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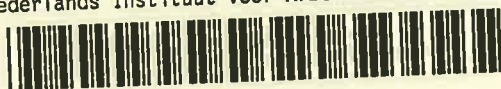
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Health in relation to — occupational exposure to pesticides in the Dutch flower bulb culture

Estimation of Long-Term Exposure to
Pesticides

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Health in relation to occupational exposure to pesticides in the Dutch flower bulb culture

Part 2

Estimation of Long-Term Exposure to Pesticides

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Summary

The cultivation of flower bulbs in the "Bollenstreek" area is a traditional, small-scaled, agricultural activity, which is attended with the use of different pesticides at all stages of the growing process. Before planting pesticides are used for soil fumigation and disinfection of the bulbs and after planting pesticides are sprayed for weed killing and for crop protection. After lifting and grading, bulbs which have to be planted the next year and thus put in stock are protected by treatment with pesticides. As a part of an epidemiological study with a cross-sectional design aiming at the establishment of possible health effects due to exposure to pesticides in the flower bulb culture, an occupational hygiene study has been conducted in order to discriminate or to rank participants according to the level of exposure.

Although there is an intensive use of pesticides by farmers in the process of flower bulb growing it was learned from observations on the spot that application of pesticides for stock protection and soil disinfection is performed by contract workers. In most cases the farmer's own exposure is limited to application for weed killing, crop protection and bulb disinfection. Exposure has been estimated to be the highest for the last two applications. So, for the purpose of this study, only exposure due to crop protection and bulb disinfection activities have been considered. For crop protection as well as bulb disinfection different techniques have been distinguished through questionnaires. Tractor boom spraying and backpack spraying are currently the most frequently used spraying techniques, whereas bulb dipping is the most popular method of bulb disinfection. The drive-in vessel and the dipping bath techniques are the most extensively used techniques of dipping.

Various pesticides have been used for crop protection and bulb disinfection during the past 40 years. Fortunately, the large majority of farmers used the same pesticides in the same periods of time, since most farmers followed strictly the advices on crop protection and pesticides given by the Flower Bulb Research Centre (LBO at Lisse, The Netherlands). Zineb/maneb, a mixture of two dithiocarbamates and the major pesticide for both fields of application has been used in flower bulb cultivation for about thirty years now.

In order to obtain a more or less quantitative estimation of exposure, a model was constructed which was applicable to crop protection and bulb disinfection, and which used zineb/maneb as a representative compound. Since the application technique affects both intensity and duration of exposure, for each technique and method of mixing and loading a "specific exposure level" was calculated, i.e. a geometric or arithmetic mean of exposure expressed as exposure per unit of acreage. These specific exposure levels were used together with information on

application technique and bulb acreage obtained from each individual farmer, to estimate the individual exposure.

"Technique- or method-specific exposure levels" of tractor boom and backpack spraying techniques were obtained by adjusting exposure data of a UK data base to the specific conditions of flower bulb farming, like application speed [ha/h] and application volume rate [l/ha]. These data were obtained from log-book records. From field studies aimed at the assessment of both respiratory and dermal exposure to zineb/maneb, method-specific exposure levels were obtained for bike spraying, three methods of mixing and loading a wettable powder, which is the most commonly used formulation of zineb/maneb for field spraying, and two major bulb disinfection techniques. Since in all these cases respiratory exposure was found to be less than 1% of the dermal exposure and respiratory exposure during bulb disinfection was considered to be neglectable, the dermal route was considered the most appropriate for estimating long-term exposure due to both forms of application.

The average dermal exposure per year for various techniques, based on an average number of 7 applications of zineb/maneb per year, ranged from 63 (tractor-boom spraying) to 4900 mg/ha (bike spraying), whereas dermal exposure due to mixing and loading ranged from 216 (direct tank filling)¹ to 2300 mg/ha (premix-scooping)². The estimated exposure during disinfection of an amount of bulbs to be planted on 1 hectare, about 10,000 kg in the case of tulips, ranged from 8 (drive-in vessel) to 430 mg/ha.

The cumulative exposure of farmers has been estimated by an exposure index. For each individual farmer the potential exposure was calculated for different years by multiplying the relevant method-specific exposure levels for the methods of crop protection and mixing and loading used by the farmer in that particular year, the acreage of farming land, and the number of applications per year. Similarly, the dermal exposure due to bulb disinfection was calculated using the bulb acreages for the different cultivated varieties. The exposure index was calculated over a five years, a ten years and the lifetime exposure period. The lifetime exposure indices of 134 flower bulb farmers, obtained from the arithmetical mean for the method-specific exposure levels, ranged from 0.25 to 2,200 in arbitrary units, with a median of 200 and a 90-percentile of 770. These figures are based on the assumption that in all applications zineb/maneb was used. If this were so, the dimension of the figures is grams of zineb/maneb.

Coefficients of correlation between the five years, the ten years and the lifetime exposure indices were relatively high ($r > 0.8$) and the agreement of classification into exposure level groups was satisfactory ($\kappa > 0.4$). These results indicate a low within-person-variation of the exposure for the years considered.

Correlation between lifetime exposure and years of exposure was moderate ($r = 0.45$) which is considered to be an indication that the use of application techniques

¹ direct tank filling: filling of the tank by pouring the powder directly from the bag.

² premix-scooping: scooping of the powder into a bucket and pre-mixing the powder with water; the suspension is poured into the tank.

and other farming-related exposure parameters, result in a long-term exposure assessment which differs from an assessment exclusively based on years of exposure.

This approach of estimating an exposure index for long-term exposure has considerable limitations in application of the exposure index for health effects studies, since it is designed to classify members of a population according to exposure and does not necessarily reflect the absolute value of individual exposure. Correlation analysis of exposure indices based on lower and upper 95-percentile confidence limits of the exposure levels indicates a relative insensitivity of the ranking to parallel changes of the method-specific exposure levels for all members of the population. However, this may not be relevant for the exposure of each individual separately. Within the series of processes which start with exposure and leads via absorption (or penetration), metabolism and distribution to a possible toxic effect, non-linear relations may lead to completely different absorbed doses in a target organ in comparison to the estimated external dermal exposure. Moreover, cumulative exposure calculated as the present exposure index suggests a linear cumulative dose-response relation which may or may not be the right concept for the health effect under consideration.

It is concluded that the exposure index is a very useful tool in an explorative study to rank populations according to exposure and is helpful in the formulation of hypotheses for further study. The exposure index itself provides the possibility to indicate the sources of low and high exposure, which is important for the implementation of hygienic measures.

General introduction

Flower bulb farming is an important agricultural activity in The Netherlands, involving about four thousand farms and more than fifteen thousand hectares of farming land. Because of the special demands of this culture, suitable farming soil is rare and hence cultivation is very intensive. Flower bulbs are very susceptible to diseases. Therefore, each stage of the growing process throughout the year needs the use of different kinds of pesticides, e.g. fungicides, herbicides and insecticides, which are applied by various methods using various work practices.

As part of an epidemiological evaluation of neurotoxic and neurobehavioural effects of pesticides on flower bulb farmers a study was performed to obtain an estimate of their long-term exposure to these compounds. The general design of this study will be reported in part I of the series of reports (Brouwer and De Wolff, 1990). Section I of this report describes the development of a method to estimate exposure and the application of this method to rank the participants in the study on health effects according to their long-term exposure. Section II contains a report of some exposure studies in the field during bulb farming to obtain exposure data for the ranking.

Introduction

The flower bulb culture has originated in an area in the western part of The Netherlands, called "De Bollenstreek". Farms in this area are relatively small and have relatively many subsidiary activities related to the cultivation of flower bulbs, such as cultivation of (bulb)flowers and commercial activities. Other aspects of small-scale farming are cultivation of a large variety of flower bulbs and a minimum of hired labour. Special labour-intensive activities, e.g. lifting, grading and planting, have always demanded large numbers of temporary workers, mostly pupils. Tulip, hyacinth, daffodil, crocus are the most cultivated bulb varieties in this region. The application of pesticides on a large scale has started after World War II. Figure 1 illustrates the application of pesticides at the various stages of the bulb growing process. Since about 1950, pesticides are applied throughout the process of bulb growing. After planting the bulbs, herbicides are applied for weed killing about two times a year. Paraquat is used frequently on fallow and other compounds like chlorpropham are used at an early stage of sprouting of the crop. In the early spring frequent field spraying is started for crop protection, mostly against *Botrytus*, but during the flowering also against insects. Dithiocarbamates like zineb and maneb have been used for about 30 years as fungicides.

Cyanide has been applied for many years for stock protection. From the late seventies onwards, fumigation of this compound may only be done by licensed applicators. Less frequently, the farmer uses an aerosol spray of dichlorvos, or naphthalene as a vapour releasing product. About six weeks before planting, nematocides are applied as soil fumigants, e.g. 1,3-dichloropropene, mostly by commercial applicators.

Just before planting bulbs are disinfected. Fungicides are applied to protect the bulbs against *Pythium*, *Fusarium* and *Botrytus*. Organic mercury compounds have been used until they were banned in 1976, thereafter captafol has been used until this compound was banned in 1982. Several other fungicides are being used frequently now. Table 1 contains a summary of the most important ones.

In epidemiology exposure means potential contact. It does not always imply a level of intensity. Most epidemiological studies dealing with pesticides lack a (semi) quantitative assessment of exposure (Corrao et al., 1989). In the absence of quantitative exposure data, a job-exposure matrix or exposure index is often used in retrospective occupational epidemiological studies (Marsh, 1987; Checkoway et al., 1987; Kriebel et al., 1988). This measure of exposure is used to link worker exposure histories with health effects. In many cases a linear dose response relation is assumed where dose is assessed as cumulative exposure, i.e. product of duration and intensity of exposure. In evaluating painters exposed to solvents, Fidler et al. (1987) used an exposure index for assessment of lifetime exposure. The basics of this exposure index are useful for application in the present study because more factors than duration of exposure are involved, such as the volume of the solvents used, and methods and rates of their application. The exposure index itself is a weighted average for all the solvents used, where the weights are based on the methods of application and the presence or absence of ventilation. However, it is focussed on respiratory exposure to volatile components, whereas exposure to pesticides is also related to the dermal route and in most cases potential dermal exposure appears to exceed potential respiratory exposure with a factor of hundred to thousand (van Hemmen, 1990).

The aim of the present study was to obtain an exposure index which reliably estimates long-term exposure to various pesticides due to application in flower bulb growing. Since usually the only source of information with regard to exposure in the past is a questionnaire completed by the worker, such an exposure index should contain items which are relatively simple to obtain but relevant for individual exposure.

Materials and methods

2.1 Observations and questionnaires

With the assistance of experts of the Flower Bulb Research Centre (LBO, Lisse, The Netherlands) a questionnaire #1 was drafted, tested in a pilot study and administered to 382 farmers registered as flower bulb farmers in the South Holland bulb district ("De Bollenstreek"). The aim of the questionnaire was to obtain an inventory of the use of pesticides in the flower bulb culture. The subjects were asked to provide farm data, such as bulb acreage and cultivated varieties, the number of employees and the nature of their jobs. Further details were asked about the methods and techniques of application, the various uses of pesticides and the frequency of application. Besides, eight farms were selected because of their variety of bulb acreage and application techniques, to view all different forms of application of pesticides. The objective of these observations was to obtain a detailed description of the use of and exposure to pesticides from which relevant exposure variables could be derived.

From the respondents of the questionnaire #1 187 farmers were selected which used zineb/maneb for bulb disinfection, to receive a more detailed questionnaire #2 on bulb disinfection techniques. Since not all different bulb disinfection techniques were observed at the eight farms selected initially, an additional survey on bulb disinfection methods was conducted at another twelve farms. Another 25 farmers were asked to keep a log-book on the spraying of pesticides for weed killing and crop protection, in order to get relevant details about dose rate (kg/ha), application speed (ha/h) and volume rate (l/ha).

2.2 Determinants of exposure and methods of exposure assessment

Two different phases can be distinguished in the exposure during application of pesticides: exposure during preparation (mixing and loading) and exposure during the application itself. The latter includes maintenance and repair. In these two phases both respiratory and dermal exposure may occur. Exposure during repair will occur incidentally.

Generally, lifetime exposure or cumulative exposure is assumed to be the product of intensity and duration of exposure. Since exposure to pesticides in flower bulb farming is irregular over the years, discontinuous throughout the year and limited

to several stages of the bulb cultivation process, years of exposure does not reflect duration accurately. Therefore another determinant was needed. Total treated bulb acreage of a particular farmer, i.e. bulb acreage times frequency of application, corrected for application technique, reflects duration of exposure more precisely, since an application technique implies a specific application speed [ha/h]. Moreover, total bulb acreage corresponds to the yearly amount of pesticides applied, since for all pesticides a specific dose rate [kg active ingredient/ha] is advised. Application technique is considered to be the most important factor in the transfer of a part of the pesticide to the worker. Since it is generally accepted that the transfer of pesticides which results in exposure is dominated by physical factors, method-specific exposure levels or face values were considered to be useful. To estimate potential exposure, analogously to the exposure index of Fidler et al. (1987) an exposure index was defined by the general formula:

$$EI_{\text{year}} = \Sigma [\{ (E_{\text{weed kill.}} + E_{\text{crop prot.}} + E_{\text{mix/load}})_{\text{method}} * N + E_{\text{soil disinf.}} + (E_{\text{stock prot.}} * N_{\text{stock prot.}}) \} * A_{\text{total}} + (E_{\text{bulb disinf.}})_{\text{method}} * A_{\text{variety}}] * (1 - P)_{\text{field of application}}$$

E = (application) specific level of potential exposure [mg-ha-1]

A = bulb acreage [ha]

N = number of applications per year

P = protection factor

Since exposure may be different for different applications, with very different volume rates (volume of spraying liquid per hectare) and frequencies, the exposure index contains method(technique)-specific levels of potential exposure instead of total amount of pesticides applied and relative emission factors for each method, in correspondence to Fidler et al. (1987). The general formula of the exposure index can be used to calculate both respiratory and potential dermal exposure.

Potential dermal exposure is defined as total body exposure (including contamination of the clothes). Actual exposure is defined as exposure of the skin. The level of actual exposure may be affected by protection measures such as the use of gloves or protective clothing. If the protection factor is equal to zero (no protection), the unprotected parts of the body are supposed to comprise hands, head and neck. It is assumed that under all conditions considered in the present study normal (work) clothing will prevent penetration of pesticides towards the skin underneath to a large degree. Analogously actual respiratory could be defined as the amount of pesticides which is available for inhalation using respiratory protection (e.g. masks)

For each individual farmer an exposure index can be calculated for each year of exposure and these can be added to give a five years, ten years or lifetime cumulative exposure index. This can be done for every pesticide in use. If sufficient data are available and a similar pesticide "profile" exists throughout these years and for all farmers, the index can be used to rank the participants in a health effect study according to the level of exposure to a relevant marker compound.

Generic data bases

It is generally accepted that generic data of pesticide exposure can be used for extrapolation to other pesticides, since the amount of exposure is largely dependent on physical factors and to a much smaller extent on chemical factors (Van Hemmen, 1990). There are bodies of dermal and respiratory exposure data available from the United Kingdom and from West-Germany. The format of the data is different in several aspects, potential versus actual exposure, exposure dose in weight per weight of handled pesticide or in spray volume per unit of time, etc. The UK data base (Joint Medical Panel, 1986) has been used to derive method-specific exposure levels since in this reference exposure is expressed as potential exposure. The German approach (Biologische Bundesanstalt für Land- und Forstwirtschaft, 1988) does not fully take into account exposure of the hands because German regulations require the use of protective gloves for mixing and loading. The gloves are supposed to protect the worker sufficiently from exposure. In The Netherlands such requirements are lacking and so the approach of the UK seems more appropriate for Dutch situations.

The UK data base contains exposure data on two field spraying techniques, i.e. tractor boom spraying and backpack spraying. These techniques are used in flower bulb farming as well and the data can be adjusted to the specific conditions of bulb farming. The adjustment includes transformation of exposure from volume sprayed per unit of time (ml/h) into amount of pesticide per unit of acreage (kg/ha active ingredient (a.i.)) and calculating a geometric or arithmetic mean of exposure. For a detailed description of the transformation, see Appendix I.

No adequate data were available on exposure during mixing and loading of wettable powders, the major formulation form used in bulb farming. Neither were data available on levels of exposure due to other techniques of spraying nor due to disinfection techniques.

Field studies

In order to obtain method-specific exposure levels of mixing and loading and of bulb disinfection, field studies have been conducted which are described in detail in section II of this report. As part of these studies some preliminary experiments were done using another spraying technique i.e. bike spraying, the results of which were used to estimate the level of exposure for this technique.

Exposure during soil disinfection has been estimated during a separate study on the biological monitoring of 1,3-dichloropropene, the major soil disinfectant in The Netherlands. The results are reported in Part 7 of this series of reports, but they were not used for the estimation of long-term exposure of the flower bulb farmers, since only contract workers were exposed.

2.3 Assessment of lifetime exposure

To estimate lifetime exposure of the selected flower bulb workers in the health effect study, a self-administered questionnaire #3 was used. Each participant was asked for details about application techniques used for crop protection, frequency

of application, method of mixing and loading wettable powders and the area treated, for each year of a ten years period (1979 - 1988). Moreover, the number of years that crop protection was exercised was established. Similar questions were asked about bulb disinfection techniques, number of barrels disinfected and number of years involved in bulb disinfection. Furthermore, details were obtained on other activities involving pesticides, e.g. on the cultivation of bulb flowers and other ornamental flowers, both in greenhouses and in open air. Finally, subjects were asked about the use of personal protective equipment. During the participation in the health effect study the answers of each participant to the questionnaire were checked for completeness. If necessary, the participant was requested to complete the questionnaire.

Lifetime exposure was estimated by extrapolating the calculated exposure index backwards from the last reported year (i.e. 1979) to previous years of exposure.

2.4 Calculations and statistical analyses

Data from questionnaires were processed by statistical software (SPSS) and the exposure indices were calculated using spread sheet software (Lotus 123). Relationships among variables, e.g. among frequency of application and acreage, and log-transformed exposure indices calculated for a ten years period were studied by multiple regression using SOLO Statistical System. Differences between groups were tested non-parametrically (Mann-Whitney U test). The sensitivity of the exposure index to variations in the method-specific exposure levels was investigated by using upper and lower 95 percentile confidence limits and comparing the resulting indices, using simple correlation (Spearman rank correlation). Agreement of exposure ranking according to different exposure indices based on geometric or arithmetic means of five years, ten years or working lifetime, were estimated by the kappa coefficient using SOLO and BMDP-package 3 D. Kappa values ranging from 0.4 to 0.7 were considered satisfactory, while a value above 0.7 was regarded as excellent agreement (Fleiss, 1981). Reliability of corresponding data of the self-administered questionnaires #1 and #3 was expressed by the Spearman rank correlation coefficient.

Results and discussion

3.1 Pesticide application and exposed population

3.1.1 Field spraying

Three techniques of field spraying for weed killing and crop protection were observed. Manual techniques, i.e. backpack spraying and bike spraying with 10 or 15 l tanks, have been used very much in the past but are still in use now. Since the seventies, tractor boom sprayers with 400 l or 600 l tanks have been introduced and about 70% of the farmers is using them nowadays. Sometimes one of the other techniques is used additionally. Thirty-two percent of the farmers changed spraying techniques during the last ten years, about half of them (17%) from bike spraying to tractor boom spraying. None of the respondent flower bulb farmers hired a commercial applicator for field spraying. The mean frequency of application for weed killing was twice a year and for crop protection seven times a year.

3.1.2 Bulb disinfection

Dipping the bulbs into a cold or heated disinfection liquid is the most common method of bulb disinfection. Small dipping baths (200 l) have been used for many years and are still in use in the smaller farms. The drive-in vessels (2200 l) are increasing in number because they save manual labour. Because some varieties (hyacinth) demand a more careful treatment of the bulbs, sometimes bulbs are disinfected by showering. Both dipping and showering require barrels to contain the bulbs. Several types of barrels, of different size are used, e.g. baskets, crates (plastic and wooden) and gauze cases.

Because of its alleged ineffectiveness and the high incidence of mechanical damage to the bulbs, disinfection by spraying during planting is a technique of minor importance in the "Bollenstreek". Thirty-three percent of the farmers changed their bulb disinfection techniques during the last ten years. Most of them (27%) used the manual dipping technique before changing. For about 5% of the farmers bulb disinfection is carried out by others, mostly other farmers.

3.1.3 Stock protection

Depending on the pesticide, four different kinds of application were distinguished. Smoke pallets are used to fumigate with hydrogen cyanide, dichlorvos is applied by an aerosol dispenser, pyrimiphos-methyl is applied by thermal fogging and naphthalene crystals are sublimed by heating. At about 65% of the farms stock protection by hydrogen cyanide and pyrimiphos-methyl is performed with a mean frequency of 3 times a year by commercial applicators, mostly employees of the pesticide supplier. Naphthalene and dichlorvos are applied by the farmer. This hardly demands entrance of the storage cell.

3.1.4 Soil disinfection

Soil fumigants are injected into the soil through a nozzle mounted under an undercutter blade. Because of this special type of equipment soil disinfection is performed by commercial applicators. Some farmers (15%) do apply soil fumigants by spraying, to disinfect the cover soil of the bulbs.

3.1.5 Re-entry

Exposure to pesticides during other activities different than application (re-entry) is very likely in flower bulb cultivation.

Generally, bulb disinfection is performed just before planting, since this is the most effective. Depending on the technique of planting, contact of hands with the bulbs may be intensive during planting. For special varieties (e.g. hyacinth) or for flower production manual planting is necessary.

In spring, during the time of flowering, flowers are cut off, which is generally performed manually. Dermal exposure by transfer of foliar pesticide residues to the hands is therefore likely to occur. After lifting, the bulbs are sorted and graded. Released pesticide residues may cause both dermal and respiratory exposure. The latter stages of the production process demand a temporary hired workforce which must be considered as the exposed population. For the purpose of the estimation of long-term exposure of bulb farmers these exposures were not taken into consideration.

3.1.6 Conclusions

From the results of questionnaire #1 and the observations it was concluded that exposure of the farmers and their permanent employees was substantial during field spraying and bulb disinfection. Exposure during weed killing was supposed to be low compared to exposure during crop protection, since frequency of weed killing is limited and dose rate (in kg/ha a.i.) and volume rate (l/ha) is much lower respectively much higher than in crop protection, therefore other nozzles and pressure are used. So, in general, spray droplets during weed-killing are coarse compared to those generated during crop protection and contain less pesticides. Consequences of differences in exposure due to the use of different spraying techniques for exposure rating are incorporated in the estimation of exposure during crop protection since each individual farmer has been using a corresponding spraying technique for both crop protection and weed killing. Exposure of farmers during stock protection is considered to be negligible, since

duration of exposure is very short (spray dispenser application of dichlorvos) or the presence of the applicator during the application is not necessary (naphthalene vapour release). Soil disinfection is generally not carried out by the farmers, so exposure due to this type of application is neglected for the purpose of this study.

3.2 Characterization of exposure

3.2.1 Route of exposure

From the observations and the results of the questionnaire #1 (see 3.1) it was concluded that the assessment of exposure to pesticides for a part of the flower bulb farmers and their permanent employees can be limited to the estimation of exposure during crop protection and bulb disinfection. From reviews of data on exposure to pesticides it is evident that respiratory exposure is in the order of magnitude of 0.001 of the dermal exposure during spraying outdoors and mixing and loading of wettable powders (Turnbull et al., 1985; Joint Medical Panel, 1986; Van Hemmen, 1990).

Bulb disinfection will result mostly in dermal exposure of the hands to pesticides in the disinfection liquid, or by transfer of pesticide residue from the surface of the bulbs or contaminated barrels to the hands. The bulb disinfection techniques have been ranked according to exposure level (see appendix II). It was concluded from this ranking that the dipping-bath technique using baskets results in the highest potential dermal exposure, significantly higher than for the other techniques, since this process of disinfection is purely manual. So far as exposure is concerned, all other techniques can be taken together. The drive-in vessel technique is considered to be a good representative of all other techniques for exposure assessment. It seems most sensible to use dermal exposure as a criterion for discrimination or ranking of the selected population according to exposure due to spraying and disinfection.

3.2.2 Potential and actual exposure

Potential exposure can be defined as the total amount of pesticide deposited on body and clothes. The estimated actual exposure, i.e. the estimated amount of pesticide deposited on the skin, depends on the measure of protection by clothing, protective gloves, etc. The UK data base (Joint Medical Panel, 1986) contains data on the distribution of external contamination experienced by operators during spray application. Sixty-five percent of the total contamination due to tractor boom spraying was located on the hands, the corresponding value for backpack spraying was 25%. From the literature it was estimated that the major contamination due to mixing and loading (45-75%) was located on the hands (see section II) and similar conclusions were arrived at exposure due to bulb disinfection. The actual deposition of pesticides was mainly on the hands, although small amounts may reach head and neck. So it was decided to use exposure of the hands to compare exposure levels due to the use of different kinds of technique.

Forty-nine percent of 127 workers participating in the health effect study reported to use gloves during mixing and loading in 1988, but only 27% did so in 1983.

Other results of the questions about personal protection are listed in table 3. From these results and the lack of adequate data on the overall efficiency of protective gloves in practice (see section II) it was concluded that for the estimation of long-term exposure protection by wearing gloves could not be taken into account and potential exposure of the hands was considered to give a reasonable estimate of actual exposure.

In table 4 the pesticides used in crop protection and bulb disinfection are ranked according to their relative contribution to the total amount of pesticides applied per hectare of bulb acreage (Liem and De Groot, 1984). Besides, the prevalences of use at the farms according to the results of questionnaire #1 are given. It was concluded that for the cultivation of flower bulbs zineb/maneb was an appropriate representative compound for assessment of exposure during crop protection and bulb disinfection, since this mixture is used in large amounts for both.

3.2.3 Method-specific exposure levels

Method-specific exposure levels were calculated for tractor boom spraying. These calculations were based on the UK data base of exposure levels and adjusted to an average application dose of zineb/maneb of 2.1 kg/ha a.i. (SD: 1.1 kg/ha; n= 42), as observed during the mixing and loading study (see Section II) and derived from log-book records). The application rate of the different spraying techniques was calculated: the mean application speeds for tractor boom sprayings equipped with a 600 l or 400 l tank were 2.6 and 1.3 ha/hr, respectively, and for backpack spraying with a 15-18 l tank and bike spraying with a 17 l tank 0.26 ha/hr and 0.34 ha/hr, respectively. Details are given in Appendix I. The volume rate was 200 l/ha, except for the tractor boom sprayings equipped with a 400 l tank which applied 400 l/ha. Calculated concentrations a.i. in the spraying liquid were 5.25 g/l for the 400 l/ha volume rate and 10.5 g/l for the 200 l/ha volume rate.

From the raw data of the UK data base both geometric and arithmetic means and the corresponding standard deviations were calculated. The number of data sets and details on the calculations are presented in Appendix I. Subsequently, potential exposure of the hands per hectare was calculated for tractor boom spraying and backpack spraying. The results of these calculations are summarized in table 5. The 95% confidence interval of the geometric mean is presented as well. Since the method-specific exposure level due to bike spraying is based on a limited number of data the range is presented and no 95% confidence interval was calculated. It is seen that the confidence interval (based on GM) of the exposure level of tractor boom spraying is relatively small compared with that of backpack spraying. Exposure levels for mixing and loading methods were calculated from the mean dermal exposure levels assessed in the field study (section II). Transformed levels (mg/kg a.i.) were adjusted for the mean application dose (2.1 kg/ha a.i.) giving levels per unit of acreage (mg/ha). In order to compare exposure due to mixing and loading of wettable powders and that of liquid formulations, transformed UK data are also presented. Details are presented in Appendix I. The results indicate that the use of liquid formulations will reduce exposure substantially. Method-specific exposure levels for the two bulb disinfection techniques were

calculated from the results of the field study. They are expressed as exposure per barrel instead of exposure per bulb acreage, since bulb disinfection is not only performed in bulb cultivation but also for the cultivation of bulb flowers, and the number (and weight) of the bulbs planted per hectare of farming land varies. The weight ranges from 8000 (crocus) to 21000 (daffodil) kg/ha. Examples of exposure levels for different methods and bulb varieties are given in table 5.

The large variance of the exposure levels and the small numbers of samples (see section II) is reflected in the large 95% CI's of the exposure levels of the bulb disinfection methods.

3.3 Exposure estimation of the participants for the health effect study

For 134 participants, farmers and employees, in the health effect study (for description of selection, see Part I of this series of reports), data on exposure were obtained by questionnaire #3. The mean number of years of exposure was 20 (SD: 7). Seven participants did not meet the selection criterion of the health effect study on exposure, i.e. being involved in application of pesticides for both field spraying and bulb disinfection for at least ten years prior to the study. Two workers were only involved in field spraying and five exclusively performed bulb disinfection. So 127 data sets on spraying and bulb disinfection over a period of 10 years, i.e. 1270 "spraying years" and 1270 "bulb disinfection years", were available for analysis.

3.3.1 Description of methods of application and mixing/loading

Spraying

In the 1270 spraying years considered, 54% of the bulb acreage was sprayed using tractor boom spraying, 17% using bike spraying and 12% using backpack spraying. In 17% of the years, combinations of techniques were used, mostly tractor boom spraying and backpack spraying (13%).

Both acreage and frequency of field spraying differed significantly between the spraying techniques (Mann-Whitney, $p < 0.05$). Details are given in table 6. Seventy-five farmers (59%) exclusively used tractor boom spraying, and 68 of them (54%) have been using this technique during the last ten years. Corresponding figures on bike spraying and backpack spraying are on 29 farmers of which 15 for more than ten years and 20 farmers of which 10 for more than 10 years, respectively.

Mixing and loading

Direct tank filling was used as method of loading in 20% ($n = 252$) of the considered 1270 years, by 32 farmers (25%). Twenty of them have used this method for more than ten years. Only two of them used bike spraying for field spraying, the others tractor boom spraying. In 52% of the spraying years the spraying liquid was prepared by premix-scooping. Seventy-one workers (45%) used this method and 58 of them (37%) used it for more than ten years. In about 28% of the spraying years the premix-pouring method was used by 40 workers (31%).

Thirty-one workers (24%) used it for more than 10 years. The figures on the current prevalence of the spraying techniques correspond well with the results from questionnaire #1 (see table 3).

Bulb disinfection

In about 45% of the 1270 disinfection years bulb disinfection was performed for growing of bulb flowers. Other techniques than manual dipping, e.g. drive-in vessel and showering, were used by 77 farmers (61%) and 53 of them (41%) for more than ten years. Fourteen out of 30 farmers have been using the manual dipping technique for more than 10 years. The prevalence of manual dipping (39%) corresponds well with the result of questionnaire #1. Twenty-two farmers using the manual dipping method did so exclusively for production of flower bulbs. In this case bulb acreage correlated only moderately ($r = 0.52$) with calculated exposure due to bulb disinfection, because the weight per hectare differed for different bulb varieties.

Summary

Field spraying was mostly performed using tractor boom sprayers. Both bulb acreage treated and frequency of spraying differ significantly between this and other spraying techniques. The backpack spraying is generally used in addition to tractor boom spraying. Sixty-five percent of the farmers did not change their spraying techniques within the last ten years.

Premix-scooping is the most popular method of preparing the spraying liquid. Only a few workers changed their method of mixing/loading during the considered period of ten years.

Total exposure due to bulb disinfection does not correlate well with bulb acreage, probably because the treatment of bulbs for the production of bulb flowers is in some cases done besides that for flower bulb production. For disinfection for flower bulb growing alone, the correlation between bulb acreage and exposure was only moderate because of differences in weight of bulbs per acreage for different bulb varieties.

3.3.2 Relative contribution of different applications to total exposure

Exposure due to crop protection was usually a dominant factor during the exposure period of ten years considered and range for the 127 farmers from 3 to 99% (median: 86%) of the total exposure. Mixing and loading contributed substantially to exposure during crop protection. It amounted from 5 up to 97% (median: 80%). The contribution of mixing and loading to total exposure ranged from 2 to 97% (median 60%).

Most of the variation of total exposure, i.e. the exposure index, could be explained in a multiple regression model by the variation of exposure due to mixing and loading (simple $r^2 = 0.83$). The variation of exposure due to mixing and loading as analysed for the group of tractor boom sprayers was dominated by the method of loading and mixing since the other contributing factors, bulb acreage and frequency of loading, only explained 36% of the variation (r^2 sequential = 36%).

3.3.3. Reliability and sensitivity

The exposure index used for the assessment of occupational exposure to pesticides does not reflect the absolute individual exposure of the farmers but ranks them. The reliability and sensitivity of the ranking depends on the reliability and validity of the calculated method-specific exposure levels and of the self-administered questionnaires. Some preliminary investigations into the sensitivity of the ranking of the studied population by exposure index to variations in the method-specific exposure levels were done by correlation analysis. Lower or upper 95% confidence limits (CI) of the method-specific exposure levels of these techniques were used to recalculate of the exposure index. With one exception ($r = 0.87$), all resulting indices correlated very well with the initially calculated exposure index ($r = 0.91$). Also a high correlation ($r = 0.91$) was observed between exposure indices using method-specific exposure levels based on geometric and arithmetic means. The above results indicate a relative insensitivity to variations and an ability of close ranking on the level of the studied population. Secondary analyses of the data, using kappa statistics and simulation techniques, seems to confirm this conclusion.

Not all specific exposure levels were obtained by exposure assessments in our own field studies, but calculated (see Appendices I and II), and an appropriate calibration was not available. Calibration could possibly be made with other techniques of assessment of dermal exposure and with adequate biological monitoring methods. These are presently not available.

By considering a representative compound, it is implicitly assumed that the use of the different pesticides (the pesticide "profile") for crop protection and bulb disinfection has been equal for all individual farmers throughout the years. According to experiences of the Flower Bulb Research Centre, the majority of the farmers has followed their advices on pesticides very strictly. Therefore, in view of the data in table 4, the use of a compound in exposure assessment is quite acceptable when no data are required on specific substances.

The exposure indices were calculated for zineb/maneb. However, there may have been participants in the study who actually did not use zineb/maneb for bulb disinfection. Captan, a frequently used alternative, is used at the same concentration in bulb disinfection liquid. So this will not result in a different ranking.

The answers to the self-administered questionnaires form another important aspect for the reliability of the exposure index. Most studies on validity of self-reported work history, e.g on job title and job duration, report a good agreement between company records and information obtained by interviewing the employees (Bourbonnais et al., 1988; Kongerud et al., 1989) and no significant differences in the validity on information of job related events over recent (2-8 years) and remote (9-15 years) periods of time (Baumgarten et al., 1983).

Data on methods of field spraying and bulb disinfection, acreage of farming land, number of applications (field spraying) and number of disinfected barrels over a period of ten years are very important for the construction of an exposure index. A high correlation was found between answers to comparable questions on techniques of spraying during the year 1987 of the questionnaires #1 and #3 ($r = 0.9$). Unfortunately, very few questions on this subject were asked in the same way,

in both questionnaires e.g. frequency of field spraying was asked per bulb variety in questionnaire #1 but unspecified in questionnaire #3.

It is remarkable that about 75% of the participants of questionnaire #3 reported the same number of field sprayings during the last ten years. This might be an indication of an inaccurate figure for a particular year, since frequency partly depends on weather conditions, but might be an accurate estimation of the mean of ten years. If a farmer had changed spraying technique during that period, exposure estimation will be less reliable.

The reliability of a lifetime exposure index, calculated by extrapolating the EI backwards from 1979 may be influenced by information bias. No details on techniques and acreage are known for the years before 1979. For a subset of 102 farmers it is known that 15 (15%) changed their spraying technique before 1979. The average number of years worked with the previous technique was 13 (range 2-24; median 13). For bulb disinfection corresponding figures are: 5 farmers changed their technique before 1979, the average number of years working with the previous technique was 12 (range 2-16; median 14).

The correlation between total number of years of exposure and the calculated exposure index over that period of exposure is low ($r = 0.45$). This is considered an indication that the use of application techniques and working methods are important aspects in long-term exposure assessment.

3.4 The use of the exposure index as a surrogate cumulative exposure for the health effect study

For all participants long-term exposures were estimated considering several years of exposure previous to the year of the study, i.e. five years, ten years and lifetime workperiods. For the health effect study a lifetime exposure is used, based on method-specific exposure levels derived from arithmetic mean exposure values and calculated by extrapolation of the exposure index from 1979 backwards. These lifetime exposure indices ranged from 0.25 to 2200 (based on grams of zineb/maneb $\times$ years of exposure; median 200; 90-percentile: 770). Figure 2 illustrates the distribution of the lifetime exposure index. Coefficients of correlation between five years, ten years and lifetime exposure indices were relatively high ($r > 0.8$) and kappa values were satisfactory since above 0.4. The data are summarized in table 7. These results indicate a low within-person-variation of the exposure (variables) for the years considered.

The process which may lead to the occurrence of health effects can be described in the following terms: exposure, absorption (or penetration), metabolism, distribution, excretion and toxic effects. In order to determine the optimum means of quantifying exposure for epidemiological studies, much consideration has been given to the use of "dose" and related ideas "borrowed" from toxicology. The analogue of total dose for occupational exposure is "cumulative exposure", defined as average exposure intensity times duration of exposure. A relationship between exposure and effects is based on the premise that exposure of the worker will

cause an effect if the exposure produces an increase of the agent (or the active metabolite) in the sensitive tissues for a sufficiently long period of time. The approach of "cumulative exposure" has the important shortcoming of not dealing adequately with the variability of exposure with time, which will produce a variable dose rate. In flower bulb farming exposure during field spraying occurs about 7 times a year (up to 2 days per spraying, within a period of about four months). On the other hand exposure due to bulb disinfection is on several subsequent days in a period of a few weeks. Knowledge of toxicokinetics (like skin absorption rate) and toxicodynamics is therefore important for the choice of an exposure assessment strategy (Smith, 1987). Since such knowledge is lacking for the present, possibly neurotoxic pesticides (see parts 3 and 4 of this series of reports) the exposure index, calculated as a lifetime exposure index, may be used as a "surrogate cumulative exposure", with the implicit assumption of a linear dose-response relationship. Since it has been demonstrated by Seixas et al. (1988) that the cumulative exposure based on the arithmetic mean is the "true" or correct measure for such a relationship, the index was calculated using method-specific exposure levels based on arithmetic means. A linear exposure-dose and thus a linear exposure-response, however, may be disputed since it is very doubtful that uptake will depend linearly on the amount of pesticide present on the skin. The application of the exposure index in an explorative study may however be helpful in the formulation of hypotheses for further studies. The exposure index itself provides the opportunity for adequate risk assessment of a specific compound if necessary corrections are introduced for toxicokinetics and toxicodynamics.

Conclusions and recommendations

From the results of this study on the estimation of long-term exposure to pesticides due to the application of these compounds the following can be concluded.

- In general, methods / techniques of application and of mixing and loading with relatively low and relatively high exposure can be distinguished. Reduction of exposure can be reached by the use of techniques resulting in relatively low levels of exposure.
- Mixing and loading of wettable powders for field spraying contributes appreciably to total exposure. So reduction of this type of exposure is of primary importance in reducing total exposure. Calculated exposures indicate a significant reduction by substitution of liquid formulations for wettable powders.
- The exposure index may provide an adequate tool for ranking populations according to exposure, but validation by other methods of exposure assessment is needed.
- Application of the exposure index as a surrogate dose for dose-response relationships, in a health effect study, without sufficient knowledge on toxicokinetics and toxicodynamics has considerable limitations.

The following goals for further research are recommended on the basis of the presented results and conclusions.

- Comparison of ranking according to estimated dermal and respiratory long-term exposure.
- Comparison of respiratory and dermal exposure in relation to absorbed dose in agricultural practice.
- Exposure studies under other circumstances of exposure to pesticides in the flower bulb culture, e.g. during re-entry.
- Estimation of the effectiveness of personal protective equipment under field conditions for reduction of exposure.

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Table 1 Synopsis of pesticides used in flower bulb farming

Field of application	Major use	Period ¹	Minor use	Period ¹
weed killing	chloropropham	since 1955	chloridazon	since 1955
	paraquat	since 1968	diquat	since 1968
	glyphosate	since 1983	amitrol	since 1972
			MCPA	since 1975
			2,4 D	since 1975
crop protection	bordeaux-mixture	between 1945 and 1960	benomyl	since 1978
			vinchlozolin	since 1980
	zineb/maneb	since 1960	carbendazim	since 1980
			synth. pyrethroids	since 1983
bulb disinfection			procymidon	since 1980
	organic mercury compounds	between 1945 and 1976	zineb/maneb	since 1980
	captafol	between 1976 and 1982	benomyl	since 1982
			carbendazim	since 1982
	captan	since 1982	thiophanate- methyl	since 1982
stock protection	formaldehyde	since 1976	prochloraz	since 1982
	hydrogen cyanide	since 1950	dichlorvos	since 1980
	pyrimiphos-methyl	since 1986	naphthalene	
soil disinfection	1,3-dichloropropene	since 1965	etridiazole	since 1980
	metam-sodium	since 1965	formaldehyde	since 1980

¹ Approximation

Table 2 Summary of the prevalence of application techniques for field spraying and bulb disinfection on farms in the "Bollenstreek"

Field of	Field spraying ¹ application	Bulb disinfection ²		
Method of application	spraying (100%)	dipping (84%)	showering (13%)	spraying (3%)
Application technique	tractor boom spraying (68%)	drive-in vessel (48%)		
	backpack spraying (45%)	dipping bath (36%)		
	bike spraying (22%)			

¹ Results questionnaire #1 (n= 295)

² Results questionnaire #2 (n =105)

Table 3 Percentage of farmers reporting the use of personal protective equipment

Equipment	Field spraying		Mixing and loading		Bulb disinfection	
	1988	1983	1988	1983	1988	1983
Glove	37	23	49	27	63	47
Respirator	31	7	32	10	-	-

- Not considered

¹ Results questionnaire #3

Table 4 Ranking of pesticides according to the amount applied (kg/ha/yr) and prevalence of use on farms in the "Bollenstreek"

Field of application	Pesticide	Amount ¹ [%]	Prevalence ² [%]
<i>Crop protection</i>	zineb/maneb	90	>95
	vinchlozolin	2	80
	procymidon	2	50
	benomyl/carbendazim	1	60
	synthetic pyrethroids	<0.5	50
<i>Bulb disinfection</i>	zineb/maneb	40	50
	captan	20	35
	formaldehyde	20	60
	benomyl/carbendazim	4	50
	procymidon	3	40
	prochloraz	2	50

¹ According to Liem and de Groot (1984)

² Results questionnaire #1

Table 5 Method-specific exposure levels

Technique		Exposure [mg/ha]	
Crop protection	AM ¹	GM ²	[CI95%] ³
tractor boom spraying	28	9	8-10
backpack spraying	850	320	130-800
bike spraying	700	(150-1300) ⁴	
Mixing & loading			
direct tank filling	45	36	27-48
premix-pouring	126	97	67-137
premix-scooping	482	294	197-437
(liquid)		27	
Bulb disinfection			
dipping bath/baskets			
[mg/barrel]	2.4	1.5	0.5-4
crocus		345	120-1000
daffodil		900	315-2650
Others			
[mg/barrel]	0.8	0.6	0.1-4
crocus		9	1-66
daffodil		23	3-197

¹ Based on the arithmetic mean of exposure levels

² Based on the geometric mean of exposure levels

³ 95% Confidence interval of the geometric mean

⁴ Range

Table 6 Bulb acreage and frequency of field spraying (times/year)

	Tractor boom spraying		Bike spraying		Backpack spraying	
	range	median	range	median	range	median
Acreage(ha)	0.4-43	5.7 ^{1,2}	0.1-4	1.4 ¹	0.07-3	1 ²
Frequency	1-16	10 ^{3,4}	1-10	6 ³	1-12	6 ⁴

^{1,3} Significant difference between tractor boom spraying and bike spraying (Mann - Whitney ($p < 0.05$))

^{2,4} Significant difference between tractor boom spraying and backpack spraying (Mann - Whitney ($p < 0.05$))

Table 7 Agreement and correlations of exposure indices calculated for 5 years, 10 years and lifetime exposure periods (n=134)

Spearman / Kappa	El 5 year	El 10 years	El lifetime	Years of exposure
El 5 year	-			
El 10 years	0.92 / 0.67	-		
El lifetime	0.80 / 0.47	0.88 / 0.54	-	0.45/ -

Figure 1 Diagram of the process of flower bulb growing and the application of pesticides

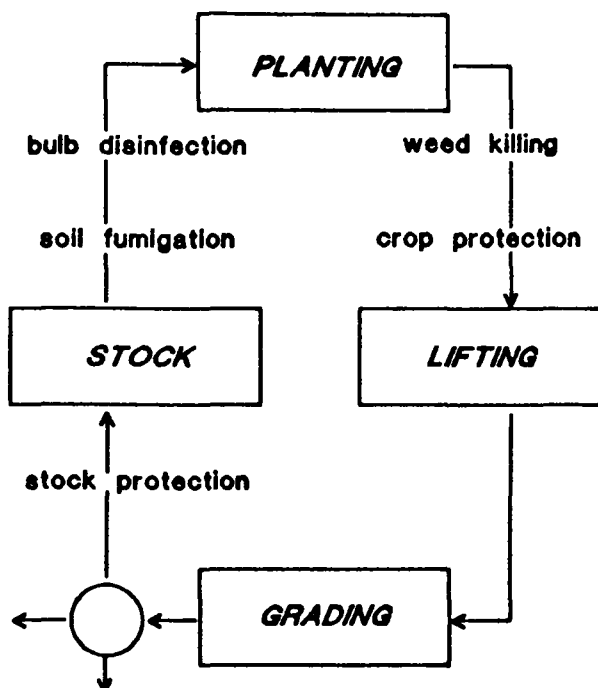
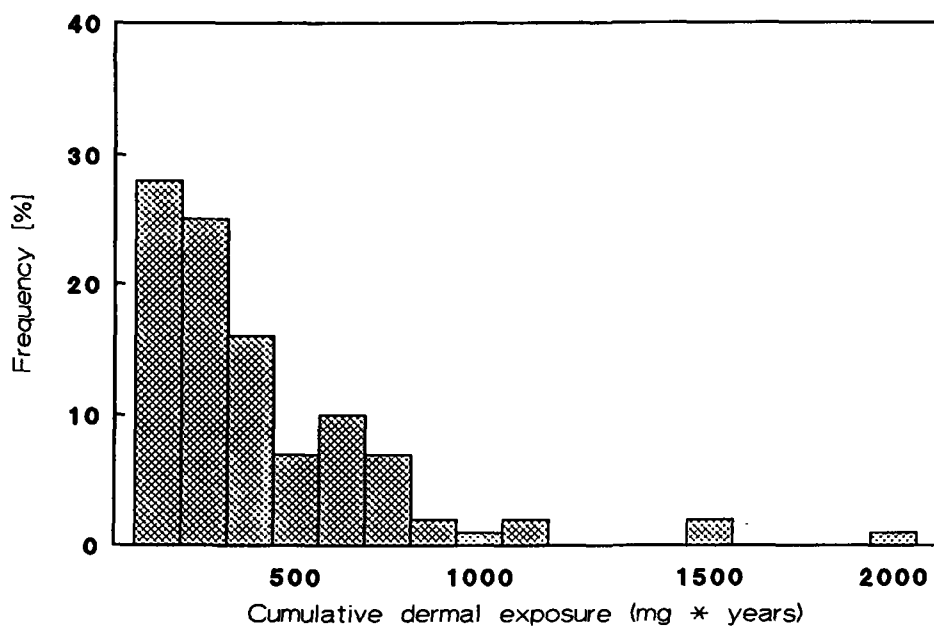


Figure 2 Frequency distribution of lifetime exposure indices (n=134), For details see text



Section 2 Assessment of exposure to zineb/maneb

Introduction

In the flower bulb culture the fungicide mixture zineb/maneb has been the major pesticide in crop protection for many years. Because this mixture is often used in bulb disinfection as well (see section 1, table 5), it was chosen as a representative pesticide for the estimation of exposure of flower bulb workers to pesticides. Method-specific exposure levels were required for several spraying techniques, for methods mixing and loading and for bulb disinfection techniques.

It has been widely accepted that exposure to pesticides is dependent on physical factors rather than on the chemical properties of these compounds. So data on exposure to a particular pesticide can to some extent be extrapolated to other pesticides with similar physical properties (Van Hemmen, 1990a). A UK data base (Joint Medical Panel, 1986) provides exposure data for spraying techniques. However, no adequate data on potential dermal and respiratory exposure during mixing and loading of wettable powders could be obtained from the literature, since most published exposure data differ with respect to method of exposure assessment and data on the amount of loaded active ingredient and on working methods are often not available (Van Hemmen, 1990b). Since data on exposure during (bulb) disinfection are lacking completely, a field study was conducted to obtain exposure levels for relevant methods of mixing and loading and for bulb disinfection techniques.

Since both the rate (2.1 kg/ha a.i.) and the average frequency (about 7 times per year) of application of zineb/maneb in crop protection are high, in most cases the relatively inexpensive formulation and packaging (10 kg bag of wettable powder) are used. A liquid formulation has been introduced in 1986, which is used mostly for bulb disinfection. Kangas et al. (1980) reported dust concentrations of maneb in the breathing zone during mixing and loading from 1 and 50 kg bags, whereas Nilsson and Nygren (1987) did the same for loading (probably by direct tank filling) of a wettable powder formulation of mancozeb. Mumma et al. (1985) reported on both dermal and respiratory exposure to mancozeb, but details about loading methods were not given. The reported dermal exposure of the hands during the mixing and loading of wettable powders ranged from about 45% (Everhart and Holt, 1982) to about 75% (Maitlen et al., 1982) of the potential total body exposure. The UK data base reports that exposure during mixing and loading is "virtually confined to the hands".

Both respiratory exposure and dermal exposure of the hands during mixing and loading of a wettable powder formulation have been investigated for the three most popular methods.

- Loading without pre-mixing with water, i.e. direct filling of the tank from the bag; this method is restricted to the use of tractor boom sprayers equipped with a 600 l tank (method 1).
- Loading with pre-mixing in a bucket; within this group two different methods for loading the bucket with the formulation were distinguished :
 - dumping or pouring (a part of) the contents of the bag into a bucket (method 2);
 - scooping (a part of) the contents of the bag into a bucket (method 3).

Bulb disinfection is an important aspect of bulb protection, since most bulb varieties are susceptible to diseases. Disinfection has proven to be very effective when performed immediately before planting. The bulbs are disinfected (with or without barrels) by dipping, showering or spraying. The disinfection liquid is prepared from 1, 5 or 10 l cans; refilling of dipping baths occurs generally one or two times a day as 1 kg of bulbs absorb 1 l of disinfection liquid. From field observations it was learned that dermal exposure was largely limited to the hands, due to contact with the disinfection liquid and the contaminated equipment during manipulation of the barrels.

Respiratory exposure was considered to be very low or even negligible for almost all disinfection methods. A similar assumption was made for respiratory exposure during the preparation of the disinfection liquid, because of the use of liquid formulations.

All observed disinfection methods, with different kinds of barrels, were rated according to the estimated potential dermal exposure (details are given in Appendix II). Manual dipping using baskets, which was the most common technique in the past, was expected to give the highest exposure, whereas the more sophisticated process using drive-in vessels was expected to give the lowest. Therefore, these two techniques were selected for the assessment of dermal exposure of the hands to zineb/maneb.

Materials and methods

2.1 Population and sampling strategy

Mixing and loading

Twelve farmers applying direct tank filling (method 1) and twelve using premix methods (2 and 3) were randomly selected from the population of flower bulb farmers which were willing to participate in the health effect study (n=272). For at least two tank filling operations both respiratory and dermal exposure of each farmer to zineb/maneb were estimated. The sampling period started at the beginning of the handling of the formulation and ended after finishing loading. Those farmers who used to wear protective gloves were asked to remove them and vinyl disposable examination gloves (TriflexTM) were available to wear underneath cotton gloves (see 2.2.2). In these cases the farmers were also asked to wear cotton gloves underneath their protective gloves during a subsequent mixing/loading operation in order to obtain some preliminary results on the effectiveness of protective gloves in reducing dermal exposure under field conditions.

Bulb disinfection

For the estimation of dermal exposure during bulb disinfection 33 farmers (13 for the drive-in vessel and 20 for the dipping-bath) were randomly selected from 105 respondents to a questionnaire (#2, see Section I) on bulb disinfection techniques using zineb/maneb for bulb disinfection.

The sampling period included at least a series of five disinfection cycles, i.e. from the introduction of the barrel into the bath or vessel until removal of the barrel after a period of about 15 minutes. Moreover, farmers who said to use protective gloves occasionally were asked to use their gloves during one cycle, in order to obtain some preliminary results on the effectiveness in reducing dermal exposure under field conditions.

2.2 Exposure assessment

2.2.1 Respiratory exposure

Respiratory exposure during mixing and loading was assessed by personal air sampling, using a personal air sampling pump (Dupont P-2500, E.I. du Pont de Nemours & Co, Wilmington, Del, USA) operating at a flow of 2 l.min⁻¹ and an IOM-sampling head (Negretti Automation, Aylesbury UK), with a 25 mm GF-filter

(Gelman Sciences Inc., Ann Arbor, MI, USA). The pesticide concentration measured near the breathing zone of the subjects was considered to reflect the inspirable fraction according to the ACGIH definition of inspirable dust (Vincent and Mark, 1987). Seven field duplicates yielded a relative standard deviation (CV analyses + sampling) of 31%. The flow rate was checked before and after sampling, using a pre-calibrated rotameter (Rotal, Dr Henning GmbH, Wehr/Baden, FRG). After sampling the filter was removed from the sampling head, transferred to a petri-dish and stored dark and cool (- 20 °C).

2.2.2 Dermal exposure

Potential dermal exposure during mixing and loading was assessed by sampling with gloves. Farmers were asked to wear cotton gloves (cotton-stretch 200 g/cm², v.d. Wee, Riel, The Netherlands) covering hands and wrists (about 1000 cm²). The recovery of zineb/maneb from the gloves was 98%. After sampling the gloves were carefully stripped off, transferred to a polyethylene storage bag (zip-lock) and stored dark and cool (- 20 °C).

Dermal exposure during bulb disinfection was assessed by a hand-washing method. After a series of disinfection cycles workers were asked to wash both hands in a polyethylene bag containing 500 ml 0.1 M EDTA. This was repeated once. After washing, the bags were closed and transported to the laboratory. The samples were analysed within 24 hours.

In a study with five volunteers the hands were spiked with 0.5 ml of a 1% homogeneous suspension of a commercial zineb/maneb formulation (TridexTM 455 g/l, Hoechst, Amsterdam, The Netherlands). After 15 minutes both hands were washed as described above. The recovery of zineb/maneb from the hands by this method was found to be 81% (SD=10%).

During mixing and loading operations climate conditions were recorded, i.e. wind speed by a thermal anemometer (Alnor, GGA 65, Turku, Finland), and temperature and relative humidity by a thermo-hygrometer (type L 0680, Thies, FRG).

2.3 Chemical Analyses

Filters

Filters were added to a headspace vial with 2.5 ml of 1.5% Tin(II)chloride in 5 M hydrochloric acid. After equilibration at 70 °C for 45 minutes, the samples were injected into the gas chromatograph and the liberated carbon disulfide was detected by electron capture.

Gloves

Gloves were transferred from the polyethylene bags to glass bottles and 300 ml of a 0.1 M EDTA was added. The bottles were shaken for about 1 hour at a frequency of 200 strokes per minute. Subsequently, the solution was sonicated for 15 minutes. Finally, 1 ml of the solution was added to a headspace vial containing the same solution as used for the analysis of filters.

Hand-washing

The polyethylene bags were shaken by hand for several minutes. Subsequently 1 ml of the solution was added to a headspace vial containing the same solution as used for analysis of filters.

Limits of detection and stability of the samples

The stability of filter samples was tested by separate analysis of two equal parts of samples obtained in the field. The first part was analysed within 24 hours after sampling, the second part after 4 days. No differences were found. The limit of detection on a filter was 20 ng active ingredient.

During a period of about three weeks a zineb/maneb formulation (TRIDEX™) was stable in the hand-washing solution when stored in the dark at room temperature. The limit of detection for hand-washing was 24 µg active ingredient.

During a period of 26 days no reduction of zineb/maneb was observed on spiked gloves stored in the dark at -20 °C (n=36). The limit of detection for one pair of gloves was 6 µg active ingredient. The coefficient of variation of all types of analysis was less than 6%. Further details are given by Ravensberg (1989).

2.4 Statistical analyses

Normality of a distribution of data was tested by the Shapiro and Wilk test and differences of exposure levels between methods were tested non-parametrically by analysis of variance (Kruskal-Wallis). After log-transformation of the data the influence of environmental parameters was tested by simple linear regression.

Results and discussion

3.1 Mixing and loading

One farmer withdrew from the study for unknown reasons and there were several misclassifications. Within the group using direct tank filling (method 1) two farmers used a different way of filling. One of them poured a part of the contents of the bag into a bucket, which he subsequently emptied into the tank. The other farmer filled the tank by scooping the powder from the bag into the tank. In the pre-mixing group (methods 2 and 3) three farmers used a different method for mixing powder and water. Instead of stirring with a stick they used a waterhose. The exposure data of these five farmers using other methods were excluded for the purpose of this study. So, finally, the data of eighteen farmers were available for analysis.

3.1.1 Dermal exposure

The exposure data were log-normally distributed. Both within and between variance was high and there were no statistically significant differences between exposure levels within each method, which indicates independent samples. Within the group filling the tank directly (method 1) five farmers were sampled during three tank filling operations and two farmers during two. The operation times ranged from 1.5 to 4.8 minutes (mean 3 minutes). Pesticide contamination of the glove ranged from 29 to 300 mg (a.i.). The amount of loaded active ingredient in one operation ranged from 4 to 8.75 kg (median 7 kg). Details are given in Appendix III.

Within the premix-pouring group (method 2) five workers were sampled three times and one worker twice. Glove loadings ranged from 14 to 360 mg a.i. The average loading time was 4.8 minutes (range 3.8 - 8.5 minutes). The median of the amount loaded was 2.7 kg a.i. (range 0.6 - 6.1 kg).

Five farmers were sampled during premix-scooping (method 3) three during three and two during two operations, and one farmer was sampled twice during scooping without pre-mixing. The operation times ranged from 2.8 to 4.8 minutes (mean 3.4 minutes) and glove loadings ranged from 53 to 2300 mg a.i. The mean amount loaded was 2.0 kg a.i. (range 1.2 - 2.4). Since the amount of active ingredient varied, the exposure levels were transformed into exposure per kilogram a.i. handled. The transformed exposure levels differed significantly between the methods. Premix-scooping (method 3) resulted in the highest levels and direct tank filling (method 1) in the lowest. Table 1 summarizes the results. Exposure levels during application of other methods (see 3.1) ranged from 70 to 3800 mg. The highest transformed

exposure levels were observed during stirring the premix with a waterhose (1100 mg/kg a.i.).

The premix operations (methods 2 and 3) involve two activities which potential exposure: (1) mixing and (2) transfer of the pesticide from the bag into the bucket. Since dermal contact with the bag during scooping is substantial, these operations resulted in the highest dermal exposures. Exposures due to direct tank filling operations fit well within the range of exposures due to the premix methods (methods 2 and 3), but differ significantly from these when exposure data were transformed into exposure per amount of loaded active ingredient.

Maitlen et al. (1982) reported two data on exposure during scooping of carbaryl, a wettable powder: 38.3 and 99.1 mg (hand-washing) per operation, which correspond well with the results of this study. A few more data are available on direct tank filling: Everhart and Holt (1982), using pads, reported a median exposure of the hands to benomyl wettable powder of 7 mg (a.i.), but these data cannot be transformed into exposure per kg a.i. loaded. Results of other studies, reviewed by Van Hemmen (1990b), show much lower exposures.

Some preliminary results on the level of protection afforded by gloves were obtained by sampling underneath protective gloves. The actual exposure ranged from 2 to 22 mg per operation, i.e. 1 to 6% of the exposure without gloves.

3.1.2 **Respiratory exposure**

The exposure data were log-normally distributed. Both within and between variance was high and there were no statistically significant differences between exposure levels within each method which indicates independent samples. Within the premix methods a subset was discerned: pre-mixing indoors, inside the barn (method 4). The results are listed in table 2.

Exposure levels during direct tank filling operations (method 1) ranged from 42 to 14700 $\mu\text{g}/\text{m}^3$, with a geometric mean of 675 $\mu\text{g}/\text{m}^3$. Wind speed during loading operations ranged from 1 to 7 m.s^{-1} (median 4 m.s^{-1}) and temperature from 15 to 26 °C (median 16 °C).

Within the premix-pouring group (method 2) the geometric mean of the concentrations was 294 mg/m^3 (range 61 to 1770 $\mu\text{g}/\text{m}^3$). Concentrations during mixing and loading by premix-scooping (method 3) ranged from 36 to 580 $\mu\text{g}/\text{m}^3$ (geometric mean 139 $\mu\text{g}/\text{m}^3$). Wind speed ranged from 1 to 5 m.s^{-1} (median 3 m.s^{-1}) and temperature from 11 to 25 °C (median 18 °C). The relative humidity was between 35 and 80%.

Pre-mixing indoors resulted in a geometric mean concentration of 2984 $\mu\text{g}/\text{m}^3$ (range 507 - 18200 $\mu\text{g}/\text{m}^3$). Wind speed inside the barn did not exceed 1 m.s^{-1} . To relate the data with the amount of active ingredient loaded (see 3.1.1), the exposure data were also transformed into concentration per kilogram of loaded active ingredient (a.i.), as shown in table 2. Although differences in respiratory exposure are observed between all methods, only those between pre-mixing outside (premix-pouring (method 2) and premix-scooping (method 3)) and pre-mixing indoors (pouring and scooping (method 4)) were statistically significant. Kangas et al. (1980) reported mean stationary air concentrations of maneb of 1.3 mg/m^3 when using 1 kg bags and 6.7 mg/m^3 when using a part of the contents of

50 kg bags. Nilsson and Nygren (1987) reported data on exposure levels by personal air sampling during loading a wettable powder formulation of mancozeb, probably during direct tank filling. The geometric mean was 0.22 mg/m³ (n=18). These levels correspond well with the present data on pre-mixing and are within the range of data on direct tank filling. The large variation reported is similar to that within the group of direct tank filling operations (method 1) (Geometric Standard Deviation (GSD): 4.1). This relatively large variation in the present study can be explained partially by environmental conditions. By simple linear regression analysis the variable wind speed appears to explain about 30% of the variation ($r^2 = 0.28$). The high concentration in the breathing zone during pre-mixing indoors (method 4) may be related to the poor natural ventilation in the barn (wind speed < 1 m.s⁻¹).

3.2 Bulb disinfection

A couple of weeks before the start of the field study, the Flower Bulb Research Centre (LBO, Lisse, The Netherlands) published a study on the (in)effectiveness of zineb/maneb for the disinfection of a number of bulb varieties. Due to this publication many farmers started to use another pesticide (mostly captan) instead of zineb/maneb. This reduced the population by fifteen farmers and the number of farmers that could be studied became very small. Moreover, six farmers withdrew from the study for several reasons (business, illness) and one farmer used another kind of barrel. Finally, dermal exposure of 11 farmers was measured. Two farmers used protective gloves during the whole period of disinfection, so for analysis of exposure data of bare hands nine farmers were available.

The dermal exposure of six farmers was measured after disinfection of bulbs with a dipping bath and baskets. Hand exposure ranged from 2.5 to 67.8 mg. The number of disinfected baskets ranged from 10 to 18 and the concentration of zineb/maneb in the disinfection liquid from 2.3 to 12.5 mg/l (median 11.4 mg/l). The exposure can be related to the number of barrels which were handled during disinfection and to the total amount of bulbs which was disinfected during the sampled disinfection operations. The transformed data are listed in Table 3. The mean geometric exposure level was 1.46 mg/barrel (GSD = 3.7) and the mean exposure level per kg of bulbs was 43 µg, when transformed to exposure per barrel (see Appendix II). The dermal exposure of three farmers using drive-in vessels was measured as well. The mean zineb/maneb concentration in the bath was relatively low (3.6 mg/l), because of the use of a mixture of captan and zineb/maneb. Hand exposure to zineb/maneb ranged from 1.5 to 4.3 mg and the number of barrels ranged from 5 to 10. Exposure data transformed into exposure per barrel and per kg of bulbs are (geometric means) 0.56 mg/barrel (GSD = 3.3) and 1.1 µg/kg bulbs, respectively. Because of the small sample sizes, the significance of the results is limited but they indicate a much higher exposure for manual dipping than for the drive-in technique. This might be due to the different concentrations of zineb/maneb in the disinfection baths. However, the manipulation of contaminated barrels and bulbs during dipping,

which is more likely to occur in the manual process, seems to be a much more probable cause. The lack of knowledge on this subject emphasizes the need of (dermal) exposure assessment *during* other activities with the contaminated barrels and, e.g., during manual planting after disinfection.

In four cases hands were washed after a series of bulb disinfection cycles performed with protective gloves. Exposure levels ranged from 1-16.4 mg. Both the number of barrels handled (range 6-220) and the way of using the gloves (full period, or limited to the actual dipping) differed considerably. Some degree of protection was observed. In the literature very few data are available on the effectiveness of protective gloves during exposure to pesticides in practice, e.g. during loading of wettable powders and dusts or during dipping. Maitlen et al. (1982) reported a reduction of 97% by wearing gloves during loading of a wettable powder. Chester et al. (1987) and Fenske et al. (1987), however, found significant amounts of active ingredient underneath protective gloves on cotton gloves or on the hands after loading a liquid formulation. The results of the present study fit well with these findings. Although the results indicate a relatively high reduction of exposure (> 94%) in a single mixing and loading operation and a moderate reduction (about 70%) during bulb disinfection activities, the amount of a.i. measured on cotton gloves and on hands is substantial. Conclusions on the efficiency of protection cannot be drawn, because in practice the same gloves will be used for many loadings and bulb disinfection cycles over a long period of time. Contact with the contaminated exterior during pulling them off will certainly occur.

Conclusions and recommendations

From the results of the field studies on the exposure to zineb/maneb the following can be concluded.

- The results of the exposure measurements during mixing and loading indicate a relatively high dermal and respiratory exposure compared to other studies. For the same amount of active ingredient, direct tank filling leads to a relatively low exposure as compared with the two methods of pre-mixing.
- Since there was no significant difference observed between the exposure of persons within one method of mixing and loading, the results indicate a minor influence of individual working hygiene on exposure, whereas an environmental factor such as wind speed has a significant influence on respiratory exposure.
- Respiratory exposure during mixing and loading inside the barn is high compared with mixing outdoors. Therefore it is recommended to avoid mixing indoors.
- The results of the exposure measurements during bulb disinfection indicate a substantial difference of dermal exposure between manual dipping and the use of drive-in vessels.
- Preliminary results on the level of exposure when using protective gloves indicate a relatively large reduction of dermal exposure. However, the remaining exposure is still substantial.

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Table 1 Results of dermal exposure during mixing and loading

Method	1 Direct tank filling [mg] [mg/kg a.i.]		2 Premix- dumping [mg] [mg/kg a.i.]		3 Premix- scooping [mg] [mg/kg a.i.]	
N	18	18	17	17	15	15
GM	107	17*	111	46*	251	140*
GSD	2.1	2.0	3.1	2.3	2.6	2.4
Range	29-300	6-66	14-360	3-179	53-2300	30-1095

* Method 1 < method 2 < method 3 (Kruskal-Wallis; $p < 0.05$)

1 Active ingredient (a.i.)

Table 2 Respiratory exposure during mixing and loading

Method	#1 Direct tank filling [g/m ³]	#2 Premix-pouring [g/m ³]	#3 Premix-scooping [g/m ³]	#4 Premix indoors [g/m ³]
	[g/m ³]	[g/m ³ /kg a.i.]	[g/m ³ /kg a.i.]	[g/m ³ /kg a.i.]
Number	18	11	9	10
Geometric mean	675	294	139	2984
Geom. stand. dev.	4.1	3.4	2.5	3.5
Range	42-14700	8-1960	36-580	507-18200

* (Methods #2 + #3) < Method #4 (Kruskal-Wallis; p < 0.05)

Table 3 Dermal exposure during bulb disinfection

	Drive-in vessel [mg/barrel]	Dipping bath [g/kg bulbs]	[mg/barrel]	[g/kg bulbs]
N	3	3	6	6
GM	0.56	1.1	1.46	43
GSD	3.3	3.6	3.7	3.7
Range	0.15-1.4	0.3-2.9	0.2-4.13	6-118

Appendix I

Calculations of method-specific exposure levels

Method-specific levels of dermal exposure were calculated from a data base (Joint Medical Panel, 1986) and from data obtained by field studies on exposure to zineb/maneb (see section I). Since the unit of exposure of the data base is ml spraying liquid per hour, exposure levels had to be transformed mg active ingredient per ha of farming land. Tables 1-3 show the stepwise transformation of these data specific conditions of spraying zineb/maneb in the bulb growing culture.

Table 1 Application speed¹ [l/ha/h] for field spraying techniques²

	Tractor boom spraying		Backpack spraying	Bike spraying
	600 l	400 l ³		
Arithmetic mean	2.6	1.3	0.26	0.34
Standard deviation	0.6	0.6	0.11	0.09
Number	35	46	14	23

¹ Volume rate 200 l/ha

² Results of log-book data

³ Volume rate 400 l/ha

Data on the application dose of zineb/maneb in crop protection were obtained from the field study on mixing and loading. From these data and the volume rate, the concentration of the active ingredient (zineb/maneb) was calculated (table 2). From the UK data base both the geometric and arithmetic mean of dermal exposure were calculated. These were transformed into method-specific exposure levels in mg/ha according to: method-specific exposure level [mg/ha] = exposure level [ml/h] × concentration of zineb/maneb in the spraying liquid [mg/ml] × spraying time per hectare of farming land (1/application speed) [h/ha]. Furthermore, these values were adjusted to exposure of the hands for tractor boom spraying and back-pack spraying, which amount to 65% and 25% of the total exposure, respectively. The results are presented in table 3.

Table 2 Mean applied dose of zineb/maneb¹ [kg/ha a.i.] and concentration in spraying liquid [g/l a.i.]

	Tractor boom spraying		Backpack spraying	Bike spraying
Tank size	600 l	400 l	15 l	20 l
Applied dose	2.1	2.1		
Stand. dev.	0.5	0.9		
Number	7	9		
Concentration	10.5	5.25	10.5	10.5

¹ Data from mixing and loading study.

In a pilot study on exposure during bike spraying, the exposure of only one person was measured twice. The samples were taken with the same method used for the assessment of dermal exposure during mixing and loading (section II, 2.2). The exposure levels were 15 and 130 mg for 0.1 ha, i.e. on the average 700 mg/ha.

Table 3 Calculated method-specific exposure levels

	Tractor boom spraying		Backpack spraying		Hands	[mg/ha]
	Whole body [ml/h]	Hands [mg/ha]	Whole body [mg/ha]	[ml/h]		
Geometric mean	3.2 ¹	13	9	32 ²	1295	320
Geom. stand. dev.	1.7	5.8				
Arithm. mean	10.5	43	28	84	3390	850
Number ³	79	13				

¹ Tractor-mounted hydraulic boom + nozzle spraying (outdoors), without cabin

² Hand-held hydraulic nozzle spraying (outdoors)

³ number of data sets

Method-specific exposure levels for mixing and loading were obtained from the results of the field studies on exposure to zineb/maneb (section II, table 1), and adjusted for an application dose of 2.1 kg/ha a.i. The results are given in table 4.

From the results of the field study on exposure to zineb and maneb during bulb disinfection (section II, table 3) exposure data were obtained, which were used as method-specific exposure levels. The results are presented in table 5.

Table 4 Method-specific exposure levels for mixing/loading methods

Method	Direct tank filling		Premix-pouring		Premix-scooping	
	[mg/kg]	[mg/ha]	[mg/kg]	[mg/ha]	[mg/kg]	[mg/ha]
Geometric mean	17	36	46	97	140	294
Geom. stand. dev.	2.0	2.3	2.4			
Arithm. mean	21	45	60	126	230	482
Number	18	17	15			

From the data in the UK data base on exposure during loading of a liquid formulation using a 10 l container, a geometric mean of 0.06 ml liquid per operation was estimated. At a volume rate of 400 l/ha, an applied dose of 2.1 kg a.i./ha and a 400 l sized spraying tank, one loading operation is required per ha. Since the liquid formulation of zineb/maneb (Tridex™) contains 455 g a.i./l, one operation would result in an exposure of 0.06 ml/ha × 455 mg/ml = 27.3 mg/ha. Spraying at a volume rate of 200 l/ha, using a 600 l spraying tank and an applied dose of 2.1 kg a.i./ha would result in two loading operations for each tank filling. Exposure per ha due to loading would be in this case:

$$\frac{2 \text{ (operations)} \times 0.06 \text{ (ml)} \times 4.55 \text{ (mg a.i./ml)}}{600 \text{ (l)} / 200 \text{ (l/ha)}} = 18 \text{ mg/ha}$$

Table 6 contains data on the density of planting of different bulb varieties. With these data bulb exposure levels specific for bulb variety from table 5, expressed in µg per kg of bulbs, can be calculated for two methods.

Table 5 Method-specific exposure levels of bulb disinfection techniques

	Manual dipping		Others	
	µg/kg bulb	mg/barrel ¹	µg/kg bulb	mg/barrel ²
Geometric mean	43	1.5	1.1	0.6
Geom. stand. dev.	3.7	3.7	3.6	3.3
Arithm. mean	70	2.4	1.7	0.8
Number	6	7		

¹ basket containing about 35 kg of bulbs

² cubic cases containing about 550 kg of bulbs

Table 6 Method-specific exposure levels adjusted to planting densities

Bulb variety [kg/ha]	Density [kg/ha]	Method-specific exposure level Manual dipping [mg/ha]	Others [mg/ha]
crocus	8,000	345	9
tulip	10,000	430	11
daffodil	21,000	900	23
hyacinth	16,000	690	175

Appendix II

Bulb disinfection and exposure

Bulb disinfection is performed to protect bulbs against diseases during the process of bulb growing or flower growing. The method of bulb disinfection is partly related to bulb variety and pesticide applied. For one method the kind of barrel may vary.

Table 1 Bulb disinfection techniques

Bulb variety	Method	Barrel
daffodil	hot water dipping — drive-in cubic case	jute bag
crocus	dipping — manual — drive-in gauze case	basket cubic case
tulip	dipping — manual — drive-in gauze case showering plastic case spraying	basket cubic case gauze case none
hyacinth	dipping — manual — drive-in gauze case showering plastic case spraying	baskets cubic case gauze case none

Table 1 lists the various methods of bulb disinfection for different kinds of bulbs. The different types of barrels have different contents, e.g. baskets contain about 35 kg of bulbs, whereas cubic cases as much as 550 kg. For manual dipping a 200 l dipping bath is used, whereas the content of most drive-in vessels is 2200 l.

Exposure is likely to occur during:

- *preparation of the disinfection liquid*

The pesticides applied in bulb disinfection are largely used in a liquid formulation. Opening of the can and splashes may lead to exposure. Since the amount of bulbs planted per ha absorb 800 to 1200 l disinfection liquid, both the dipping bath and the drive-in vessel need refilling; for the dipping bath the frequency of refilling is about 4/ha and for the drive-in vessel 0.5/ha.

- *maintenance and cleaning*

Most equipment does not need frequent maintenance and within a period of bulb disinfection no cleaning is performed. So exposure will be limited.

- *handling bulbs, barrels and operation of equipment*

Manual dipping involves manual immersion of the basket in and removal from the dipping bath. To prevent the bulbs from floating, generally a basket is covered with a jute bag with a stone on top. After removal, the contaminated baskets are transported to the bulb fields for planting.

In most cases disinfection by drive-in vessels involves the use of a lift-truck to lift a cubic case or a palette with gauze cases or jute bags. Special equipment has been constructed to prevent floating. After disinfection, both barrels and bulbs have to be manipulated to plant the bulbs. Generally, the contaminated barrels are re-used.

Showering, another method of disinfection is very well comparable with dipping using drive-in vessel and gauze barrels. Disinfection by spraying does not demand the use of barrels. Mostly, spraying of bulbs occurs just before covering the planted bulbs with soil. Very coarse droplets are used in this kind of spraying and exposure will probably be limited, although no exposure data are available.

Another type of spraying is a combination of spraying and planting, but this technique is not popular any longer. Both respiratory and dermal exposure considered to be relatively high in this case.

Table 2 lists a ranking of the different combinations of disinfection technique and type of barrel with regard to dermal exposure. The overall ranking was obtained by merging the independent rankings performed by three occupational hygienists. This ranking was considered to be reasonably good according to two "experts" on bulb disinfection techniques. It was concluded that manual dipping could be considered to give the highest exposure and that exposure during application of the drive-in vessel technique was representative for exposure by all other techniques.

Table 2 Ranking of bulb disinfection techniques in decreasing order of exposure

Technique	Barrel
manual dipping bath (200 l)	basket
spraying in combination with planting	none
shower	gauze case
drive-in vessel	case
drive-in vessel	jute bag
shower	plastic case
back-pack spraying	none
drive-in vessel	cubic case