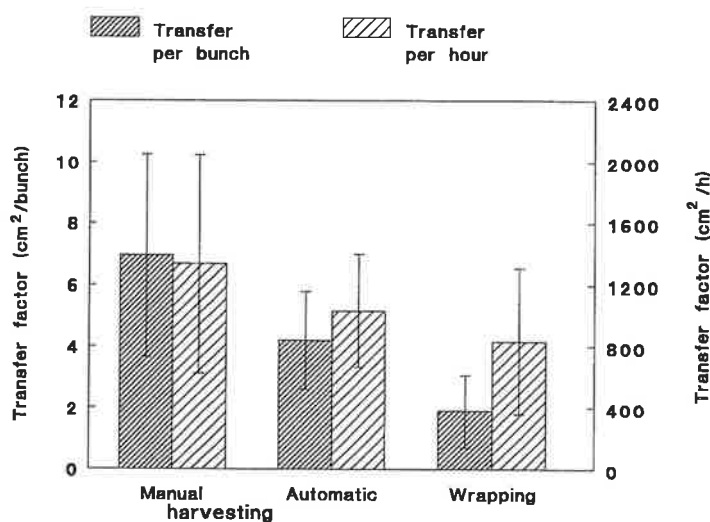


Figure 1 Task specific transfer factors, expressed per bunch (left) and per hour (right).

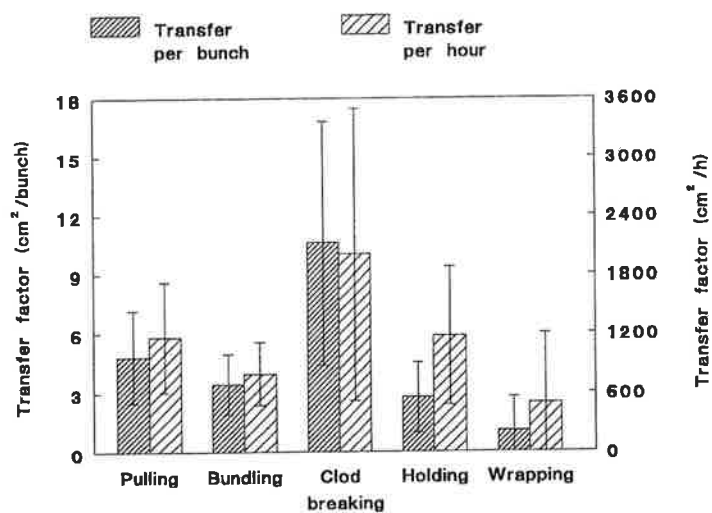


Figure 2 Action specific transfer factors, expressed per bunch (left) and per hour (right).



#### ANNEX 6 Correlation of Leaf Area Index and plant height in chrysanthemums.

The leaf area index (LAI) is a measure of crop density that indicates how many square metres leaf surface cover one square metre of ground. LAI depends on leaf growth, which is crop and probably season specific, and planting density, i.e. the number of plants that have been planted per ground area. In fall, chrysanthemums are usually planted wider apart because of less day-light.

Leaf area index is rather cumbersome to measure and it generally requires the destruction of several plants. Since leaf growth is generally correlated with plant height, this parameter may be used instead. Figure 1 illustrates the association of LAI and plant height. The relation between LAI on the one hand and plant height and planting density on the other hand has been studied by linear regression analysis on data from four greenhouses. The relation is linear from a height of 10 cm onwards and for planting densities ranging from 45 to 57 plants per square metre. The corresponding regression equation reads as follows:  $LAI = 0.076 * \text{plant height} + 0.086 * \text{plant density} - 5$  ( $n = 46$ ).

This equation explains 94% of the variation in LAI. The 95% confidence intervals for the regression coefficients of the explaining variables are respectively 0.070-0.082 for plant height and 0.052-0.12 for plant density.

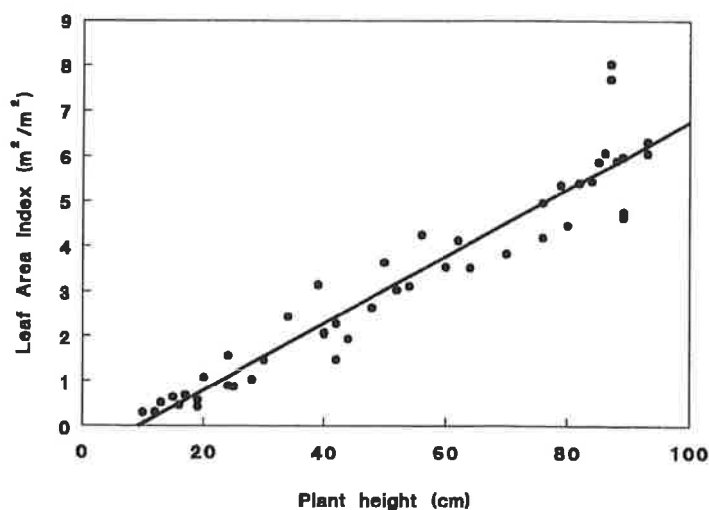


Figure 1 Association of Leaf Area Index and plant height in chrysanthemums.



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