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# Assessment of occupational exposure to pesticides in agriculture

Part I General aspects

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Labour Inspectorate

# Assessment of occupational exposure to pesticides in agriculture

Part I General aspects

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## SUMMARY

For registration of pesticides data on toxicology and on occupational exposure are required. In this series of reviews the exposure data available in the published literature for mixing and loading, application and re-entry are considered for the establishment of generic/surrogate data bases with which for a specific case (e.g. a new pesticide) the exposure can be estimated for use in a first step for risk assessment.

In the first part of the series the general aspects of the determination of exposure to pesticides under field conditions are considered, as well as the published proposals for generic data bases. It is concluded that the development of data bases is in principle possible, although in many cases (depending on the type of technique for mixing/loading and application) not enough data are available. Data bases on re-entry have not been proposed in the literature, although many studies on exposure after re-entry have been reported.

Various relevant variables that may affect the dermal and inhalation exposure to pesticides in various agricultural settings are mentioned in the literature. The most important groups of variables are: (1) type of formulation and packaging, (2) application technique and working method (personal hygiene), (3) agricultural and climatic conditions. It is noted, however, that many published reports are insufficient in reporting data on the relevant variables. Therefore, it is concluded that every single report on exposure has to be evaluated before inclusion in a data base can be considered. Furthermore, it is concluded that the exposure data should be available in amounts of volume (for liquid formulations and sprays) or in amounts of weight (for solid formulations and dusts) and preferably given as a ratio to the amount of pesticide handled.

A general approach to risk assessment using (surrogate) exposure data is described.

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## 1 INTRODUCTION

Use of pesticides<sup>1</sup> is widespread in agriculture and elsewhere. The hazards of these compounds depend on their toxicity and on the level of exposure. This is not only true for workers but also for the general public, although the route(s) of uptake may be quite different.

Toxicity is an intrinsic property of a particular compound. It has to be assessed for each pesticide as completely as possible. In fact, (inter)national regulation prohibits the use of pesticides unless sufficient knowledge on their toxicity has been obtained, although the quality of the evaluation and interpretation of toxic properties may vary among countries and, certainly, changes throughout time.

The other component needed for evaluating the risks due to contact with pesticides is the level of exposure. Exposure is largely dependent on the job being done, how it is done, the physical form of the pesticide and the ambient weather conditions, rather than on the chemical nature of the product.

Pesticides are biologically active compounds. Most of them are considered toxicologically relevant. It is not surprising, therefore, that governments and workers question the risks due to working with diluted as well as with highly concentrated solutions (or suspensions) or dusts of these compounds. In some cases contact with pesticides occurs frequently or even daily. The activities include not only application of pesticides but also picking of fruits and cutting of vegetables and flowers from treated crops.

In this series of papers the assessment of occupational exposure will be described as well as the possibilities to use published data on exposure for the estimation of exposure in situations which have not been investigated experimentally. This process of risk assessment on so-called generic data (also called surrogate data) is very important for the registration of new pesticides and for re-evaluation of pesticides which have already been used for several years or decades.

In part I of this series the general aspects of the assessment will be described. In following parts the open literature will be reviewed for relevant data on exposure assessment during mixing and loading, application and re-entry.

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<sup>1</sup> Pesticides : all agents (substances, mixtures of substances or microorganisms) causing a herbicide, insecticide, fungicide, or rodenticide action. Furthermore, the legislation in The Netherlands covers against ectoparasites on cattle, growth regulators for plants and agents to control pests in buildings used to house animals, in means of transport for animals or in milking equipment [1]. For this paper only chemical agents are considered.

## 2 GENERAL CONSIDERATIONS

The safety of workers is a key consideration in the registration of pesticides for agricultural practice. The safety measures that have to be taken to match the levels of (occupational) exposure and the toxicologically allowed levels of uptake, must be based on a critical evaluation of the various factors that may affect the level of exposure.

Working with chemicals can be done under conditions which do not lead to exposure. The risk will then be (close to) zero. However, in practice, especially in agricultural practice, this is not very likely to occur. Usually, the farmer is his own 'specialist' for matters of occupational hygiene and safety.

In the present paper it is assumed that the toxicity of the individual pesticides is known and therefore needs no further consideration, i.e. sufficient knowledge exists on 'no-effect' levels. In many cases this will only be true for oral exposure as determined in long-term studies with animal species. Since occupational exposure occurs through inhalation, along the dermal route and - to a generally smaller extent - via the oral route, knowledge is needed of the degree of absorption through the skin and via inhalation. Especially for skin exposure this degree varies considerably amongst pesticides.

The oral exposure in agricultural practice is generally not considered, since it is hard to determine. However, in the case of a high dermal exposure, or inhalation exposure to relatively large particles, one should consider the possibility of secondary ingestion.

The processes which may lead to the occurrence of health effects can be described in the following terms : exposure, absorption (or penetration), distribution; elimination (deposition<sup>2</sup>, metabolism and excretion) and toxic effects. This paper will only deal with the first element of this chain of processes, although it should be stressed that for the occurrence of systemic effects only that part of the exposure needs to be considered which is relevant for absorption (or penetration). Therefore, a further distinction is introduced: potential and actual dermal exposure. Potential dermal exposure is the total dermal exposure of an individual, including the exposure of the clothing. The actual dermal exposure is the amount on the skin, either directly or after penetration of clothing.

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<sup>2</sup> Deposition : formation of a depot (e.g. in fat or bone).

### 3 APPROACHES TO THE MEASUREMENT OF EXPOSURE

Exposure can be estimated by measuring contamination of the skin, or of substitutes like pads or special suits, and by monitoring the ambient air. Furthermore, exposure can be estimated by measuring the compound or its metabolites in body fluids or excreta (biological monitoring), or can be inferred from measurements of some physiological or biochemical change in the body (biological-effect monitoring or health surveillance). The former procedures measure external exposure, the latter internal exposure or (early) health effects. For risk assessment, it is certainly of more value to know the internal exposure and early health effects rather than the external exposure. However, external exposure may be interpreted in terms of internal exposure if sufficient data on absorption or penetration are available.

For registration of new pesticides, knowledge on health effects in man is completely lacking. In fact, the aim of the registration procedure is prevention of health effects. Biological monitoring is a very efficient way to estimate the relevant levels of internal exposure. It is, however, compound-specific and resultant exposure data can, therefore, not easily be used for exposure assessment of other compounds. The same argument is valid for biological-effect monitoring in the sense that the relation between exposure and biological effect is compound-specific. As an example, cholinesterase inhibition by various organophosphorous compounds and carbamates may be mentioned. In a recent technical monograph GIFAP [2] has presented a similar approach for exposure monitoring as is given in this paper.

In the following sections only the assessment of external exposure will be discussed, although some remarks will be made on biological monitoring. The main reason for this is the possible use of data on external exposure for extrapolation to other compounds, i.e. the tabulation of generic data.

#### 3.1 Measurement of external exposure

Exposure may occur at almost all parts of the (clothed) body. A differentiation has to be made between exposure of the skin (directly or through the clothing), which is called actual exposure, and exposure of the whole body (covered and not covered by clothing), the potential exposure. This differentiation is very important, since only the amount that reaches the skin eventually is hazardous. In the literature the actual exposure is usually taken as the exposure of hands, forearms, head, neck and the "V" of the chest.

Techniques for determining the deposition of pesticides on clothing and skin include the use of pads (or patches) and suits (coveralls), washings or wipings of parts of the skin, as well as tracer techniques.

Inhalation exposure is determined by measuring the concentration of the



compound in the breathing zone. The air collection devices used are designed in such a way that only particle sizes are collected that may enter the airways. For solid and liquid aerosols the collection system is based on filtration and for vapours on adsorption (e.g. charcoal). In the early days of exposure assessment respirators were used to trap the particles and vapour present in the inhaled air. Presently, the availability of 'personal air samplers' largely precludes the use of respirators and stationary air samplers for the assessment of individual exposure.

### 3.1.1 Dermal exposure

#### *Washing methods*

Chemicals can be dissolved in solvents and thus it is possible, in principle, to estimate the amount of deposited pesticides on the skin by washing, wiping or swabbing the skin. Mild solutions should be used for human skin, such as dilute solutions of detergents. However, many pesticides do not dissolve in these solvents. So the use of alcohols and ketones, such as methanol, ethanol and acetone, have also been described in the literature.

With these methods one can only obtain the amount of pesticide that has not already been absorbed by the skin and even the unabsorbed pesticide may not be removed completely. There have been no reports in the literature on the effectiveness of the washings for specific compounds. In practice, complete removal of the amount present on the skin at the time of (thorough) washing is assumed. For compounds that readily penetrate the skin the amount of exposure will thus be underestimated, unless sampling is done after short periods of exposure. Frequent sampling will also lead to inadequate data, since the skin will become more penetrable. In one or two cases this method for the determination of dermal exposure has been compared to another technique, the deposition on cotton gloves [3,4]. For the compounds investigated, the data indicate that washing may show lower levels of exposure than the use of gloves.

It should be noted that the quantitative estimation of skin exposure by washing is only practical for the extremities of the body such as hands, arms, feet and legs, although the use of a bath for complete immersion of the body is not precluded for compounds that dissolve readily in water.

#### *'Pseudo-skin' methods*

The use of pseudo-skin for the quantification of dermal exposure presumes that the matrix will contain as much pesticide as would have been retained by the skin itself. The methods in use are pads or patches, coveralls, caps and gloves made of various materials. The materials that are frequently used are cotton, denim and (for pads) paper. Especially the use of gloves is important, since the level of external exposure of the hands is generally high in comparison to other parts of the body. No reports have been published in which it has been attempted to estimate the correlation between the actual

exposure of the uncovered skin of the hands and the exposure of gloves made of various materials. One can imagine that the level of exposure as observed with gloves will be an overestimation since all residues which might have contacted the skin are collected. Special attention should be given to the situation where liquids are involved. It seems quite possible, depending on the type of material used, that a glove may act as a sponge and collect more liquid than would have adhered to the uncovered skin. For these factors some allowance should be made, although for the moment this cannot be done quantitatively. Further, it should be noted that during work part of the pesticide on the skin may be removed again. This amount is not considered when exposure is measured over short periods of time using gloves, etc.

Workers wear clothing, even in the hottest conditions. This may protect them from contamination through splashes, mists and dusts. Especially splashes may, nevertheless, lead to exposure of the skin underneath. These splashes may be indicated experimentally by the introduction of coloured (*e.g.* fluorescent) compounds in the spray liquid.

The amount of pesticide on the clothing can be determined directly by analysing the clothing. The amount that permeates the clothing may be estimated from the amount on t-shirts and underwear worn underneath. It is emphasized again that it is assumed that the level of exposure measured is identical to the exposure of the skin in the absence of this underclothing.

For the estimation of the potential exposure of the body pads can be used or coveralls, which cover the whole body and which may be analysed in parts to estimate the exposure of different parts of the body. The pads are made of absorbing material and are placed at various positions on the clothing to estimate potential exposure, or underneath the clothing to estimate actual exposure. The use of pads may lead to erroneous estimates, since splashes are by nature local and may or may not be 'observed' by the pads. Estimation of exposure underneath the clothing can also be done by pads that are made up of two layers. The amount of the pesticide on the lower layer is used as a measure for the actual exposure.

The use of pads has been described in detail by Durham and Wolfe [5]. For the location of the pads on the body, different guidelines and reports recommend different numbers and positions [6,7]. The estimates of contamination of the pads have to be extrapolated to the whole body. This may be done by using standard surface areas of the various parts of the body, such as those proposed by WHO [6] and EPA [7]. In some papers the exposure of the hands is measured with wrist pads. This will very likely give erroneous results since relatively large amounts of pesticides will adhere to the fingers and palm of the hand during the work and will not be 'observed' by wrist pads.

### *Tracer techniques*

The use of tracers is widespread in experimental chemistry. A tracer is used which is chemically (almost) identical, but far more easy to detect or to

analyse. As was noted before, fluorescent tracers to indicate the distribution of splashes of liquid on the clothing have been used in several exposure studies. A relatively new development is the use of a quantitative method based on fluorescence. With this technique the fluorescent tracer is quantified at the various parts of the skin (covered and uncovered). The technique is laborious, but gives accurate data on actual dermal exposure and thus allows the study of the effectiveness of protective clothing. It should be noted, however, that the interpretation of the measurements of the fluorescent tracer requires knowledge on the (differences in) properties of the pesticide under study and the tracer itself, such as degree of permeation through clothing. In this respect the large body of work of Fenske and co-workers should be mentioned [8,9, see also 10].

### 3.1.2 Inhalation exposure

The concentration of pesticides in the breathing zone of the worker can be estimated by means of 'personal air samplers' (PAS) which have been designed in such a way that inspirable particles are retained on filters and gaseous pesticide material on adsorbents and/or impingers. It should be noted, however, that not all PAS-measurements described in the literature have been obtained with equipment validated for inspirable liquid and solid particles. This may lead to an unknown degree of overestimation of the inhalable exposure

The amount of pesticide on the filter, on the adsorbents and/or in the impinger liquid must be determined. This amount and the sampling time yield the average concentration in the breathing zone.

In the early days of exposure assessment, air samples were collected in the area where the application took place. This so-called stationary sampling has since been shown to be generally less adequate for the estimation of worker's exposure.

The work of Durham and Wolfe [5] has stimulated the use of respirators. This method involves the use of filter pads to determine the amount of pesticide that is 'drawn in' by the workers. Although this method is thought to measure the exact amount of pesticide that is inhaled by the worker, this may not be true for several reasons: (1) the pads are easily contaminated in field experiments, (2) leakage along the mask, (3) the breathing rate may change and (4) particles that would not enter the airways because of their size, may also be trapped on the filter. The latter is relevant for aerosols which consist (partly) of particles of a high mass and will therefore give an overestimation of the personal exposure.

### 3.2 Measurement of internal exposure

Measurement of the amount of pesticide absorbed by the body is done by the assessment of the parent compound or one of its metabolites in urine, blood,

faeces, exhaled air or sweat. In some cases (metal-containing compounds) even measurements in hair may be used. A great advantage of the estimation of internal dose is the integration of exposure along all routes: inhalatory, dermal and oral. As has been noted above, oral exposure will not be taken into account when the external exposure is estimated.

The so-called biological monitoring is based on detailed toxicokinetic knowledge of the compound and should reflect the external exposure or, preferably, the amount that entered the body (absorbed dose). Biological monitoring based on dose-excretion studies will generally be developed in the case of pesticides which are widely used and for which the levels of exposure in agriculture are such that more precise knowledge on internal exposure is required. Recent developments and applications of biological monitoring have been described in many papers and several proceedings of symposia [e.g. 11,12]. In this paper the use of biological monitoring will not be given further attention, since biological monitoring data cannot easily be used as surrogate data for exposure to other compounds. The importance of biological monitoring for the assessment of internal exposure (absorbed dose) is evident.

#### 4 USE OF SURROGATE/GENERIC DATA FOR EXPOSURE ASSESSMENT

There are about 500 active ingredients on the international market of pesticides in about 10,000 different formulations. This indicates that with regard to assessment of exposure to pesticides a search for generalisations, theoretically or pragmatically, is quite useful. The quality of registration procedures would increase enormously if this search would be successful and the high economic costs involved in assessing different field conditions could then be used for the most important agricultural settings and for the most toxic compounds and most hazardous situations. This line of reasoning has been used in several countries, at the level of regulation as well as in research institutes.

Fortunately, exposure to pesticides depends on various variables that are not related to the chemical nature of the compounds. For a systematic approach of the problem, a definition of the variables that may affect the level of exposure is necessary, as well as verification in field experiments. Fundamental research has been done by Batel in Germany. It is a pity that almost none of this work has been published in English [13]. This has hampered the dissemination of his findings.

Popendorf *et al.* [14,15] developed a so-called 'unified field model for re-entry hazards' for the exposure during re-entry, i.e. the transfer of pesticides from the crop to the skin due to contact with the crop during picking of fruits, cutting of flowers and similar activities, as well as contact with whirling dust (*e.g.* from the soil). So far, however, generic data have not been considered in the literature for extrapolation to various other situations. The only attempts for extrapolation are related to the transfer from the so-called dislodgeable residue from crop to skin [16]. These considerations, as well as a survey of the relevant literature, will be described in that part of this series that will deal with exposure after re-entry, and will not be described here.

For further considerations, one should dissect the actual processes in which exposure to pesticides occurs. Generally, this is done by defining three categories: (1) mixing and loading, (2) the actual application and (3) re-entry. In the literature on exposure to pesticides the categories (1) and (2) are not always differentiated, especially for work in enclosed spaces, *e.g.* greenhouses, with relatively small amounts of active ingredient.

It is evident that also a differentiation should be made between work in enclosed areas (inside) and work in the open field (outside). This is important for inhalation exposure (to volatile compounds, mists and dusts) as well as dermal exposure to mists and dusts. Further differentiation should be based on upward (*e.g.* in orchards) and downward (*e.g.* for weed-killing) application. Climatic conditions as well as agricultural conditions may largely affect the

levels of exposure. The most important variables are temperature, wind conditions and factors such as field or plot size, level of mechanization and aspects of hygiene. Therefore, one should be careful in using data obtained in one part of the world for extrapolation to other parts with totally different conditions. In the following section (4.1) several of these aspects will be treated in more detail. In section 4.2 some analytical and agricultural aspects of exposure will be described. In section 4.3 published papers on the use of surrogate data will be reviewed.

#### 4.1 Factors that affect exposure under field conditions

Batel [13] has studied the exposure due to application of pesticides under laboratory conditions, using a suitable model compound. The levels of inhalation exposure were examined with a glass model of a head containing an air sampler and contamination of the skin with a glass model of a hand. From this work, which will not be described in detail, it is concluded that exposure at a certain site (in *e.g.*  $\text{mg}/\text{m}^3$ ) is largely determined by three groups of variables: (1) emission of the compound(s), (2) conditions in the field and (3) equipment used and type of crop. The various variables are described in the following terms :

- (1) the emission rate, which is a function of the applied dose (application dose,  $\text{kg}/\text{ha}$ ), the forward velocity of the equipment relative to the crop and the width of the spraying zone;
- (2) the relevant air conditions, such as relative direction of the wind (determined by angle), relative velocity of the wind and relative humidity;
- (3) the liquid pressure at the nozzle, the velocity of the supporting air stream (if present), the height of the spraying nozzles relative to the ground, the distance between nozzles and worker (measuring site) and droplet size. Further, secondary aspects such as height and density of the crop may affect exposure.

Details on the effects of these variables on the immission at a work site have been gathered in model experiments. This has led to general formulae and from the emerging exposure immitted at a work site, the actual inhalation and dermal exposure can be calculated, at least in principle. In principle, because the calculations are for the moment related to the glass models described above. This procedure has been described in detail [13] and, with some related aspects, in a publication of the Landbauforschung Völkenrode [17]. Some general conclusions from this work are that the amount of exposure is linearly related to the application dose. Exposure increases with smaller droplet size. The effect of wind velocity on the level of exposure is complex. Wind direction affects exposure to a large extent, as would be expected. Exposure increases with decreasing horizontal distance between nozzles and driver of a tractor-driven equipment. The use of a cabin on a tractor decreases exposure, although cabins used properly are not observed frequently in actual practice.

The equipment that is used for different types of application, as well as the methods themselves, vary widely. For this reason it is important to categorize the various types of application in groups. For the agricultural practice in The Netherlands this has been done by a group of agricultural experts (data not published). It may not be possible to find exposure data in the literature for all these groups. Nevertheless, the distinctions are made. The main reason is, that relatively large differences in techniques may be involved. For registration procedures, however, it might be possible to rank the level of exposure during handling of the pesticides between the groups. The different groups that have been distinguished are given in table 1.

Table 1 Differentiation between methods of application

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1	Downward spraying (outdoors)
	1a Tractor mounted equipment
	1a.1 Treatment of soil (spraying boom low position)
	1a.2 Treatment of crop (spraying boom high position)
	1b Aerial equipment
2	Soil fumigation (tractor mounted equipment)
3	Application of granules (outdoors)
	3a Tractor mounted equipment
	3b Manual
4	Upward spraying outdoors (tractor mounted equipment)
5	Spraying outdoors (manual; upward and downward)
	5a Stationary equipment
	5b Knapsack
6	Spraying indoors (upward and downward)
7	Ultra-low volume spraying (indoors)
8	Soil fumigation under plastic cover (indoors)
9	Dusting (indoors)
10	Application of granules (indoors)
11	Fumigation of enclosed locations
12	Disinfection of seeds and bulbs
	12a Dipping (mechanical)
	12b Encapsulating/coating
	12c Dipping (manual)
13	Spraying of animals
14	Dipping of animals

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The measure of exposure is generally expressed in units of weight (or concentration) per unit of time. This seems appropriate, since the total mass

taken up by the body over a relevant time period will determine the toxicological risk. If extrapolation to other situations is requested, it is much better to use other measures, however, such as the amount of compound related to the actually handled amount (*e.g.* mg/kg). Another approach is the use of a measure of the amount of formulation (in units of volume or weight, depending on the nature of the formulation) or spray liquid (in units of volume). The rationale behind this is the very practical consideration that workers deal with the formulation and spray liquid and the amount of external exposure will in most cases not depend on the chemical nature of the active ingredient, but largely on the physical nature of the dust or granule of the formulation or on the droplet size of the spray liquid. Therefore, exposure has to be expressed in relation to the solid or liquid that is dealt with. This implies the use of volume of spray or weight of formulation. It should be noted that the exposure during mixing and loading is generally expressed in terms of the formulation assuming no contact with the spray liquid. The exposure during application is counterwise expressed in terms of the spray liquid, assuming no contact with the formulation. Knowledge on the percentage of the pesticide present in the formulation and of the concentration in the spray liquid can be used in registration procedures to determine the exposure of the active ingredient involved. Especially with regard to inhalation exposure, volatile compounds may behave differently. Such compounds, especially gasses, should be treated separately.

The work of Batel only deals with application of pesticides [13]. The exposure during mixing and loading cannot be easily captured in theoretical terms, since part of the exposure is due to contact with the undiluted and diluted formulation of the pesticide, as well as with contaminated surfaces. No theoretical analysis of the physical processes of transfer of material through contact has been published to our knowledge and, therefore, at present only a pragmatical approach can be taken.

### *Mixing and loading*

Mixing and loading is normally done manually (although closed transfer systems have been developed and put into use) and takes a relatively small amount of time compared to the actual application. In Dutch agricultural practice mixing and loading is generally done by the applicator himself.

The most important formulations can be subdivided into concentrates for (1) dilution with water, (2) products to be applied undiluted and (3) products for seed treatment [18].

(1) The products to be diluted with water are either emulsifiable concentrates, emulsions in oil, suspension concentrates, soluble concentrates, water soluble powders, water soluble grains and suspo-emulsions. Besides, there are a few other types of formulation of minor importance.

(2) The ready-for-use products are dustable powders, granules, electro-chargeable liquids, spreading oils, tracking powders, ultra-low-volume liquids



or suspensions. Liquids for soil injection are the most important formulations in The Netherlands (about 50 % of total volume). Some other products are of minor importance.

(3) The products for seed treatment comprise powders for dry use, suspension concentrates and emulsions, as well as water soluble and water dispersible powders and solutions.

These different formulations can be summarized for the present purpose by the categories liquids, powders and granulates.

Studies of the distribution on the body of the contamination of workers during mixing and loading show, in general, that the larger part of the exposure concerns the hands. However, the possibility of exposure elsewhere on the body should always be considered.

It is expected that the amount of exposure will depend on conditions of wind and ventilation, especially when powders, dusts or small droplets are used. Therefore a distinction between working indoors and outdoors is worthwhile. There have been some developments for the introduction of closed transfer systems. The use of these systems, which are meant to decrease the level of worker exposure, is at present only marginal in The Netherlands.

#### *Application*

Application is still mainly done with relatively large amounts of water or with dusts. For a number of applications the amount of water is small, or even nil. In table 2 a classification is given, as presented in [19].

Table 2 Classification of sprays according to volume

Volume classification		Application dose (l/ha)	
		Field crops	Trees and bushes
High volume	HV	> 600	> 1000
Medium volume	MV	200-599	500-999
Low volume	LV	50-199	200-499
Very low volume	VLV	5-49	50-199
Ultra-low volume	ULV	< 5	< 50

Evidently, the droplet size depends on the nature of the spray. With smaller volumes, the droplet size has to be decreased to obtain a homogenous 'covering' of the crops or other surfaces. Since with decreasing droplet size

the drift of the spray (mist) will increase, application of ultra-low volume applications will occur mainly in enclosed spaces such as greenhouses. As is noted above, a further differentiation has to be made between applications outdoors and indoors, as well as between upward and downward spraying. Since weather conditions vary throughout the year, and even throughout a day, there is not much point in further differentiation of exposure in relation to weather conditions such as wind speed, temperature and relative humidity, for registration procedures. The climatic conditions may, however, differ even more between different parts of the world, such as between Western Europe (*e.g.* The Netherlands, England and Germany) and large parts of Africa, Asia and Latin America. These differences will be effective at the level of exposure, especially for application of volatile pesticides outdoors. This is an important factor to consider when extrapolating data from different countries. Extreme conditions should be excluded from a data base.

Exposure may also occur during cleaning of the spray tanks and during trouble-shooting (*e.g.* cleaning of nozzles). These activities are generally included in field observations but may not always be included in exposure studies done solely for registration procedures. On the other hand, cleaning of a tank will usually lead only to exposure to very diluted liquids and, therefore, probably contribute only to a minor degree.

Exposure of the different parts of the body depends on the type of application, the climatic conditions during application and the work methods. The direction of the wind, for instance, is an important variable. In [19] several studies have been considered in this respect and some data are given on the distribution of dermal contamination. These data are summarized as a rough guide in table 3.

Table 3 Distribution of dermal contamination<sup>1</sup> during application

Application technique (outdoors)	Body part		
	hands	upper body	legs
Downward spraying (tractor)	65-75 %	10-15 %	10-25 %
Spraying (manual)	10-25 %	5-65 %	25-85 %

<sup>1</sup> Data based on potential exposure [19].

## 4.2 Analytical and agricultural aspects of exposure data

As is shown in chapter 3, dermal and inhalation exposure data are generally obtained with techniques that have not been validated, in the sense that it has not been shown that the data are a good measure for the true amounts that one seeks to know. Furthermore, the majority of the published exposure data are given without sufficient information on the analytical chemistry involved. Reproducibility and accuracy of the procedures, recoveries from absorbents, the placement of the absorbents on the body and the precise method of calculation of exposure from the distribution of absorbents are hardly discussed. In a few cases relevant measurements (for dermal exposure) of the hands are lacking.

Some pesticides are unstable under various conditions. This means that the treatment of the field samples is very important. In many papers no data are given to indicate the possible effects of decay on the exposure data presented. It has been shown, for instance, that pesticides in absorbents, such as cotton pads may disappear through volatilization or through hydrolysis. A possible solution of this problem for measurement of dermal exposure to volatile chemicals is given in [20]. Hydrolysis when the pesticide stays on leaves of the crop may be very important for exposure after re-entry. Because of the possibility of formation of other -maybe even more toxic- products, this aspect merits much more attention in exposure studies than is generally given.

In several papers the agricultural data are not described extensively, neither are the climatic conditions. Data should be given on field or plot size and on application equipment, such as tank volume, tank geometry (location of filling site), type of nozzles, liquid pressure and tank-cleaning procedures. Furthermore, details on the amount of pesticide handled, as well as on the nature of the formulation are not always given. This means that the necessary transformation for comparison of the data is not always possible.

The work methods and personal hygiene of a mixer/loader, applicator or picker (re-entry) may affect the exposure to a large extent. This aspect is not considered in the work of Batel, as noted in section 4.1. It has in fact not been described in any detail in published exposure studies. Calamities, such as large splashes of liquid reaching the body are mentioned in several study reports, however. Extreme levels of exposure due to sudden changes in wind speed and other environmental changes are indicated. Such exposure data are generally omitted from final analysis of the data.

## 4.3 Review of the development of generic/surrogate data bases

The way data are presented varies largely among different publications. Exposure data are given as a range, as average values (arithmetical or geometrical) and variation in the data is sometimes given as standard deviation. In many publications data are defined as non-detectable, without indicating the detection limit. In some cases the methods of calculation for

obtaining the final exposure data are not given. In addition, it is noted that in many instances exposure data are collected underneath the protective garment. These data may not be relevant for generic data bases for (potential) external exposure as they are proposed here.

In an unpublished paper entitled "Estimation of exposure and absorption of pesticides by spray operators" of the Joint Medical Panel of the Scientific Subcommittee on Pesticides from the U.K. Ministry of Agriculture, Fisheries and Food and the Toxicology Committee of the British Agrochemical Association, the exposure during mixing and loading as well as during actual application was considered on the basis of an unpublished series of experiments [19]. In this paper a well-developed scheme is presented along which the generic data that can be used to predict the actual exposure for another pesticide.

A similar approach has been made in Germany [21]. The original study reports are, however, not available.

In the literature only a few attempts have been made to describe a data base from which extrapolations can be made. The most explicit one has been presented in a book entitled: "Occupational Hazards of Pesticide Use" [22]. Crome evaluated a large part of the literature on worker exposure and Turnbull *et al.* summarized these exposure data. Hackathorn and Eberhart [23] made a proposal for a data base for future application in registration procedures. For the time being (1984) they concluded that the available data in the open literature are insufficient. Pependorf and Franklin [24] made a crude estimate of the ranges for dermal contact exposure, which they consider relevant for risk assessment. No underlying data base is presented.

Reinert and Severn [25] describe the viewpoint of the Environmental Protection Agency of the USA. They consider it quite possible, with the increasing number of exposure studies with time, to use surrogate data for risk assessment. It is stated that the Agency will continue to pursue the development of a generic data base [7,25]. This data base is to include for use by others the exposure data obtained in confidential studies presented for registration procedures. So far, however, such a data base has not been made available.

Honeycutt [26] does not give generic data, but presents a scheme with some examples of how to use generic data for predicting mixer/loader and applicator exposure. Franklin [27] has described the registration procedure used in Canada. It is emphasized that much additional knowledge is needed on exposure and on percutaneous penetration. Maddy *et al.* [28] describe the registration procedures in California (Department of Food and Agriculture) as well as several studies that have been carried out in California to estimate worker exposure. The values they present are considered (by the authors themselves) low in comparison to other data, mainly due to the fact that the workers in the Californian exposure studies were well-trained and worked strictly according to regulations.

Batel and Hinz [13] have presented some data which they consider to be the best available estimates of worker exposure during application. The data are taken from published and unpublished studies that were done in accordance to procedures considered acceptable in view of the work of Batel [29]. The underlying data base is not given.

### *Mixing and loading*

Exposure of the hands during mixing and loading is considered very important. In general this comprises much more than 50% of the total body exposure [27]. Some studies even consider dermal exposure to be virtually confined to the hands [19]. The proper use of protective gloves may decrease exposure of the hands. They are considered to prevent actual exposure to the skin completely [21]. Maddy *et al.* [28] state that even with gloves exposure of the skin of the hands represents about 40% of the total body exposure. Three possible explanations are given: (1) contamination of the inside of the gloves, (2) removal of gloves during mechanical adjustments and (3) wrong handling of contaminated gloves during putting on and taking off. In fact, in many field studies it has been observed that in daily practice the use of clean gloves is exceptional instead of a rule. Further, it should be noted that pesticide on the skin underneath a glove (occlusion) may well penetrate more easily due to humidity and warmth and, therefore, pose an additional hazard.

The available data bases for dermal and inhalation exposure from different data sets are given in table 4. It is obvious that a large spread in exposure levels occurs. Especially, the data on dermal exposure from [25] are relatively high and those from [28] seem indeed relatively low. Furthermore, it is clear that the format of the data differs considerably between the publications. Data are given per average working day or per hour of work and for potential or for actual exposure. The units of exposure are in weight of active ingredient or of formulation and some data are given per amount of handled pesticide. Most data bases presented in this table use unpublished reports. For these reasons it seems imperative to review all available data in the literature to establish a generic data base. This has in fact already been done by Hackathorn and Eberhart [23], but their publication does not provide enough detail. Similarly, a review has been made by Crome and the data base has been constructed by Turnbull *et al.* [22]. Both data bases were established several years ago and apparently did not review all the published literature available at that time.

### *Application*

The differences between potential exposure and actual exposure may be quite large for the various techniques of application, much larger than for mixing and loading. In section 4.1 some data have been given on the distribution of exposure of the body as observed in the work described in [19]. According to Maddy *et al.* [28], for all applications considered (tractor-driven and

airplanes), the average contamination measured on clothing is about 23%. On the average 43% is measured on the hands. The other 34% is found on head, face and neck. It should be mentioned, however, that these data pertain to well-trained workers. Further reasons for differences shown in the literature may be due to differences in working methods and to the large differences in equipment used for application (size of machinery, presence of a cabin). Reinert and Severn [25] present data showing that on the average 60% of the total dermal exposure during ground-boom application pertains to the hands. Further, on the basis of a broad data set, it is shown that the total dermal contamination per unit of time during airblast spraying in orchards shows a good correlation with the application dose. This once more indicates the importance of knowledge of application dose or amount of pesticide handled. In table 5 the published data bases for exposure during application are given with reference to the technique used. It is evident that for many application techniques no data base has been published. Furthermore, it may be seen that the data format is quite different for the various data bases. This makes comparison difficult. It should be noted, however, that for a registration procedure, the requested application doses for specific applications are available and, therefore, the various data bases can be used to calculate the expected dermal and inhalation exposure.

From the spread in the data it can be concluded that a critical evaluation of the data to be used in the development of generic or surrogate data is very important. Every single exposure study should be considered before inclusion in such data bases can take place.

Table 4 Data bases for exposure during mixing and loading

Dermal* exposure	Inhalation* exposure	Comments	Reference
<i>Outdoors (powders)</i>			
<b>Actual exposure</b>			
15 (1-40) mg/h	0.03 mg/h (0.003-0.3)	mean (range) liquids and powders	[13]
considered 0	0.09 mg/kg	geometric mean	[21]
	0.44 mg/kg	90-percentile	[21]
10.6-624 mg/h f.	<0.22-12.5 mg/h f.	range	[22]
10-100 mg/h	no data	range liquids and powders	[24]
510 mg/h (39-3000)	no data	mean (range)	[25]
<b>Potential exposure</b>			
no data	<1 mg /oper.	75-percentile	[19]
5.8 (0.3-94.5) mg/day (7 h)	no data	median (range) liquids and powders	[28]
<i>Outdoors (liquids)</i>			
<b>Actual exposure</b>			
15 (1-40) mg/h	0.03 mg/h (0.003-0.3)	mean (range) liquids and powders	[13]
considered 0	0.0005 mg/kg	geometric mean	[20]
	0.0014 mg/kg	90-percentile	[20]
0.70-615 mg/h f.	0.004-0.6 mg/h f.	range	[22]
10-100 mg/h	no data	range liquids and powders	[24]
7800 mg/h (27-32000)	no data	mean (range)	[25]
<b>Potential exposure</b>			
<1 ml /oper.	undetectable	75-percentile (10-20 l containers)	[19]
5.8 (0.3-94.5) mg/day (7 h)	no data	median (range) liquids and powders	[28]
<i>Indoors</i>			
no data base available			

\* active ingredient, unless stated otherwise;

oper. = handling of a single product pack;

f. = formulation.

Table 5 Data bases for exposure during application

Dermal* exposure	Inhalation* exposure	Comments	Reference
-----			
<i>1a Downward spraying (tractor mounted equipment)</i>			
<u>Actual exposure</u>			
1 (0.2-4) mg/h	0.015 mg/h (0.004-0.08)	median (range) appl. dose 0.3 kg/ha	[13]
0.53xAxR mg/day	0.002xAxR mg/day	geometric mean	[21]
0.003-17.3 ml/h	0.00003-0.014 ml/h	40-1000 l/ha	[22]
2.6-18.8 ml/h	0.002-0.04 ml/h	> 1000 l/ha	[22]
3.5 mg/day (0.175-7523)	no data	median (range)	[28]
<u>Potential exposure</u>			
10 (0-50) ml/h	0.01 (0-0.2) ml/h	75-perc. (range) hydraulic nozzles	[19]
2 (0-10) ml/h	0.005 (0-0.01) ml/h	75-perc. (range) rot. disc atomizers	[19]
24.4 (1-130) mg/h	no data	low boom	[25]
<i>1b Downward spraying (aerial equipment)</i>			
<u>Actual exposure</u>			
0.0009-0.016 ml/h	0.00005 ml/h	10-100 l/ha	[22]
0.57 mg/h (0.06-2.3)	0.015 mg/h (0.001-0.199)	mean (range) appl. dose 0.7-1.7 kg/ha	[23]
6 (0-17.7) mg/h	no data	mean (range)	[28]
<i>2 Soil fumigation (outdoors)</i>			
no data base available			
<i>3 Application of granules (outdoors)</i>			
no data base available			

the table is continued on the next page



Table 5 Data bases for exposure during application (continued)

Dermal* exposure	Inhalation* exposure	Comments	Reference
-----			
4 <i>Upward spraying outdoors (tractor mounted equipment)</i>			
<u>Actual exposure</u>			
20 (5-100) mg/h	0.08 mg/h (0.02-0.4)	median (range) appl. dose 0.3 kg/ha	[13]
1.7xAxR mg/day	0.02xAxR mg/day	geometric mean	[21]
2.2-436 ml/h	0.002-0.4 ml/h	> 1000 l/ha AB	[22]
1.37-178 ml/h	0.018-0.12 ml/h	> 1000 l/ha	[22]
1.02-13 ml/h	0.012-0.039 ml/h	50-1000 l/ha	[22]
0.013-1.41 ml/h	0.0011-0.063 ml/h	< 50 l/ha	[22]
0.7-69.7 mg/h	0.01-1.6 mg/h	range {OP}	[23]
		appl. dose 0.56-3.4 kg/ha	
0.4-150.6 mg/h	0.001-0.17 mg/h	range {CHC}	[23]
		appl. dose 1.1-3.4 kg/ha	
0.3-250.1 mg/h	0.003-0.29 mg/h	range {OP}	[23]
		appl. dose 3.4-4.5 kg/ha	
<u>Potential exposure</u>			
2.4-755 mg/h	no data		[27]
5 <i>Spraying outdoors (manual; upward and downward)</i>			
<u>Actual exposure</u>			
2-353 ml/h	0.002-1.4 ml/h	> 1000 l/ha	[22]
0.016-3.1 ml/h	0.00008-0.012 ml/h	50-1000 l/ha	[22]
<u>Potential exposure</u>			
50 (0-200) ml/h	0.015 (0-0.2) ml/h	75-perc. (range)	[19]
6 <i>Spraying indoors (upward and downward)</i>			
<u>Actual exposure</u>			
1.2 (0.2-6) mg/h	0.009 mg/h (0.015-0.45)	median (range) appl. dose 0.3 kg/ha	[13]
0.19-3.19 ml/h	0.00007-0.142 ml/h	100-1000 l/ha	[22]
7 <i>Ultra-low volume spraying (indoors)</i>			
no data base available			

the table is continued on the next page

Table 5 Data bases for exposure during application (continued)

Dermal* exposure	Inhalation* exposure	Comments	Reference
	8	<i>Soil fumigation under plastic cover (indoors)</i>	
no data base available			
	9	<i>Dusting (indoors)</i>	
no data base available			
	10	<i>Application of granules (indoors)</i>	
no data base available			
	11	<i>Fumigation of enclosed locations</i>	
no data base available			
	12	<i>Disinfection of seeds and bulbs</i>	
no data base available			
	13	<i>Spraying of animals</i>	
no data base available			
	14	<i>Dipping of animals</i>	
no data base available			
<hr/>			
*	active ingredient (mg) or spray liquid (ml)		
{OP}	=	organophosphorous compounds	
{CHC}	=	chlorinated hydrocarbons	
AB	=	air blast application	
A	=	area (ha/day)	
R	=	application dose (kg/ha)	

## 5 USE OF SURROGATE EXPOSURE DATA FOR REGISTRATION PROCEDURES

For registration procedures, data on potential exposure must be transformed to actual exposure and subsequently to uptake into the body. The result must be compared with toxicological data. This is a difficult process. It requires as much specific knowledge about the compound and its use in practice as can be obtained.

From the data of Batel [13] it is evident that many factors affect the level of exposure in the field and it appears that many uncontrollable variables (*e.g.* climatic conditions) vary in such a way that a large spread (see the data in tables 4 and 5) in exposure will result. This is a very important conclusion for the use of generic data in the process of registration of pesticides, as such variations have to be considered as occurring in the actual farming practice. This is a good reason for not using the average exposure values as a guide for registration procedures, but for using high percentiles, *e.g.* 90-percentiles. A further reason for the use of high percentiles is given by the fact that higher exposures are unlikely to occur in field conditions with good agricultural practice. The proper use of a high percentile (*e.g.* 90-percentile) in registration procedures will lead to work conditions in which the health and safety of the worker are not affected, as is required by Dutch law (see *e.g.* [1]).

The collection of data from different sources leads to the statistical problem that these data cannot be considered as a homogenous data set and it is therefore not possible to obtain valid percentiles and arithmetic or geometric means. Nevertheless, for pragmatic reasons that pertain to registration procedures, an estimate can be made of the 90-percentile, being a value on the higher side of the range of exposure data but not the highest value measured. This requires again that the data base be developed with care, excluding unreasonably high exposure values due to use of non-validated methods or obtained during unusual or incorrect work practice. This may introduce some degree of subjectivity, which it is hoped goes hand in hand with professional judgement.

Assuming that the toxicological information is such that acceptable levels ('no-effect' levels) for oral, inhalation and dermal daily intake are available for man, the comparison with surrogate inhalation exposure data is relatively simple and straightforward, using available knowledge on the percentage active ingredient in the formulation. On the basis of such a comparison it may be necessary to get more precise information on actual exposure for more specific conditions of the processes involved. As a second step it may be useful to consider the original literature for directly comparable situations. In a third step a study of exposure in representative situations for the particular

compound may be required.

If this does not lead to a health-based acceptable 'matching' of exposure and toxicological level, further measures must be taken to lower the level of exposure, which reflect aspects of form of formulation, packaging, techniques and, finally, use of specific protective measures.

If an inhalation 'no-effect' level is not available, the oral level may be considered for comparison, using data on differences in percentages of oral and inhalation absorption (taken as 100 %, if no data are available) and data on possible differences in toxicokinetics for these different routes of exposure.

For dermal exposure the situation is more complex. First of all, the actual exposure has to be estimated from the potential exposure. In practice this means that equality has to be assumed, unless adequate data are available on differences between potential and actual exposure under conditions in practice for the specific processes involved. From here, the analysis is straightforward unless a dermal 'no-effect' level is lacking. In this case a strategy may be followed as indicated above for inhalation. This will always mean that data are required on dermal penetration. If these are not available, it may be assumed, for safety reasons, that the amount of the compound under consideration, which is deposited on the skin, will be completely absorbed. For most compounds complete absorption is unlikely to happen in practice, especially for the relative high amounts taken as surrogate exposure (such as the above-mentioned 90-percentile). An absorption of 10 % is sometimes used in calculations if no data are available indicating higher or lower dermal absorption [19,21]. It should be noted, however, that the dermal penetration process cannot be described adequately in terms of percentage absorption, since the amount penetrating will depend on the area of the skin involved, the amount of pesticide present on the skin acting as a 'driving force' for penetration, the duration of the presence on the skin as well as on many other aspects related to the worker (skin) and the work situation.

If detailed information on dermal toxicokinetics is available, more elaborate and direct techniques, such as biological monitoring, can be used to obtain relevant data on internal exposure.

## 6 CONCLUDING REMARKS

Several data bases on dermal and inhalation exposure to pesticides during mixing and loading as well as during application have been presented in the literature. No data bases are presented for exposure after re-entry.

The spread in the data is relatively large. This is mainly due to the use of different data sets. Possible other reasons for the variations among the data bases are different criteria for inclusion of exposure data and differences in the climatic and agricultural conditions during the exposure studies which are included.

It is considered very important to review all exposure studies that are open for careful evaluation. For this evaluation the following aspects are important:

- differentiation according to the various types of (1) mixing/loading (see table 4), (2) application (see table 5) and (3) re-entry (not considered so far);
- agricultural conditions (area treated per day, crop, level of mechanization, application dose, formulation type, type of packaging);
- climatic conditions (temperature, relative humidity, wind conditions);
- work aspects (protective equipment, work practices, personal hygiene);
- analytical chemical aspects (sampling methods, reproducibility, recovery, treatment of samples).

In following parts of this series the open literature on exposure studies in agricultural practice will be reviewed, with emphasis on the various factors that may affect exposure and that have been described in this first part of the series: General aspects.

It may well be that a large majority of the published papers does not meet at least some of the standards that can be derived from the above-mentioned criteria, due to insufficient details given. The exposure data should be in units of volume (liquid formulations and spray liquid) or units of weight (solid formulations or applied dusts). It would be best if the data are also given as a ratio to the amount of pesticide handled, since there appears to be a strong correlation between level of exposure and amount of pesticide handled.

Many reports contain data on exposure, but the majority of these publications is not freely available to researchers since the data belong to firms which produce or trade pesticides in pure form or in formulations. The reports are presented to governments for registration procedures but stay confidential, and thus cannot be considered by the author. The picture that emerges from a review of only the open literature may, therefore, be distorted to a certain extent.

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