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Exposure to pesticides

Part I. The cultivation of carnations in greenhouses

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Labour Inspectorate

Exposure to pesticides

Part I. The cultivation of carnations in greenhouses

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FOREWORD

By request of the Directorate-General of Labour of the Ministry of Social Affairs and Employment the TNO Medical Biological Laboratory investigates the health risks of exposure to chemical substances in the working environment. These investigations are performed in a framework called 'Health Hazard Surveys' (HHS). The present study report is the first part of a series on exposure to pesticides and concomittant health risk.
In the present survey the health risk of exposure to pesticides after re-entry (crop tasks after pesticide application) of the greenhouses with carnations which has been treated with pesticides is emphasized. It was intended to perform this survey in both carnation production farms and carnation nurseries, but because of lack of cooperation of the majority of the latter, the study was only performed in production farms.

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SUMMARY

On 18 farms for carnation culture in greenhouses in The Netherlands respiratory exposure and dermal exposure of hands and forearms to chlorothalonil, thiophanate-methyl, thiram and zineb during application and following crop activities have been measured. Farms have been selected by a 'worst case' strategy with regard to dermal exposure to pesticides during cutting of the flowers. For 95 workers the average dermal exposure during cutting was 10.6 mg/h (geometric mean; active ingredient) and during sorting/bundling of these flowers by 38 workers dermal exposure was 7.3 mg/h. Dermal exposure during cutting was strongly related to the amount of dislodgeable foliar residue (DFR). A measure for transfer of the pesticides from leaves to hands has been calculated of approximately 4500 cm^2/h. Differences have been noticed between the different pesticides. The DFR was mainly determined by the amount of pesticide remaining from previous applications and by the latest applied dose, and not by the application technique (spraying or dusting).
An average concentration in air of 0.07 mg/m^3 (personal air sampling) has been measured during cutting after a pesticide had been dusted.
For the same treated area spraying of a pesticide yielded a 7 times higher dermal exposure than during dusting. On the other hand during dusting respiratory exposure was significantly higher.
It is concluded from the observed levels of dermal exposure after re-entry of greenhouses that a health risk may occur, especially after application of relatively toxic pesticides with good skin-penetrating properties. The respiratory exposure to dusted pesticides after re-entry is as high as during application of these pesticides. In some situations this may also lead to health risks.

SAMENVATTING

BLOOTSTELLING AAN BESTRIJDINGSMIDDELEN IN DE ANJERTEELT IN KASSEN.

Rudi Brouwer, Derk H. Brouwer, Gerrit de Mik en Johannes J. van Hemmen

Blootstelling aan bestrijdingsmiddelen in de land- en tuinbouw vindt meestal plaats tijdens het zogenaamde mengen en laden (het klaarmaken van de spuit- of nevelvloeistof en het laden van de apparatuur), de toepassing van het middel en de werkzaamheden die na de toepassing in de betreffende ruimte of in het gewas worden uitgevoerd (re-entry). Uit de literatuur blijkt dat blootstelling aan bestrijdingsmiddelen kan leiden tot nadelige gezondheidseffekten. In Nederland worden in de glastuinbouw ongeveer 25000 mensen potentieel blootgesteld aan bestrijdingsmiddelen tijdens werkzaamheden in met deze middelen behandelde gewassen. Slechts een beperkt aantal personen wordt blootgesteld tijdens het mengen en laden en de feitelijke toepassing van de middelen. Geschat wordt dat de meest intensieve blootstelling plaatsvindt in de sierteelt onder glas, omdat nagenoeg het jaar rond dagelijks wordt geoogst en omdat strenge eisen worden gesteld voor wat betreft afwezigheid van ziekten en plagen in bloemen voor de export.

Het doel van het in dit rapport beschreven onderzoek was het bepalen van de blootstelling aan bestrijdingsmiddelen in de anjerteelt in kassen, met speciale aandacht voor de blootstelling van de huid tijdens werkzaamheden in gewas dat met bestrijdingsmiddelen is behandeld. Op basis van de mate van gebruik en de toxiciteit van de bestrijdingsmiddelen werden chloorthalonil, thiofanaat-methyl, thiram en zineb gekozen om de niveaus van blootstelling te bepalen.

De volgende vragen zouden met het onderzoek beantwoord moeten worden:

- Wat zijn de blootstellingsniveaus (via huid en ademhaling) voor de vier geselekteerde bestrijdingsmiddelen tijdens toepassing en na re-entry?
- Welke relatie bestaat er tussen de huidblootstelling en de hoeveelheid op het blad aanwezig bestrijdingsmiddel (afveegbaar residu)?
- Kunnen de verkregen resultaten voor de vier geselekteerde bestrijdingsmiddelen worden geëxtrapoleerd naar andere, niet onderzochte bestrijdingsmiddelen?
- Vormen de gemeten blootstellingsniveaus een risico met betrekking tot systemische gezondheidseffekten?
- Zijn er aanwijzingen voor effekten op de huid van handen en onderarmen door

blootstelling aan bestrijdingsmiddelen?

Hieronder worden de resultaten en de konklusies en aanbevelingen kort samengevat.

Op 18 anjerteeltbedrijven is de blootstelling van handen en onderarmen aan chloorthalonil, thiofanaat-methyl, thiram en zineb bepaald tijdens de toepassing van deze middelen (inklusief mengen en laden) en tijdens gewaswerkzaamheden. In een oriënterende studie kon geen blootstelling via de ademhalingswegen worden gemeten bij gewaswerkzaamheden na een spuittoepassing. Daarom werd de blootstelling via de ademhalingswegen tijdens gewaswerkzaamheden alleen gemeten nadat de middelen over het gewas verstoven waren en verder bij alle vormen van toepassing. Het onderzoek werd verricht op bedrijven die geselekteerd waren op basis van 'worst case' overwegingen, waarbij als uitgangspunt gold een hoge huidblootstelling tijdens het snijden van de anjers. Omdat het kontakt met het gewas intensiever is naarmate het gewas dichter en hoger is, zijn de metingen voornamelijk verricht op standaardanjerbedrijven in het tweede jaar van de teelt.

Van alle aanwezige werkers werd de blootstelling gemeten gedurende de eerste gewaswerkzaamheden - snijden en sorteren/bossen - na een toepassing van een pesticide. Tevens is de hoeveelheid bestrijdingsmiddel bepaald die tijdens het snijden op het gewas aanwezig was (afveegbaar residu).

De huidblootstelling die tijdens het snijden van anjers werd bepaald bij 95 werkers was gemiddeld 10,6 mg/uur (geometisch gemiddelde; aktieve stof; spreiding: 1-100 mg/uur). Tijdens het sorteren/bossen van deze bloemen werd bij 38 werkers een huidblootstelling gemeten van gemiddeld 7,3 mg/uur (figuur 1 en tabel 2). De hoeveelheid bestrijdingsmiddel die tijdens het snijden op de handen en onderarmen werd aangetroffen bleek samen te hangen met het afveegbaar residu. De variantie van de huidblootstelling tijdens het snijden kon voor 66% verklaard worden door de variatie in de hoeveelheid afveegbaar residu. Er kon een overdrachtsfaktor (d.i. een maat voor de overdracht van het bestrijdingsmiddel van gewas naar hand en onderarm) van ongeveer 4500 cm^2/uur worden berekend. De verschillen tussen de overdrachtsfaktoren voor de vier bestrijdingsmiddelen zijn relatief klein; zij varieerden van 2900 tot 10000 cm^2/uur (figuur 2 en tabel 3).

Het afveegbaar residu werd voornamelijk bepaald door de hoeveelheid bestrijdingsmiddel die nog aanwezig was van voorgaande toepassingen en door de laatst gebruikte dosering (gram aktieve stof/1000 m^2), maar niet of nauwelijks door de toepassings-

techniek (hoog-volume spuiten of stuiven) of de formulering (tabel 4). De gevonden verschillen in de huidblootstelling tijdens het snijden na spuiten of stuiven worden dan ook aan verschillen in de toegepaste dosering toegeschreven. De afhankelijkheid van het afveegbaar residu van eerdere toepassing(en) is een sterke aanwijzing voor ophoping van deze middelen op het blad. Chloorthalonil en thiofanaat-methyl blijven tot ongeveer 60 dagen na toepassing detekteerbaar (figuur 3). Dit betekent dat deze middelen na een toepassing binnen 60 dagen na de vorige waarschijnlijk op het gewas zullen ophopen.
De inhalatoire blootstelling (persoonlijke monstername) tijdens het snijden van anjers nadat een middel was verstoven, bedroeg gemiddeld 0,07 mg/m^3.
De blootstelling van de handen en onderarmen gedurende het bespuiten van een bepaald oppervlak was 7 maal hoger dan tijdens het bestuiven van eenzelfde oppervlak. Daar staat echter tegenover dat tijdens stuiven de inhalatoire blootstelling significant hoger was dan gedurende spuiten (tabel 5).
Op basis van de beschikbare gegevens over de toxiciteit en de huidresorptie van de vier onderzochte middelen is een 'indicatieve limietwaarde'afgeleid, d.i. de hoeveelheid bestrijdingsmiddel waarvan op grond van de huidige kennis mag worden aangenomen dat die geen negatieve gevolgen heeft voor de gezondheid. Aan de hand van de gemeten blootstellingsniveaus kan hiermee een schatting van het gezondheidsrisiko worden gemaakt.
Gezien de gemeten niveaus van huidblootstelling kan worden gesteld dat, tijdens gewaswerkzaamheden na toepassing van bestrijdingsmiddelen gezondheidseffekten kunnen optreden (tabel 7), vooral na toepassing van relatief toxische middelen met goed penetrerende eigenschappen in relatief hoge doseringen. De inhalatoire blootstelling aan verstoven bestrijdingsmiddelen gedurende gewaswerkzaamheden is vergelijkbaar met de inhalatoire blootstelling gedurende toepassing van deze middelen. In sommige situaties kan dit leiden tot gezondheidsproblemen. In het licht van deze resultaten worden maatregelen aanbevolen om de blootstelling te verlagen. De dermale blootstelling kan worden verminderd door te zorgen voor een geringer afveegbaar residu. Dit kan bereikt worden door het gebruik van bestrijdingsmiddelen te minimaliseren, d.w.z. door de (waargenomen te hoge) dosering en de frekwentie van toepassing te verlagen. Vanuit het oogpunt van inhalatoire blootstelling tijdens gewaswerkzaamheden wordt het verstuiven van middelen ontraden. Over het algemeen is de gewaswerker zich niet bewust van de blootstelling aan bestrijdingsmiddelen

gedurende een groot deel van de dag. De kennis van de gewaswerker t.a.v. de (toxicologische) eigenschappen van bestrijdingsmiddelen en de (persoonlijke) hygiëne die bij het werk (toepassing én gewaswerkzaamheden) in acht genomen moet worden, behoeft de nodige verbetering door training en scholing.

1 INTRODUCTION

Exposure to pesticides in agriculture occurs mainly during loading, mixing and application of pesticides and during manual activities in treated crops, usually referred to as re-entry. In greenhouses for flower culture nearly all workers are daily exposed to pesticides because of manual crop activities. A limited number of persons is involved in the actual application (generally one person per farm). 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.

Different tasks with frequent dermal contact with foliage treated with organophosphorous pesticides have led to incidents of illness among fieldworkers in citrus and peach crops in the USA (Gunther et al., 1977). Local effects such as contact dermatitis due to contact with pesticide residues have also been reported (Fregert, 1967; Ketel, 1976; Bruynzeel & Ketel, 1986). In several other studies a relationship between pesticide use and prevalence of skin disorders was found (Veien et al., 1980; Peachy, 1981; Willems et al., 1984). In 1970 the occurrence of incidents has led in the USA to regulations on fieldworker re-entry intervals for various organophosphorous and carbamate pesticides, as a way of reducing the exposure of workers to acceptable levels (Spear et al., 1977). Foliar residues and dermal exposure of fieldworkers have been determined in order to estimate safe levels of foliar residue (re-entry intervals) of, e.g., phosalone in apple orchards (Davis, et al., 1982) and peach orchards (Popendorf et al., 1979), of parathion (Spear et al., 1975; 1977) and chlorthiophos (Iwata et al., 1982) in citrus groves, of organophosphorous compounds in cotton (Burns & Parker, 1975), of maleic hydrazine in tobacco (Herman et al., 1985), and of captan, benomyl, carbaryl, methiocarb and vinchlozolin in strawberries and blueberries (Zweig et al., 1983; 1984; 1985). Several of these authors have tried to predict the dermal exposure of the worker by calculating the ratio of dermal exposure and the so-called dislodgeable foliar residue (DFR) (Popendorf & Leffingwell, 1982; Nigg et al., 1984; Zweig et al., 1985). They state that there is a relative constancy of this factor for a variety of crops and different pesticides. A constant fraction of dislodgeable foliar residues is transferred to the skin or clothing of workers during manual activities in treated fields (Zweig et al., 1985).

This transfer of the pesticide residue from the surface of the leaves to the skin of the worker is most important for dermal exposure (Popendorf & Leffingwell, 1982; Popendorf, 1985) 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 (DFR) depends on the formulation, application technique and the dose applied per unit surface, and on the frequency of use and the re-entry interval. The frequency and intensity of contact of the body with the treated crops is determined by the particular crop (density and height), by the frequency of manual activities and by worker-dependent factors such as work practice. Uptake of pesticides may be affected by personal hygiene (Popendorf & Spear, 1974).

As Morse et al. (1979) showed for commercial florists who were exposed to contaminated cut-flowers, and Liesivuori et al. (1988) for workers in greenhouses with roses exposed to dimethoate and benomyl, a potential health hazard exists for workers having prolonged and continuous contact with foliar surfaces of cut-flowers bearing pesticide residues.

In Dutch greenhouses a population of about 25,000 persons is potentially exposed to pesticides during application or after re-entry during crop activities. In flower culture pesticides are used more frequently and in greater amounts than in the cultivation of vegetables. Most flower species have to be harvested daily. This means that workers have to work in crops within several hours after treatment with pesticides. In the Dutch flower culture about 24,000 man-years are needed yearly for the production of cut-flowers and pot-plants. The economically most important cut-flowers are roses, chrysanthemums and carnations. These species have an annual turnover of more than a milliard guilders and account for approximately 40% of the total acreage for flower culture in greenhouses (CBS, 1990). The main production areas are found in the western part of the country. About 60% of the total acreage is found in the area between Rotterdam, Delft and The Hague, called Het Westland. Another 20% is found in the region around Aalsmeer.

The aim of the present study was to estimate the health risks due to exposure to pesticides in greenhouses for carnation culture, with special emphasis on dermal exposure during crop activities because dermal exposure after re-entry is considered to be the most important type of exposure. On the basis of toxicity, frequency of application and amounts used in carnation culture, the pesticides chlorothalonil, thiophanate-methyl, thiram and zineb were selected for the estimation of the level of exposure.

The following questions had to be answered:

- What are the levels of dermal and respiratory exposure to the four selected pesticides at application and after re-entry?
- What is the relationship between dermal exposure during the major manual crop activities and the amount of pesticide on the leaves (dislodgeable foliar residue)?
- Is it possible to extrapolate the results of the four selected pesticides to other pesticides?
- Can systemic health risks be expected at the observed exposure levels?
- Is there evidence for skin disorders on hands and forearms due to exposure to pesticides?

2 MATERIALS AND METHODS

2.1 Selection of farms

In 1988 the total acreage for carnation culture in The Netherlands was about 320 ha spread over 800 farms. About half of these farms used more than 0.25 ha for carnations and formed the major source of their income. The average acreage of these farms was 0.8-0.9 ha. In total 2,000 to 2,500 workers are involved in the production of carnations.

Carnations are cultured for one or two years. Two types of flowers are distinguished: the spray carnation with a cluster of flowers and the standard carnation with one central flower. To obtain a spray carnation the central flower-bud has to be removed once, while a standard carnation is obtained by removing the side-flower-buds regularly. The most important daily manual crop activities are harvesting by cutting off the flowers with a knife, collecting them into a bundle on arm or on cord above the crops (called cutting), sorting according to quality and bundling them to a bunch. These last two tasks are usually performed by the same person in one action (called sorting/-bundling). From our own observations as well as from a worker-crop contact analysis (Wicker & Guthrie, 1980), it appears that during the activities mentioned contact beteen skin and the treated flowers is limited mainly to hands and forearms.

Exposure to pesticides depends on many variables. This makes it necessary to perform a study in a large representative population. Another approach is to study cases in which the highest levels of exposure are expected: the 'worst case' strategy. A study according to this strategy can be performed in a relative small sample selected on criteria defining the 'worst case'. The farms in the present study were selected with regard to the worst cases of dermal exposure to pesticides during cutting of flowers. The following criteria were used:

- farms for which carnations form the major source of income;
- farms producing standard carnations, because contact with the treated crops during cutting was considered to be more intensive for these flowers than for spray carnations;
- the crop had to be relatively old, because the older the crop the more dense and high it is, leading to more intensive contact during cutting than with young crops.

Some species of the standard carnation may have a greater height and density than others, so dermal exposure during cutting could be different for different species. However, according to the opinion of experts on carnations the differences in practice are very small and have to be attributed more to culture conditions and not to varieties.
The four selected pesticides are mainly applied between September and December, with the shortest re-entry time in September. Production and growing speed are declining in the months thereafter and the re-entry time increases, which may lead to lower dermal exposure. Measurements were, therefore, carried out between September and December at the first re-entry after application, so exposure was assumed to be highest for that particular situation.
Spraying of pesticides gives rise to higher levels of dermal exposure than fogging (misting), since spraying is crop-directed and thus leads to more deposition of pesticide on the leaves (Lindquist et al., 1987). No distinction could be made beforehand between dermal exposure after re-entry for spraying or dusting. Two of the selected pesticides are only dusted in carnation culture (thiram, zineb). Thiophanate-methyl is usually sprayed. Chlorothalonil is sprayed, fogged and sometimes dusted with the dry formulation (wettable powder) meant to be sprayed.

By means of a questionnaire, sent to 208 carnation farms situated in Het Westland, farms which met the 'worst case' criteria were selected. From the 115 farmers who returned the questionnaire 18 refused to participate and 36 farms were not suitable for various reasons. Finally, 'worst case' farms could be selected from 61 remaining farms. Measurements were carried out on 18 carnation production farms. The 18 selected farms had an average of 0.8 ha for carnation culture and employed on the average 4 workers. To extend the number of measurements, exposure to chlorothalonil was also measured on 4 spray carnation farms. This also allowed comparison of the levels of exposure on standard carnation and spray carnation farms.

2.2 Assessment of dermal exposure.

Dermal exposure of all workers on the selected farms was measured by means of (pre-washed) cotton gloves covering hand and forearm (cotton: 200 g/m^2, surface (one sided) 370 cm^2; J. van der Wee B.V., Riel, The Netherlands). Measurements were performed during the whole period of cutting and sorting/bundling. In order to minimize break-through of the gloves followed by transfer of pesticide from glove to hand, a pair of gloves was not worn longer than one hour. In a pilot study the breakthrough was found to be about 5% for chlorothalonil after 1 hour. After a break in the work period the workers received a new pair of gloves.

During application exposure was measured for a whole application period including loading and mixing of the pesticide. Samples were stored in the dark at 4 °C awaiting chemical analysis for the amount of pesticide.

2.3 Assessment of respiratory exposure

Respiratory exposure during harvesting was determined only after dusting thiram and zineb. Exposure of the workers involved was measured during the same period as defined for dermal exposure during cutting and sorting/bundling. Respiratory exposure after re-entry after spraying was thought to be negligible. For all four pesticides respiratory exposure was also measured during the whole application period, including loading and mixing.

Measurements were carried out using an IOM personal air sampler (IOM, Negretti Automation, England) with a Mixed Cellulose Ester filter (25 mm, pore diameter 8μm, Millipore Corporation, USA) attached to a constant-flow pump operating at 2 l/min (P2500A Air Sampling Pump, Dupont, USA), estimating the inspirable fraction according to ACGIH (ACGIH, 1985; Mark & Vincent, 1986). Flows were checked before and after the exposure period by using a precalibrated rotameter tube (ROTA, Dr Hennig GmbH, West-Germany). For chlorothalonil an adsorption tube (XAD_2, SKC Inc., USA) was placed between the filter holder and the pump in order to trap chlorothalonil vapour evaporating from the filter. Filters in polypropylene tubes (Greiner und Söhne GmbH, West-Germany) and XAD_2-tubes were stored in the dark at 4° C awaiting chemical analysis.

2.4 Assessment of dislodgeable foliar residue

Just before application and after re-entry twelve leaves were sampled in duplicate from six adjacent beds at the height the flowers are cut. After collection dislodgeable residues were obtained as described by Iwata et al. (1977). Total leaf surface (one-sided) was measured with a surface area meter (LI-COR, 3100). The solution containing the dislodgeable residue was analysed for pesticides. The coefficient of variation (CV_t) for leaf sampling and analysis was between 10 and 14%, depending on the pesticide.

2.5 Chemical analysis of pesticides

Chlorothalonil

Solutions of dislodgeable foliar residue or methanol extracts of gloves and filters containing chlorothalonil were extracted with hexane. Analytical recovery from filters and gloves was more than 90%. Recovery from DFR solutions was 100%. Chlorothalonil was then quantified by means of liquid chromatography with 1% dioxane in n-hexane as eluent on a non-modified silica column with UV detection at 254 nm. The between-day coefficient of variation (CV) was less than 5% for all matrices (Jongen et al., 1990).

Zineb

Zineb was converted into its soluble sodium form with Na_4EDTA and extracted into chloroform/hexane as an ion pair with tetrabutylammoniumhydroxide. After derivatization with methyliodide, the derivative was quantified by reversed phase HPLC with UV detection at 272 nm. Analytical recovery from filters, gloves and DFR solutions were more than 90%. The between-day CV was approximately 7% for all matrices (Engel, 1988a).

Thiophanate-methyl and thiram

Thiophanate-methyl and thiram in DFR solutions were analysed directly by reversed phase HPLC and UV detection at 254 nm. Gloves and air sample filters were extracted with acetonitril before HPLC analysis. Recoveries from filters and gloves were more than 90%, the between-day CV's less than 5% (Engel, 1988b, 1988c).

2.6 Data on skin effects and on subjective effects

A physician visited 17 of the 18 carnation production farms in which measurements were performed and took a history of 84 workers in order to registrate experienced (local) health effects. Further, the hands and forearms were examined carefully for local effects on the skin.

2.7 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). Differences between groups were tested by the Mann-Whitney test (Snedecor & Cochran, 1982) on the non-transformed data. The relation between dermal exposure and dislodgeable foliar residue was studied with linear regression using least squares estimates.

3 RESULTS

3.1 Dermal exposure after re-entry

Dermal exposure of 95 workers was measured during cutting and of 38 workers during sorting/bundling. The work was carried out in crops which had been treated with pesticides 35 h earlier on the average. Gloves were worn for an average of 72 minutes. In this period the workers cut about 400 m^2 of plants. Sorting/bundling of these carnations took 52 minutes. More details are presented in table 1. These data for spraying and dusting of pesticides differed little.

Table 1 General data on dermal exposure measurements during cutting and sorting/bundling of carnations.

	Cutting	Sorting/bundling
Number of measurements	95	38
after spraying	43	13
after dusting	52	25
Exposure period (min)(1	72 (34)	52 (27)
after spraying	87 (33)	57 (31)
after dusting	62 (31)	49 (25)
Re-entry time (h)(1	35 (26)	--(2
after spraying	38 (30)	--(2
after dusting	33 (23)	--(2
Harvested area (m^2)(1	397 (312)	--(2

1 Expressed as mean and standard deviation (in parentheses)

2 Irrelevant

The results of the dermal exposure measurements during cutting are presented in figure 1. For all four pesticides the dermal exposure is generally within the range of 1 to 100 mg/h. The geometric means (GM) for the different pesticides vary between 4.4 and 16.1 mg/h (GSD: 2.0-3.3). The results presented in table 2 show that dermal exposure during crop tasks after application of pesticides by spraying is significantly higher than that after dusting. This is seen for both exposure during cutting and exposure during sorting/bundling. The average dermal exposure during sorting/bundling (7.3 mg/h) is not

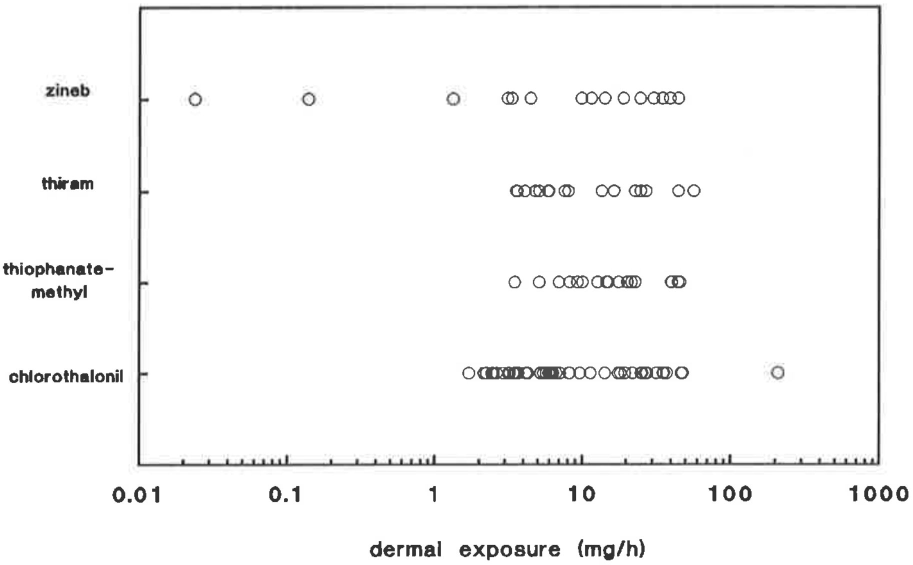

Figure 1 Dermal exposure of hands and forearms to pesticides during cutting of carnations.

significant lower than that during cutting (10.6 mg/h). Differences in dermal exposure to the different pesticides are small for a particular application technique. Dermal exposure after re-entry was compared for the two types of carnations only after spraying chlorothalonil. Exposure during cutting of spray carnations was significantly lower than that during cutting of standard carnations (11.7 (GSD: 2.6) and 21.0 (GSD: 1.3) mg/h, respectively), which supports one of the assumptions for 'worst case' conditions.

In order to investigate to what extent dermal exposure during cutting is related to the DFR linear regression analysis was carried out ignoring application technique and pesticide used. The result is shown in figure 2. The regression coefficient or transfer factor, which is a measure of the transfer of the pesticide from leaves to hands, is approximately 4500 cm^2/h. Separate regression coefficients for different pesticides and different techniques are presented in table 3. Transfer factors vary from 2900 cm^2/h for spraying chlorothalonil to 10,000 cm^2/h for dusting thiram. Noticeable is the difference in the transfer factors of chlorothalonil for spraying and dusting.

Table 2 Dermal exposure of hands and forearms to pesticides after re-entry in greenhouses for carnation culture.

Pesticide	Application technique	Dermal exposure (mg/h)[1]			
		n[2]	Cutting	n[2]	Sorting/bundling
chlorothalonil	spraying	22	14.4 (2.3)		
thiophanate-methyl		21	16.1 (2.0)		
total spraying		43	15.2 (2.1)*	13	20.8 (1.9)+
chlorothalonil	dusting	19	4.4 (3.3)		
thiram		17	10.4 (2.5)		
zineb		13[3]	11.9 (3.1)		
total dusting		49	7.7 (3.2)*	22	3.9 (5.3)+
total		92	10.6 (2.8)	35	7.3 (4.6)

1 Expressed as geometric mean (GM) and geometric standard deviation (GSD)

2 Number of observations

3 After log-transformation 3 observations were obtained as missing value because dermal exposure was close to zero

*, + $p < 0.05$ (Mann-Whitney Test for 2 groups)

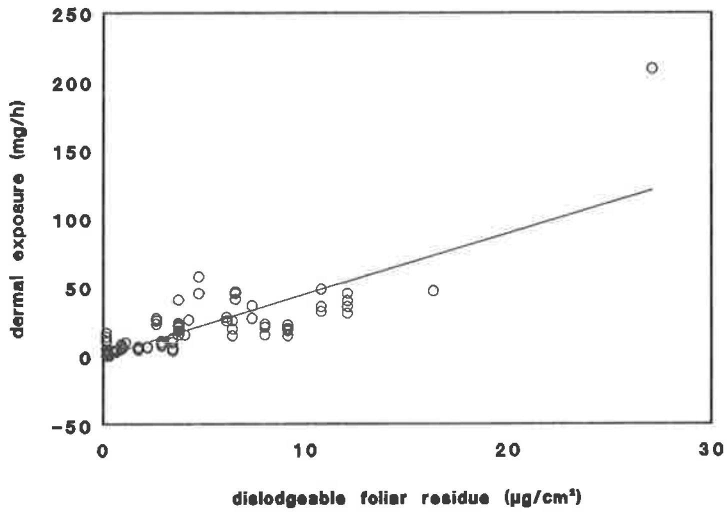

Figure 2 Association of dermal exposure of hands and forearms to pesticides during cutting of carnations and the dislodgeable foliar residue.

From table 3 it is obvious that dermal exposure during cutting depends strongly on the amount of dislodgeable foliar residue. The variance in dermal exposure can be

Table 3 Relationship between dermal exposure (μg/h) of hands and forearms to pesticides during cutting of carnations and dislodgeable foliar residue (μg/cm^2).

Pesticide	n[1]	Regression coefficient[2]	R^2
chlorothalonil (sprayed)	22	2,865 (465)	0.65
chlorothalonil (dusted)	19	6,475 (560)	0.88
thiophanate-methyl (sprayed)	21	3,665 (1,405)	0.26
thiram (dusted)	17	10,005 (1,100)	0.85
zineb (dusted)	16	2,840 (245)	0.91
total	95	4,465 (325)	0.66

1 Number of observations
2 Standard error (s.e.) in parentheses

explained for more than 65% by the regression on DFR. For most pesticides, sprayed or dusted, this is even higher: up to 90%. For spraying thiophanate-methyl it is only about 25%, which may be due to the relative large variance found in the dermal exposure over a small range of DFR.
The observed overall difference between dermal exposure during cutting after spraying and that after dusting (table 2) might be due to a difference in DFR after application (DFR_1) between spraying and dusting (table 4). In order to investigate to what extent

Table 4 Dislodgeable foliar residue after application (DFR_1), dislodgeable foliar residue before application (DFR_0) and applied dose for spraying and dusting.

Application technique	DFR_1[1] (μg/cm^2)	DFR_0[1] (μg/cm^2)	Applied dose[2] (g/1000 m^2)
spraying (n=14)	4.79 (1.7)	0.59 (5.1)	307 (149)
dusting (n=22)	1.56 (5.6)	0.34 (8.0)	162 (231)

1 Expressed as geometric mean (GM) and geometric standard deviation (GSD)
2 Expressed as mean and standard deviation (in parentheses)

this difference may be ascribed to the application technique itself, a regression analysis was carried out with other possible determinants of DFR_1. The results are reported in table 5. It appears that DFR_1 is mainly determined by dislodgeable foliar residue already present on the leaves before the application in question (DFR_0) and by the applied dose of the pesticide, and not by the technique itself. The regression coefficient

Table 5 Relationship between dislodgeable foliar residue after application (DFR_1), applied dose, dislodgeable foliar residue before application (DFR_0) and application technique.

	Regression coefficient[1]			
	DFR_0 ($\mu g/cm^2$)	Applied dose ($g/1000\ m^2$)	D_{appl}[2]	R^2_{adj}[3]
Total (n=36)				
DFR_1 ($\mu g/cm^2$)	0.88 (0.06)***	0.009 (0.002)***	0.21 (0.87)	0.89
Spraying (n=14)				
DFR_1 ($\mu g/cm^2$)	0.71 (0.27)*	0.012 (0.003)**	----	0.59
Dusting (n=22)				
DFR_1 ($\mu g/cm^2$)	0.89 (0.08)***	0.009 (0.003)*	----	0.90

1 Standard error (s.e.) in parentheses
2 Dummy variable for application technique: 0=spraying 1=dusting
3 Amount of variance explained by regression
* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

for application technique (D_{appl}) plays no significant role in the model; the separate models for spraying and dusting are more or less the same. Only the amount of variance in DFR_1 explained by the models (R^2_{adj}) is different (59% vs. 90%).
The importance of previous applications is illustrated in Figure 3 for chlorothalonil and thiophanate-methyl. Both remain detectable on the foliage for about 60 days after application of a dose of 398 g and 327 g active ingredient/1000 m^2, respectively. For about 20 days after application the amount of DFR remains more or less the same.

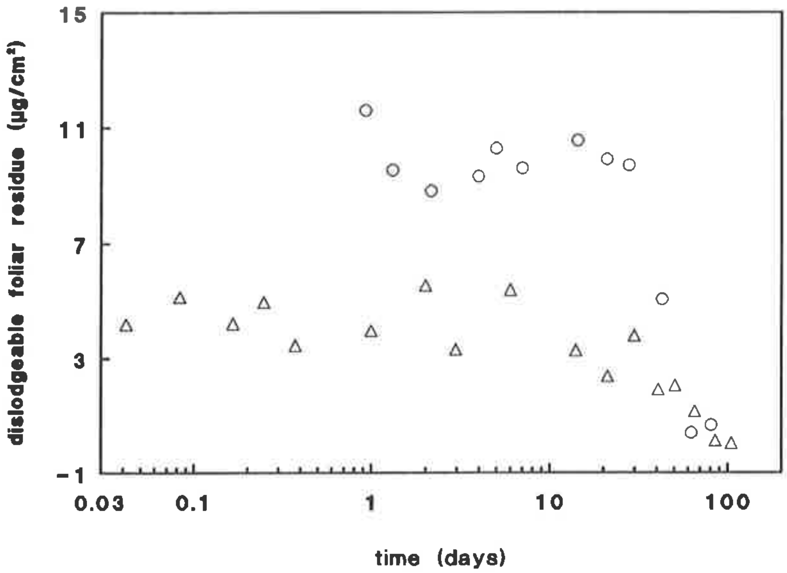

Figure 3 Decrease of the dislodgeable foliar residue of chlorothalonil (○) and thiophanate-methyl (△) on carnations with time.

3.2 Respiratory exposure after re-entry

The respiratory exposure after re-entry was only measured for dusting a pesticide (field strength dust). During cutting of the carnations a geometric average concentration of 0.07 mg/m^3 ($n=34$) was measured. Small but significant ($p<0.05$; Mann-Whitney test) differences were found after application of thiram ($n=13$) and zineb ($n=14$): 0.04 (2.3) and 0.11 (3.5) mg/m^3, respectively. Only a few measurements ($n=4$) could be performed during sorting/bundling; the exposure was about 0.01 mg/m^3.

3.3 Dermal exposure during application

Spraying was performed with hydraulic sprayers (spray gun, spray lance) for an average of 50.4 min per 1000 m^2. The applied dose was approximately 300 g/1000 m^2. Field strength dust was applied using backpack dust-blowers. Application of about 160 g/1000 m^2 took an average of 7.1 min per 1000 m^2.
The dermal exposure of hands and forearms during spraying and dusting was different (table 6), the exposure per hour being much higher for dusting than for spraying.

Expressing dermal exposure per unit area instead of per hour, shows that exposure during spraying is 7 times higher than that during dusting a pesticide (table 6).

Table 6 Dermal exposure of hands and forearms and respiratory exposure to pesticides during application by different techniques in greenhouses for carnation culture.

Application technique	n[1]	Dermal exposure[2] (mg/h)	Dermal exposure[2] (mg/1000 m^2)[3]	Respiratory exposure[2] (mg/m^3)
spraying	13	17.8 (4.3)*	13.5 (5.1)+	0.04 (2.3)#
dusting	10	39.4 (4.2)*	2.0 (9.1)+	0.67 (5.2)#
total	23	25.1 (4.4)	5.7 (8.2)	0.15 (6.4)

1 Number of observations
2 Expressed as geometric mean (GM) and geometric standard deviation (GSD)
3 Calculated with average exposure time per 1000 m^2 for spraying (50.4 min.) and dusting (7.1 min.)
*, +, # $p < 0.05$; Mann-Whitney test for two groups (spraying vs. dusting)

3.4 Respiratory exposure during application

Spraying leads to a significantly lower respiratory exposure than dusting (table 6). The variance in respiratory exposure during dusting is large (GSD=5.2). This may be explained by the fact that measurements included loading and mixing of the pesticide, an important source of exposure which exerts its influence during a relatively large part of the total application period. This is not the case for spraying.

3.5 Effects on the skin and subjective effects

On the 17 farms visited, 84 persons were interviewed using a standardized procedure to report experienced skin symptoms, and their hands and forearms were inspected for skin disorders. Of these 84 persons 57 were male and 27 female. Results of the interviews are presented in table 7.

Twelve of the 34 persons who reported dry hands did not consider this phenomenon a skin disorder. Altogether 21 persons reported dry hands to be a symptom occurring primarily during the winter season. One person related his dry hands to his work and

Table 7 Prevalence of reported symptoms and the actual skin disorders among 84 workers in greenhouses for carnation culture.

Reported symptoms	n	(%)[2]	Registered skin disorders[1]	n	(%)[2]
at skin/mucous membranes	23	(27)	total	39	(46)
dry skin	34	(40)	eczema	7	(8)
eye irritation	13	(15)	skin injuries	3	(4)
at respiratory tract	26	(31)	chapped hands	9	(11)
			dry hands	16	(19)
			dry hands with nail-rim damage	4	(5)

1 By visual inspection
2 Percentage of total number of observations (n=84)

6 (18%) definitely did not. All those who reported eye irritation and respiratory tract problems, related their disorders to their work. In many instances eye irritation was said to be caused by rubbing the eyes with dirty hands; respiratory problems were mainly related to 'incorrect' use of pesticides or activities causing dust exposure (clearing out the crop after the production period).

On examination, a disorder of the skin of the hands was diagnosed in 39 persons (46%). The different types of skin disorders found, are presented in table 6. The disorders most commonly found, were dry skin (16), chapped hands (9) and eczema (7).

4 DISCUSSION

4.1 Re-entry

The levels of dermal exposure of about 10 mg/h during harvesting (cutting) of carnations grown in greenhouses are in the range found by Davis et al. (1982; 1983) for apple thinning, using the same exposure assessment method. Others, like Popendorf & Leffingwell (1982) and Zweig et al. (1985), used pads to measure exposure, which makes comparison more difficult.

The maximum difference of a factor 5 in the levels of dermal exposure to the four different pesticides during cutting are small compared to the results obtained by Zweig et al. (1985). For harvesting of strawberries they found differences of up to a factor of 60 depending on the pesticide used. They ascribed this difference to the large variances in re-entry intervals (up to 14 days) and DFR. In the present study re-entry intervals were much shorter, 12-119 h (mean 35 h), and for the pesticides investigated differences in DFR were relatively small (range of a factor of 5). The overall results of dermal exposure after dusting a pesticide are strongly influenced by the low values for dusting a wettable powder of chlorothalonil (4.4 mg/h; n=19, table 2). But the levels of dermal exposure to the field strength dusts (thiram and zineb; 11 mg/h) after re-entry are still lower than those for spraying. This is also observed for dermal exposure during sorting/bundling.

As shown in table 3 the variance in dermal exposure during cutting of carnations can be explained for more than 65% by differences in DFR. The latter is higher after spraying than after dusting (table 4). DFR is determined by dislodgeable foliar residue remaining from previous applications (DFR_0) and the applied dose (table 5). From the results it appears that the mentioned difference in dermal exposure after spraying and dusting is not due to the application technique and formulation, but probably to the different applied doses of active ingredient. The finding that DFR_1 is also determined by DFR_0, points to cumulation of pesticides. This is supported by the data in Figure 3. When chlorothalonil or thiophanate-methyl is applied within 60 days after the prior application it will, apparently, cumulate on the foliage. Therefore, levels of dermal exposure and concomitant health risks will increase in such cases, assuming that the efficiency of transfer from foliage to skin is not time dependent. The observed decline

of chlorothalonil and thiophanate-methyl after 20 days is probably not due to physical or chemical decay but to growth and harvesting. More than 60 days after application levels of DFR of leaf samples taken from lower parts of the plant than the contact zone during cutting are equivalent with those shortly after application. During the 60 days, leaves in the contact zone from which DFR samples are taken disappear due to harvesting and young untreated leaves appear on the fast growing crop.
The observed difference between dermal exposure during sorting/bundling after spraying and that after dusting is more pronounced, which may be due to dislodging of dry (dusted) pesticide from the leaves during cutting and handling of flowers before sorting/bundling. Further, during sorting/bundling contact with the treated material is probably more intense. This assumption is supported by the observation that after spraying a pesticide dermal exposure is higher during sorting/bundling than during cutting.

The transfer factor of approximately 4500 cm^2/h established in this study is of the same order of magnitude as that for strawberries and blueberries (Zweig et al., 1985), citrus, peaches and grapes (Popendorf & Leffingwell, 1982). Zweig et al. (1985) calculated from their own studies and from studies by Popendorf transfer factors ranging from about 1000 to 60,000 cm^2/h depending on pesticide and crop, with an average of 7800 cm^2/h. For fruit (citrus) harvesters Zweig et al. (1984) obtained a factor of 5000 cm^2/h. Since 65% of the variance in dermal exposure in the present study regarding the carnation culture can be explained by the amount of DFR after application, it seems appropriate to use this parameter for a first estimation of dermal exposure of field-workers during harvesting, although some differences have been found in the transfer factors of different pesticides (table 3). These relatively small differences can probably be ascribed to differences in physical and chemical properties of both the formulation and the active ingredient. The estimated dermal exposure can be used to establish, for instance, (safe) re-entry intervals without the need for studies involving workers.
The difference in respiratory exposure to zineb and thiram during cutting of treated carnations is remarkable and unexplicable at present, because formulation and the dosage used, as well as the dermal exposure are not much different.

4.2 Application

Dermal exposure per hour during application of field strength dusts (thiram and zineb) was significantly higher than that measured during spraying, although the applied dose (g/1000 m^2) during dusting is much lower. An explanation may be the contribution of dermal exposure during mixing and loading, which is relatively large in short exposure periods such as in the case of dusting. Fenske et al. (1987) reported a higher exposure during loading/mixing of a wettable powder than during its application. Another cause may be that a spraying tank is usually loaded with one liter jars of liquid formulation while field strength dusts are loaded from 5 or 10 kg bags. Handling large packages may result in relatively high exposure. Expressing dermal exposure in amounts per unit area allows a better comparison of dermal exposure for different application techniques. The total dermal exposure per 1000 m^2 is about 7 times smaller for dusting than for spraying, because spraying takes more time.
Respiratory exposure is significantly higher during dusting than during spraying. This difference can be ascribed to differences in formulation and application technique which determine particle size (Matthews, 1985) and to different concentrations of the active ingredient in the aerosol. The latter is much higher during application of field strength dusts. Also the difference in formulation during loading/mixing may play a role; powders may lead to a higher emission than liquids.

4.3 Potential health risks due to exposure to pesticides

The present results indicate a relatively high potential exposure for manual activities in crops treated with pesticides compared to application of pesticides, the duration of manual activities on a working day being many times the duration of application. Furthermore, for all workers manual crop activities recur daily whereas application is limited to one person, maybe a few times a week.
It should be noted that the observed levels of exposure are for unprotected hands and forearms on which pesticides are accumulating during the day. Health risks due to the levels of exposure can be estimated for the separate pesticides of this study from toxicological data such as 'no-effect levels' (NEL) and dermal and respiratory absorption data. Using the measured intensity and duration of exposure and so-called safety factors (Genderen et al., 1988) based on differences in extrapolation from animal to man (Vocci & Faber, 1988) and on the seriousness of the effects under conside-

ration, health-based exposure limit values may be extrapolated.
Frequently, certain assumptions have to be made for such estimations because of lack of the necessary data. For example, NEL values may be based on oral toxicological data. Exposure to pesticides after re-entry, however, is mainly limited to skin and airways, although oral exposure has also to be considered. For extrapolation from an oral NEL established in animal studies to man, data on absorption (dermal and respiratory) are required. For dermal absorption knowledge is necessary on the dermal penetration flux, which depends on the amount on the skin and the exposed area. Generally, these data are not available. This is the major reason to use the percentage absorption, although this procedure lacks a theoretical basis. The Joint Medical Panel (1987) uses in the absence of penetration data three indicative calculation measures for dermal penetration, i.e. 100%, 10%, 1%.
In table 8 'indicative limit values' (ILV: the amount of pesticide considered to be the highest level of exposure which, supposedly, does not lead to an adverse health effect) for the four pesticides chosen in this study are presented. The ILV's are based on the observed no toxic effect level (Deutsche Forschungsgemeinschaft, 1976; Hayes, 1982; FAO, 1987) by using a safety factor of 10 for intra- and interspecies differences. Only for chlorothalonil a safety factor of 30 was used following FAO (1987) and because of the uncertainty in the extrapolation of the effects found in the kidneys of rats and mice.
In table 8 the observed average potential levels of exposure to the selected pesticides are compared to the ILV's. It is concluded that for chlorothalonil a health risk exists both during application and after re-entry. The respiratory exposure to chlorothalonil after dusting a wettable powder may also lead to health risks. Contact with thiram has to be avoided because of its sensitizing properties. Available data on dermal absorption for zineb are inadequate, precluding a proper estimate of the dermal ILV. The respiratory exposure to zineb during application is close to the ILV and may lead to health risks, especially if it is considered that in table 8 average levels of exposure are given.
In the preceding part dermal and respiratory exposure has been evaluated separately. However, one should realize that both types of exposure occur simultaneously. It should further be considered that the analysis has been based on average levels of exposure. Ninety-percentiles of the observed levels of exposure are a factor 3 to 5 higher.
A chronic health risk appears to exist after re-entry of greenhouses for carnation culture after application of pesticides that are relatively toxic (NEL $<$ 1 mg/kg) and show a relatively good dermal absorption.

Table 8 Measured average exposure per task on a working day in greenhouses for carnation culture, and indicative limit values (ILV) for a day for some pesticides.

Pesticide		Dermal exposure[2]				Respiratory exposure[2]			
	NEL (mg/kg.day)	ILV[1] (mg/day)	A	C (mg/task)	S/B	ILV[1] (mg/day)	A	C (mg/task)	S/B
chlorothalonil (sprayed)	0.4	10	69	58	54	1	0.3	n.m.	n.m.
chlorothalonil (dusted)			11	18	7		0.2	0.5	n.m.
thiophanate-methyl (sprayed)	8	560	37	64	23	56	0.1	n.m.	n.m.
thiram (dusted)	4	280[3]	20	42	9	10	0.2	0.2	n.c.
zineb (dusted)	1	7-700	19	48	8	1	0.9	0.6	n.c.

ILV = indicative limit value; A = application; C = cutting; S/B = sorting/bundling

n.m. = not measured; n.c.= not calculated

1 ILV is calculated for a man of 70 kg, using of a safety factor 10, except for chlorothalonil(= 30);
Dermal absorption is assumed to be 10%; for zineb data on absorption were inadequate: 1% and 100% are used.
Respiratory absorption is assumed to be 100% and a respiratory volume of 10 m^3 is taken for a working day.

2 Average task times on a day : spraying 85 min (1700 m^2)
dusting 25 min (3600 m^2)
cutting 4 hours
sorting/bundling 2 hours

3 Sensitizing compound

4.4 Effects on the skin and subjective health effects

The lack of overlap of the medical observations and the self-reported symptoms is probably due to the fact that reported skin disorders were not necessarily present at the time of investigation. Observations have been made in a relatively short period of the year (September to November) in which a particular group of pesticides (fungicides) is frequently used. The prevalence of skin disorders in this study was higher than would be expected from observations in the general population (5%) and an agrarian population (6.5%) (Coenraads, 1983). However, the population investigated was small and some selection bias may have occurred.

Some of the problems reported are related by the workers themselves to the use of certain pesticides. In this context chlorothalonil is mentioned frequently, which is in agreement to skin irritating and sensitizing properties of chlorothalonil reported in the literature (Bruynzeel & Ketel, 1986). In a previous epidemiological study among 1355 Dutch farmers in eight agrarian sectors, Willems et al. (1984) found also a relation between skin problems experienced and pesticide use, notably in mushroom culture and horticulture.

4.5 Conclusions and recommendations

The levels of dermal exposure on a working day due to manual crop activities were found to be higher than those due to application of a pesticide. The respiratory exposure to pesticides applied as a field strength dust is after re-entry of the same order of magnitude as during the application itself when adjusted for exposure time.

The dermal exposure during cutting, the major manual crop activity, depends on an average for about 65% on the amount of dislodgeable pesticide on the foliage (DFR). In view of this strong dependence and the relatively small differences in the transfer factors of the four pesticides, it seems allowable to extrapolate the present results for cutting of carnations in greenhouses to other pesticides. This means that 'worst case' predictions of dermal exposure to other pesticides can be made by measuring dislodgeable foliar residue. Probably the dependence of dermal exposure on DFR and the magnitude of transfer factors are task- and crop-specific. More research on other manual activities and other types of horticulture in greenhouses is needed.

From the observed levels of dermal exposure during cutting (about 10 mg/h) and sorting/bundling (about 7 mg/h) it can be concluded that during crop activities a health

risk may exist for pesticides with a low 'no-effect level' and good skin-penetrating properties. More knowledge about simultaneous exposure by different routes, and about body uptake must be gathered, e.g. by using biological monitoring to determine the internal exposure, independent of the routes of exposure.
In general the levels of exposure during cutting of carnation will be lower, because measurements were carried out in 'worst case' situations for cutting. The results of other activities were not obtained in 'worst case' situations. To what extent the latter situations were representative for the cultivation of carnations in greenhouses is difficult to estimate. The influence of the selection criteria on the representativeness of other activities is unknown.
The prevalence of skin disorders on hands was found to be higher than expected. Some of the problems are related to certain pesticides by the workers themselves. General conclusions cannot be drawn, since the population was too small and probably biased by selection. It seems worthwhile to initiate a separate study on skin disorders among growers of ornamental flowers.

The levels of exposure during manual crop activities found and the emerging potential health hazards for some pesticides are of importance in health risk evaluation for pesticide approval procedures. Data on dermal and respiratory exposure after re-entry and on skin-penetrating properties are required for proper health risk evaluation.
The necessary decrease in the present levels of exposure may be achieved by minimizing the use of pesticides and lowering the dose and frequency of application. This will lead to a lower DFR and thus to a lower dermal exposure after re-entry. Furthermore, lowering the amount of pesticides on the skin by good personal hygiene is strongly recommended. Respiratory exposure after re-entry may be reduced by using liquid formulations instead of powders. Further research on cumulation and dissipation of pesticides on foliage is desirable. Research on cleaning of the skin from pesticides during work has to be undertaken.
Generally, the workers in treated crops were not aware of the intensive contact with pesticides occurring during a large part of the day. Especially the crop worker appears not to be informed on work procedures for handling pesticides as present on the product labels, since he does not handle the package. Therefore, the knowledge of the greenhouse worker about pesticides should be improved by training and education on toxicological properties of the relevant pesticides and on (personal) hygiene measures to take.

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