



# Hand Wash and Manual Skin Wipes

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Hand wash and skin wipes are major techniques that have been used for dermal exposure sampling. Both techniques remove chemicals either deposited on or transferred to the skin contaminant layer by a combination of chemical and mechanical actions. The paper overviews identified methods and techniques, with emphasis on sampling parameters and sampling efficiency. It is concluded that identified sampling protocols, including sampling techniques, deviate at possible key issues, which hampers comparisons of study results. It is recommended to conduct sampling efficiency studies prior to field sampling, under conditions that are quite similar to conditions of exposure regarding exposure process, levels of skin loading, and time of residence of the compound on the skin. Harmonization of sampling protocols will be a first step in creating a database for better understanding the influence of sampling parameters on the performance of removal techniques to assess dermal exposure. © 2000 British Occupational Hygiene Society. Published by Elsevier Science Ltd. All rights reserved.

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## GENERAL INTRODUCTION

Removal techniques, i.e. removal of chemicals deposited on the skin by washing, wiping, or stripping and consequently chemical analysis of the amount of chemical recovered from the washing solution, or wiping or stripping medium, have been used for decades to assess dermal exposure. Referring to the proposed conceptual model for assessment of dermal exposure (Schneider *et al.*, 1999), the amount of chemical removed actually represents the amount of the chemical which is present in the skin contaminant layer compartment at the time of sampling, i.e. the result of *loading*, e.g. by direct emission from the source, deposition, and transfer, *unloading*, e.g. by removal or resuspension/ evaporation, and *uptake* processes.

Since the techniques have the clear advantage of low capital costs and ease of use (Anon, 1998), a

widespread use of removal techniques can be observed. Removal techniques have been used mostly in studies to assess dermal exposure to pesticides (Durham and Wolfe, 1962; Davis *et al.*, 1983; Fenske, 1993; Ness, 1994). In addition, these techniques have also been applied in studies to assess exposure to metal compounds, e.g. Roels *et al.* (1980); Linnainmaa and Kiilunen, (1997), PAHs, e.g. Cheng (1981), and PCBs, e.g. NIOSH (1986). However, most studies present no or very limited information on sampling performance characteristics, e.g. sampling efficiency.

The objective of this paper is to evaluate removal techniques with emphasis on sampling parameters, sampling efficiency, and sampling strategy by reviewing exposure studies which provide information on the removal technique that has been used. Because of the relevance of the dermal route most of these studies have been done in the context of exposure to pesticides during and following use. However, the evaluation of the removal techniques will not be limited to its use to assess pesticide exposure.

Since skin stripping has been used primarily for skin absorption studies, the evaluation is limited to

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hand wash sampling and skin wipe sampling. Both techniques are discussed separately.

## HAND WASHES

### *Sampling principles*

Removal of contaminants from the skin surface is accomplished by providing an external force that equals or exceeds the force of adhesion. For hand washing three categories of external forces can be distinguished: mechanical action, hydrodynamic drag, and wet chemical action.

Generally, two basic methods can be identified:

1. (*Hand*)*washing* can be defined as scrubbing the skin by mechanical agitation exercised by movements and pressure of both hands in liquid in a routine washing fashion. The contaminant is detached from the skin by a combination of mechanical forces and wet chemical action (dissolution).
2. (*Hand*)*rinsing* or pouring can be defined as liquid–skin contact, where the contaminant is removed by a combination of hydrodynamic drag, and wet chemical action (dissolution).

Clearly, the basic distinction between both methods is the presence or absence of mechanical forces in the process of detachment. Within both methods subcategories can be distinguished using flowing or contained liquid (determines the strength of hydrodynamic drag), and the kinds of liquid (determines the strength of solubility). Often detergents are introduced in the process to enhance the detachment of insoluble particles.

### *Materials and methods*

*Wash/rinse procedures.* During bag rinsing one hand is immersed in solvent and a technician holds the bag tightly just above the wrist to prevent leakage. The person should cup the hand slightly and hold the fingers a short distance apart during most of the shaking operation. Occasionally, the thumb and fingers should be rubbed against one another and against the palm. The hand should be shaken vigorously, either by the person or by the technician during a fixed time, e.g. 30 s (Fenske and Lu, 1994; Fenske *et al.*, 1998; Brouwer *et al.*, 1992, 2000a, b), a fixed number of shakes, e.g. 50 times (US EPA, 1986), or a fixed number of shakes (60) in a fixed time (30 s) (Fenske *et al.*, 1999).

During hand washes the subject is asked to wash his hands thoroughly in a routine fashion or according to a 6-step procedure (CEN, 1996). Brouwer *et al.* (1992, 2000a, b) used a procedure for solvent based routine fashion hand washing in a bag, where workers were asked to wash their hands for 30 s. After removal from the solvent the hands were allowed to

dry above the solvent for 10 s before removal from the bag, and the procedure was repeated a second time in a fresh handwash solution. Marquart *et al.* (2000) also used the routine fashion hand washing for a tap water/soap-based method. After transfer of approximately 1.5 ml of a hypoallergenic soap from a dispenser onto the palm, the hands are moisturized by supplying a small amount of water. A hand wash procedure is performed by the worker in a routine fashion for 15 s. Consequently the hand wash procedure is repeated for 30 s during which the hands are kept in a water flow of approximately 3 l/min, after which the hands are allowed to drain above the funnel for 10 s. After rinsing the funnel using deionized water and replacing the container the 30 s hand washing in the water flow is repeated.

*Solvents and equipment.* Wash liquids may vary between tap water, distilled or deionized, possibly in combination with a commercially available surfactants, e.g. Sur.Ten, (US EPA, 1996), commercial (hypoallergenic) liquid hand soaps, and organic solvents. Organic solvents with mild skin irritative effects, such as neat alcohols, e.g. ethanol, 2-propanol (isopropanol), may be used pure or as a solution (10% or 40% w/w in water) (US EPA, 1996).

Bags are selected that are sturdy enough to hold 250 or 500 ml of solvent, e.g. commercially available Zip-Loc™, or 'home made' polyethylene bags (0.025 mm thickness) (US EPA 1986, 1996; Brouwer *et al.*, 1992; Brouwer *et al.*, 2000a). No information is available on the type of bowls used.

A specially developed hand washing device has been designed for hand washing with tap water flow (Marquart *et al.*, 2000). This device consists of a tube attached to the water tap of the water supply, an adjustable flow control set at a flow rate of approximately 3 l/min and a timer, a tap, a funnel and a 5 l. polyethylene bottle to collect the rinse water.

### *Sampling efficiency*

Hand wash sampling efficiency tests are recommended (OECD, 1997; Fenske and Lu, 1994; Fenske *et al.*, 1998), but a standard approach has not been adopted. Two different approaches have been identified.

*Mass balance approach* (Fenske and Lu, 1994; Fenske *et al.*, 1998). The contaminant is transferred from a surface with a known amount of contaminants to the hands of human volunteers by a standardized contact. The amount of contaminant that has been transferred is estimated from the difference between the amount of contaminant which was not removed from the surface and the amount initially spiked. This procedure has been designed to mimic non-liquid contamination by surface to skin transfer.

*Direct spiking* (Brouwer *et al.*, 1992, 2000a, b). 0.5 ml of a water-diluted (pesticide) formulation

is spiked on the hands of a human volunteer. This procedure mimics exposure resulting from liquid exposure (spills, aerosol deposition).

In Table 1 hand wash efficiency results derived by different procedures for several compounds are summarized. Generally, results are from three human volunteers. For 12 compounds (all but one pesticide) 28 sampling efficiency data were reported, ranging from 23 to 96% (median 73%).

#### Sampling parameters

An important variable within the sampling efficiency validation procedure is the residence time, i.e. the time elapsed between contamination of the hands and the start of the wash sampling. Fenske and

Lu (1994) and Fenske *et al.* (1998) used 0 and 1 h, whereas Brouwer *et al.* (2000a, b) and Marquart *et al.* (2000) standardized the time of residence to 0.5 h. Fenske and Lu (1994) and Fenske *et al.* (1998) reported a significant decrease of sampling efficiency with prolonged time of residence for chlorpyrifos and captan, respectively. The mean sampling efficiency of two washes of chlorpyrifos decreased from 43% immediately following exposure to 23% 1 h after exposure, whereas for similar conditions the removal efficiencies for captan decreased from approximately 78% to approximately 68%. Therefore, residence time is considered to be of major importance for contaminants that are well absorbed by, or adsorbed to the skin.

The level of skin loading, i.e. mass of contaminant present on the hand(s), may also affect the sampling

Table 1. Results from hand wash sampling efficiency studies

Substance	Method <sup>a</sup>	Loading ( $\mu\text{g}$ )	Wash efficiency (%)		N	References
			Mean	STD		
Captan	B	1500 <sup>b</sup>	94	11	4	Brouwer <i>et al.</i> (2000a)
		15 000 <sup>b</sup>	63	13	4	
	B	5620 <sup>c</sup> (1 h)	68	5	6	Fenske <i>et al.</i> (1998)
	B	5250 <sup>c</sup> (0 h)	78	14	3	
Carbendazim	B	500 <sup>b</sup>	94	8	3	Brouwer <i>et al.</i> (2000a)
		5000 <sup>b</sup>	59	13	3	
Chlorothalonil	A	4400 <sup>b</sup>	74	11	4	Brouwer <i>et al.</i> (2000a)
Chlorpyrifos	B	1700 <sup>c</sup> (0 h)	43	24	6	Fenske and Lu (1994)
	B	1570(1 h)	23	9	6	
	C	1100(1 h)	27	5	6	
Mancozeb	A	2275 <sup>b</sup>	81	10	4	Brouwer <i>et al.</i> (2000a, 1992)
	B	2275 <sup>b</sup>	66	5	5	
	D	5000, 15 000, 30 000 <sup>b</sup>	86	5	12	
Methiocarb	D	500 <sup>b</sup>	77	3	4	Brouwer <i>et al.</i> (2000a)
		1800 <sup>b</sup>	84	3	4	
		7000 <sup>b</sup>	84	6	4	
Methomyl	D	300 <sup>b</sup>	71	3	4	Brouwer <i>et al.</i> (2000a)
		1490 <sup>b</sup>	70	4	4	
Prochloraz	B	500 <sup>b</sup>	95	14	4	Brouwer <i>et al.</i> (2000a)
		5000 <sup>b</sup>	96	6	4	
Propoxur	D	175 <sup>b</sup>	66	8	4	Brouwer <i>et al.</i> (2000a,b)
		575 <sup>b</sup>	71	13	4	
		1400 <sup>b</sup>	72	10	4	
	D	500,5000,7500 <sup>b</sup>	46	3	12	Marquart <i>et al.</i> (2000)
Vinclozolin	D	59.2, 227, 384 <sup>b</sup>	81	5	3	Brouwer <i>et al.</i> (2000a,b)

<sup>a</sup>(A) 2-propanol rinsing, 2 hands 500 ml, PE-bag; (B) 2-propanol rinsing, 1 hand 250 ml, PE-bag; (C) ethanol rinsing, 1 hand 250 ml, PE-bag; (D) water-soap rinsing, 2 hands, tap water; N=number of test subject per level of loading.

<sup>b</sup>Direct repeated spiking of 0.5 ml on the hands.

<sup>c</sup>Mass balance approach from transfer of a contaminated tube.

efficiency. Decrease of sampling efficiency for lower levels of skin loading is hypothesized on the assumption that a fixed amount of the contaminant may bind to the skin strongly, whereas on the other hand at very high levels of skin loading sampling efficiency may be affected by the limited ability for a complete dissolution of the contaminant in the wash liquid. In particular, this may be the case for bag rinsing with a limited amount of liquid. A thorough evaluation of some studies shows some, but not consistent, evidence for the assumption of decrease of removal efficiencies for low skin loadings (Boeniger and Brouwer, 2000). No data are available to evaluate the influence on sampling performance for similar (total) mass of contaminant present on the hand but for different surface area exposed, or similar surface area exposed for different mass of the contaminant, i.e. different amounts of contaminant per surface area contaminated ( $\mu\text{g}/\text{cm}^2$ ). This relates to the inability of the hand wash/rinse method to assess the area exposed. For assessment of skin absorption of a compound the surface area contaminated is one of the key parameters (Cherrie and Robertson, 1995; Bos *et al.*, 1998).

The method of contamination of the hands and the chemical and physical state of the contaminant may also affect the efficiency of removal. Theoretically, mechanical transfer of a dry contaminant (Fenske and Lu, 1994; Fenske *et al.*, 1998) and liquid spiking (water diluted (Brouwer *et al.*, 1992; Brouwer *et al.*, 2000a, b), or solvent diluted (pure substance or formulation) may affect the strength of binding of the contaminant to the skin, and therefore the sampling efficiency. However, no data are available to draw conclusions on this issue.

Another variable is the number of consecutive washes performed. In the sampling efficiency protocol of Brouwer *et al.* (1992, 2000a, b) and Marquart *et al.* (2000) three consecutive wash procedures are used in a laboratory setting, but in field practice two consecutive washes are performed. Fenske and Lu (1994) and Fenske *et al.* (1998) used two washes. From analysis of the washes the contribution of the first wash to the total removal is in the range of 75% to approximately 90%.

The wash time is another variable which may determine the sampling efficiency. Both Fenske and Lu (1994) and Brouwer *et al.* (1992, 2000a, b), standardized the washing time at 30 s. However, EPA bag rinsing methods standardize the number of strikes (50) instead of time (US EPA, 1996). Recently, Fenske and coworkers (Fenske *et al.*, 1999) conducted a study where the wash protocol includes number of shakes (60) within 30 s.

As has been indicated washing fashion/time and rinsing time are potential variables. Brouwer *et al.* (2000a,b) standardized the washing time, i.e. the soap-contact duration, at 30 s, but allowed the volunteers to use their own routine fashion of washing. In

a protocol for claims and tests for chemical disinfectants in human medicine, the wash routine has been prescribed but no limitations are given to the soap-hand contact time (CEN, 1996).

For solvent rinsing the type of solvent will also affect sampling efficiency (Fenske and Lu, 1994). In a study with chlorpyrifos the authors observed at  $t=0$  a significantly higher sampling efficiency for the isopropanol/water wash compared to the ethanol wash, whereas the reversed were observed at  $t=1$  h.

For water/soap methods the type and amount of soap may also account for variation of the sampling efficiency. Brouwer *et al.* (2000a,b) and Marquart *et al.* (2000) tried to standardize the soap type by using Sporex (Kimberly Clark) which is a mildly acidic, hypoallergenic liquid soap, and the amount of soap used to approximately 1.5 ml by application using a dispenser.

Finally, the water hardness (differences between laboratory and field situation) and the temperature of the solvent may be variables which may affect removal efficiency, however no data are available to substantiate this.

#### *Sampling strategy*

OECD (1997) recommends performing hand wash sampling whenever the worker normally washes his/her hands (i.e. every two to three hours before breaks and at the end of the working day). However, the aim of the sampling may also be to assess hand exposure during specific tasks or consistency or variability through consecutive replicate sampling of the same workers, which demands end of task or periodic sampling.

Both OECD and US EPA (1986) recommend a (solvent) pre-washing prior to the start of the task, to remove background contaminants.

No recommendations are given on the sampling interval or number of rinses using organic wash solvents. However, washing or rinsing the hands with a solvent is considered by some to disrupt the barrier function and enhance percutaneous absorption of the contaminant (Froebe *et al.*, 1990; Hilton *et al.*, 1994; Hatanaka *et al.*, 1995; OECD, 1997). In case of exposure to relatively volatile or rapidly penetrating compounds, short sampling intervals should be considered.

#### **SKIN WIPING**

Wipe sampling methods for skin and surfaces have been reviewed by McArthur (1992) and Ness (1994). Basically, most methods used by different investigators are similar to or adjustments of the general methods published by OSHA (1984, 1990) and US EPA (1985, 1996). Most of these adjustments are primarily related to surface sampling, e.g. the sampling medium, i.e. type, surface size are related to

surface sampling, the absence or presence of 'wetting liquids' in the sampling medium, i.e. a dry medium, a wetted medium or a soaked medium and the kinds of liquids used (water (deionized/distilled), organic solvents), the size of the sampled surface area and the use of templates, and the type of wiping, i.e. rectangularly or circularly, and the number of transverses. Despite similarities of skin wiping with surface wiping, the variation of the skin surface (location, roughness, porosity, shape) is limited compared to surfaces. For skin sampling the locations of sampling mostly is limited to the hands (palms, fingertips), forearms, and forehead. In addition, the variation of sampling media and wetting liquids are limited, because skin wiping requires special consideration to protect the skin layer.

#### *Sampling principle*

Skin wiping can be defined as the removal of contaminants from skin by providing manually an external force to a medium that equals or exceeds the force of adhesion over a defined surface area. Similar to hand washing, the contaminant is detached from the skin by a combination of mechanical forces and wet chemical action (dissolution).

#### *Materials and methods*

*Skin wipe procedures.* Fogh *et al.* (1999) used two different methods to wipe tracer particles from the forearms of volunteers. In one experiment cotton fabric wipes were used with a surface area (of the wipe) of approximately 10 cm<sup>2</sup>. Wipes were soaked in distilled water. After exposure, a rigid steel template with a circular hole of diameter 4 cm (within which the skin is wiped), was placed on the arm of the exposed person and held in place by the unexposed individual. The unexposed individual blots excess moisture from the wipe, and conveys it, via forceps, to the gloved hand of the exposed individual, who wipes his own arm. Consequently the used wipe was transferred to a clean labelled plastic bag and the wiping was repeated five times. In another experiment for similar exposure a second procedure was followed. Whatman 542 filter paper, soaked in ethanol, was used as wipe medium with an area of the individual wipes of approximately 8 cm<sup>2</sup>. After exposure a template is placed on the arm of the exposed person and held in place by an unexposed individual. The template is made of flexible aluminium, with a rectangular hole of 8 cm<sup>2</sup>, within which the skin is wiped. The unexposed individual blots excess moisture from the wipe, and rigorously wipes the arm of the volunteer. The used wipe is transferred to a clean labelled plastic bag, and the wiping is repeated three times.

Meuling *et al.* (1991) used a cotton ball as a wipe medium with a surface area of approximately 4.8 cm<sup>2</sup>. Prior to exposure a flexible template 20×5 cm is used to mark a surface area at a flat skin surface, e.g. fore-

arms. Cotton roll plugs were soaked in ethanol and wiped over the skin surface by the operator in one stroke. The operator is provided with vinyl gloves. After wiping the plug is transferred to a labelled disposable PE-tube, and the wiping was repeated ten times.

For wiping hands Geno *et al.* (1996) use commercial wetting sponges. The sponge was wetted with 10 ml 2-propanol and the subject was instructed to perform a general wipe of the hands, after which the subject placed the sponge in a glass jar. A second sponge was wetted and the subject was instructed to thoroughly wipe each individual digit and the palm of the hand.

Fenske *et al.* (1999) used 12-ply cotton surgical pads. Pads were wetted with water containing 1% surfactant. For assessment of hand exposure three separate wipes were used, i.e. one for the palm, one for the back of the hand, and one for the fingers and the thumb.

*Sampling medium, 'wetting' liquids and templates.* For skin wipe sampling a cotton fabric-based sampling medium has been used most typically, however cellulosic Smear Tabs has also been used to collect PCBs from hands of transformer repair workers (Lees *et al.*, 1987). For skin sampling wetted or soaked sampling media are used. In most cases the sampling medium was soaked in deionized water or neat alcohols, e.g. ethanol and isopropanol, since these solvents have good solubility for most compounds and are not overly irritating to the skin. However, skin irritation may not be excluded with repeated exposure, which may be indicated by a remarkable increase (twofold) of sampling efficiency over a period of three consecutive days of sampling, also reported in a study conducted by Geno *et al.* (1996). In a study to compare four solvents for their ability to remove selected agricultural pesticides from an in vitro porcine model, polyethylene glycol, soap and water, and a commercial decontamination product (D-TAM™) were used, in addition to 1-propanol (Campbell *et al.*, 2000). In that study repeat wipe sampling also appeared to affect subsequent recovery.

With the exception of sampling hand skin surface, for most other skin surfaces templates are used to mark the surface area that should be wiped. The surface area depends on the body part. Circular, rectangular or square templates are used (Meuling *et al.*, 1991; Campbell *et al.*, 2000; Fogh *et al.*, 1999). The shape of the sampling medium is often related to the choice of shape of the template.

#### *Sampling efficiency*

As stated before, sampling efficiency is a key factor using wipe sampling results for assessing dermal exposure. In general, similar approaches are needed as used for the determination of hand wash efficiency, i.e. a mass-balance approach and direct spiking. Geno *et al.* (1996) used a mass-balance approach to deter-

mine sampling efficiency for two pesticides. A solution of pesticides was spiked on a 15×15 square aluminum foil. After drying volunteers pressed the foil and the residue remaining on the foil was analysed. The difference between applied and recovered mass was considered the mass transferred to the hands.

Meuling *et al.* (1991) used direct spiking of 0.3 ml of a pesticide solution (50% v/v water/methanol) on 100 cm<sup>2</sup> surface area of the forearms of 15 volunteers. They conducted an experiment with volunteers (*N*=15) using commercial swabs soaked in a water/ethanol mixture. Wiping was performed by one single operator. Since these experiments were conducted as part of a dermal penetration study, the time of residence was 4 h.

In the study by Campbell *et al.* (2000) the wipe sampling efficiency for four (radio-labeled) pesticides, at three levels was tested for four solvents. A direct spiking method was used to apply the pesticides to *in vitro* porcine skin. Two drops (40 µl) of a formulation were spread over 6.45 cm<sup>2</sup> skin. Each solvent, pesticide, and skin loading combination was repeated three times. The time of residence (interval between application and wiping) was 90 min.

The overall recoveries (mean of four pesticides) for the solvents (*N*=36) showed considerable differences and ranged from 45% (D-TAM™) to 57% (1-propanol), but was pesticide/wipe solvent dependent. For 14 out of 16 pesticide–solvent combinations a significantly lower sampling efficiency was observed for the lowest skin loading level compared to the highest level. The most water soluble pesticide (glyphosate) could be removed best by water/soap wipe solvent, but showed to be removed less effectively by the wipes compared to the other pesticides.

During the Risø/Imperial College studies (Fogh *et al.*, 1999), volunteers wiped excised rat skin which has been exposed to 4.5 µm particles labelled with neutron activated tracers, and which has consecutively fixed to the volunteers skin. Removal efficiencies ranged from 15 to 30%.

In Table 2 wipe sampling efficiency results derived by different procedures for several compound are summarized. For seven pesticides 19 sampling efficiency data were reported, ranging from 36 to 104% (median 51%).

#### *Sampling parameters*

The number of passes, i.e. the number of contacts of the wiped area with a single wipe, varies from 1 (Meuling *et al.*, 1991; Fogh *et al.*, 1999) to 15 (Campbell *et al.*, 2000). However, in field practice one pass seems to be quite common, but in these cases consecutive samples, i.e. the same surface is wiped using different wipes, are taken, e.g. two by Geno *et al.* (1996), three or six by Fogh *et al.* (1999), and ten by Meuling *et al.* (1991). It is clear that the performance of the method, e.g. the recovery or wipe

efficiency, strongly depends on the number of passes and/or number of consecutive samples.

Between-operator variability, e.g. resulting from pressure differences, has been recognized to be an important variable, so often only one operator will be involved in a study to maintain consistency. For skin wipe sampling, sometimes the volunteer involved in the study acts as the operator (Geno *et al.*, 1996; Fogh *et al.*, 1999), which may introduce an unknown within-person variability to removal efficiency.

In addition to the type of wiping which may include repeated contact between the sampling medium and the (skin) surface, the surface area of the contact surface could be a variable which affects removal efficiency. For example, the use of a 4.8 cm<sup>2</sup> cotton plug for wiping a 100 cm<sup>2</sup> surface (Meuling *et al.*, 1991) will result in a ratio of approximately 1:20, whereas the use of 8 cm<sup>2</sup> filter paper for a 8 cm<sup>2</sup> surface area (Fogh *et al.*, 1999) results in a ratio of 1:1. Surface loading combined with absorption capacity of the sampling medium will determine the amount of contamination that can be removed.

#### *Sampling strategy*

Timing and frequency of sampling and sampling locations etc. depends on the objectives of the sampling. Exposure assessment is the major objective for skin wipe sampling, so post exposure sampling is more or less standard. Therefore, wipe sampling efficiency tests to assess exposure should be conducted at times of residence relevant for realistic duration of exposure in practice, e.g. 90 or 240 min. Since the body parts that can be wiped are limited to hands, forearms, and facial and neck region, e.g. V-of the neck and forehead, the spatial distribution of contamination at the body parts is irrelevant if the entire area is wiped. When templates are used, spatial variability of exposure distribution becomes more relevant and under- or overestimation of the body part is possible by extrapolation of the results from the area sampled.

#### *Evaluation of the removal techniques*

*Hand wash/rinse techniques.* Identified sampling protocols show a reasonable similarity of procedures. However, they deviate at possible key issues, e.g. amount of liquid and duration of rinsing (bag rinsing), amount of liquid, amount of soap, duration of washing (water/soap methods). Identified protocols for the determination of sampling efficiency differ substantially, especially in the way the contaminant is applied to the hands. For almost all reported sampling efficiencies no within-substance comparison of the mass balance approach/dry transfer and direct spiking of liquids is possible. Only for captan sampling have efficiency studies been conducted using the same hand wash method (Table 1) and different loading methods; however time of residence and levels of loading differed.

Table 2. Results from wipe sampling efficiency studies

Substance	Method <sup>a</sup>	Dose level ( $\mu\text{g}/\text{cm}^2$ )	Wash efficiency (%)		References
			Mean	STD	
Alachlor	A1	0.5;2.0;8.0	57	13	Campbell <i>et al.</i> (2000) ( $N=9$ , for each method)
	A2	0.5;2.0;8.0	55	8	
	A3	0.5;2.0;8.0	52	12	
	A4	0.5;2.0;8.0	51	6	
Chlorpyrifos	B	4 <sup>b</sup>	104	11	Geno <i>et al.</i> (1996) ( $N=12$ )
Glyphosate	A1	0.5;2.0;8.0	44	12	Campbell <i>et al.</i> (2000) ( $N=9$ , for each method)
	A2	0.5;2.0;8.0	41	11	
	A3	0.5;2.0;8.0	49	14	
	A4	0.5;2.0;8.0	36	9	
Methyl parathion	A1	0.5;2.0;8.0	57	17	Campbell <i>et al.</i> (2000) ( $N=9$ , for each method)
	A2	0.5;2.0;8.0	41	18	
	A3	0.5;2.0;8.0	50	19	
	A4	0.5;2.0;8.0	41	15	
Propoxur	C	13;27;53;107;214	48	14	Meuling <i>et al.</i> (1991) ( $N=15$ , 3 for each dose group)
Pyrethrin I	B	38 <sup>b</sup>	92	28	Geno <i>et al.</i> (1996)
Trifluralin	A1	0.5;2.0;8.0	69	10	Campbell <i>et al.</i> (2000) ( $N=12$ )
	A2	0.5;2.0;8.0	51	15	
	A3	0.5;2.0;8.0	56	13	
	A4	0.5;2.0;8.0	53	14	

<sup>a</sup>(A) Cotton gauze sponge, circularly movement, 6.45  $\text{cm}^2$  contact area, wipe 90 min after application. (1) 1-propanol; (2) polyethylene-glycol; (3) soap/water; (4) D-TAM<sup>TM</sup>. (B) Cotton plugs, rectangularly movement, 4.8  $\text{cm}^2$  contact area, wipe 240 min after application. (C) Cotton gauze sponge, rectangularly movement, unknown contact area, wipe immediately following application.

<sup>b</sup>Total amount applied: no surface area exposed is given.

Because of the limited data set on removal efficiencies and large differences in components (related to physical properties), wash methods, and levels of loadings, no general conclusions can be drawn on the strengths of the variables which have been distinguished.

Limited data are available on between- and within-person variability of the sampling efficiency. Brouwer *et al.* (2000a) observed, in an experiment with four test subjects, three levels of (mass) loading and three replicates for each loading significant between-person difference in the sampling efficiency for one compound (propoxur;  $P=0.0018$ ,  $N=12$ ), but no difference for another compound (methiocarb,  $P=0.806$ ,  $N=12$ ). For the time being, data sets on sampling efficiency are too limited to afford a thorough analysis. Harmonization of procedures and protocols will be helpful to generate data which are appropriate to compare methods and compounds.

The overall uncertainty of the methods is determined by the between-person variability of the sampling efficiency, the difference between the observed sampling efficiency in the laboratory and the actual sampling efficiency (similarity of the dermal exposure process, time of residence of the contaminant on the

skin, influence of dirt and so on), the level of standardization of wash procedures, the extraction of recovery efficiency from the wash solution, and the accuracy of the chemical analysis.

All hand wash/rinse methods reveal an amount of a contaminant detached from the hands (or parts of the wrist) expressed in units of mass per body part (hand or hands). The use of other units of exposure, e.g. mass per surface area exposed ( $\mu\text{g}/\text{cm}^2$ ) introduces additional uncertainty, since in most cases the surface area is unknown or contamination of the complete surface area is assumed and the surface area of the hands is estimated (or defaults are used).

*Manual wipe sampling methods.* Identified sampling protocols deviate at possible key issues mentioned above, and a harmonized protocol is not available. Sampling efficiency will not only be determined by the sampling method, but also by the chemo-physical properties of the compound and wipe solvent. Time of residence will be essential, as indicated by the experiments of Meuling *et al.* (1991), and it can be argued whether the results of these experiments, compared to the results from the study by Geno *et al.* (1996), reflect more potential absorption of the substance by or to the skin than sampling efficiency.

Between-operator variability of the sampling efficiency cannot be determined from the experiments by Geno *et al.* (1996), because skin variables are also included. The same holds for the experiments by Meuling *et al.* (1991) for within-operator variability. The only data on within-operator variability can be determined from the experiments by Campbell *et al.* (2000), since the skin (type) was similar and the tests were performed by a single operator. The observed coefficient of variation was very low (4.7%). An overall uncertainty of the manual wipe methods is not indicated, but is estimated to be substantial, regarding different solvents, sampling media and contaminants, and different skins. Much research is needed to clarify the influence of the variables within the sampling procedures.

The use of templates implies the sampling of a discrete surface area, which enables expression of the mass removed from the skin in terms of mass per surface area exposed, e.g.  $\mu\text{g}/\text{cm}^2$ . However, extrapolation of the sampling result to non-sampled surface areas implies the assumption of homogeneous distribution of the contamination.

#### GENERAL REMARKS

Because of their ease of use and their low capital costs, the application of removal techniques is widespread to assess dermal exposure. In spite of their potential to be used for all body parts, usually the uncovered parts of the body are monitored. Especially for wipe sampling, relatively high resolution of exposure per surface area can be achieved; however for hand washing this is not the case. Repeated sampling is possible, but the exposure process is disturbed and skin surfaces may be affected.

There is clear evidence that wipe sampling is less effective to remove contaminants from the skin, despite the high removal efficiencies of wipe sampling reported by Geno *et al.* (1996) In a pesticide re-entry study Fenske *et al.* (1999) compared hand exposure rates determined by hand wash sampling and wipe sampling. They observed on average a six-fold lower hand exposure rate for wipe sampling compared to hand wash sampling. Incomplete removal by hand wipes has also been demonstrated by McCurdy *et al.* (1994) and Fogh *et al.* (1999). In both studies hand wash sampling was conducted after wipe sampling and in both studies substantial amounts of a contaminant could be washed off from the hands compared to the amount removed by hand wipes [up to 57% (McCurdy *et al.*, 1994)].

Referring to the proposed conceptual model for assessment of dermal exposure (Schneider *et al.*, 1999) the amount removed from the skin actually represents the amount of the contaminant which is present in the skin contaminant layer compartment at the time of sampling. However, the interpretation of the amount removed for exposure assessment is not

simple, since it is obvious that the sampling efficiency is affected by the time of residence of the contaminant on the skin surface. The amount removed by washing or wiping only represents the amount which is accessible by the sampling method at the time of sampling. Therefore, interpretation of these results to dermal exposure over the sampling interval includes uncertainties if a residence time-sampling efficiency relationship is unknown. Moreover, in view of percutaneous absorption processes the relevance of the amount which can be removed, not only for sampling purposes but also due to normal hygienic decontamination practice, is questionable. However, in practice it has been shown that the amount removed from the skin correlates with the amount absorbed (Meuling *et al.*, 1991; Brouwer *et al.*, 1998). Current gaps in scientific knowledge indicate that the results of sampling by removal techniques should take into consideration incomplete removal of the contaminants which needs careful interpretation.

#### RECOMMENDATIONS

When removal techniques are used to assess dermal exposure monitoring for risk assessment purposes, it is recommended to conduct sampling efficiency studies as a key issue for method performance.

Sampling efficiency studies should be designed to mimic (i) the relevant exposure process, (ii) the relevant time of residence, and (iii) the relevant levels of skin loading.

For exposure processes where surface-skin transfer predominates other processes, e.g. deposition, the mass-balance/dry transfer approach, as proposed by Fenske and Lu (1994), or Geno *et al.* (1996), seems to be appropriate. For exposure due to deposition direct liquid spiking could be appropriate; however, the composition of the spiking liquid should be close to the composition of the aerosol. In practice, e.g. for a pesticide spraying exposure scenario, water-diluted pesticide spray liquids can be used, but for some formulations, e.g. (micro-encapsulated) granules, this will not be easy to achieve.

The studies of Fenske and Lu (1994) and Fenske *et al.* (1998) clearly demonstrate the influence of time of residence on sampling efficiency. Therefore, sampling efficiency studies should address typical durations of exposure for the exposure scenario that will be evaluated.

The results of sampling efficiency studies indicate that sampling efficiency is unlikely to be constant across loading levels on skin. More levels of loading, both in terms of total mass ( $\mu\text{g}$ ) and mass per surface area contaminated ( $\mu\text{g}/\text{cm}^2$ ), should be used to determine sampling efficiency values over the expected range of skin loading. A similar approach could be followed as recommended by the OECD guidelines for fortification of field spikes for surrogate skin sam-

plers (OECD, 1997). The low and high levels of skin loading should be in the range of the anticipated level of exposure. If the highest expected level is more than 100× the lowest level, it is recommended that a mid-level loading is included. The different amounts of contaminant should be applied to a similar surface area of skin to achieve different loadings in terms of mass per surface area contaminated ( $\mu\text{g}/\text{cm}^2$ ).

To reduce variability of sampling efficiency preference should be given to those procedures that limit subject or operator depended variability. For hand washes a standardized hand wash procedure as prescribed by CEN prEN 1499 for claim testing of chemical disinfectants and antiseptics (CEN, 1996) seems more appropriate to reduce variability than a routine fashion washing by individuals.

For interpretation of hand wash sampling results for risk assessment purposes it is necessary to estimate the true surface area exposed. The use of (fluorescent) tracers in different (mass transport) exposure processes could be helpful to estimate the surface area exposed (Cherrie *et al.*, 2000).

For skin wipe sampling the influence of the operator on specific sampling parameters, e.g. pressure and wipe speed, on sampling efficiency has been recognized, but not quantified. The results of the study by Campbell *et al.* (2000) showed relatively low within-skin, and within-operator variance ( $CV < 5\%$ ), the study by Geno *et al.* (1996) showed relatively high coefficients of variation between operators for different skins (up to 39%). Therefore, it seems quite obvious to limit the number of operators within a study. However, standardization of operator related skin wipe variables, e.g. pressure and traverse speed, is needed to enable comparison of data from different studies.

Ideally, the ratio of contact surface area/skin surface area wiped should be one, to achieve the highest sampling efficiency. However, in field practice this may be difficult to meet since, often, fine-tuning of this parameter is necessary to the skin surface area that is sampled regarding the surface area exposed, homogeneous distribution of exposure, and other sampling strategic considerations. Therefore, we propose a minimum ratio of contact surface area/skin surface area wipe of 1:5, but no hard data are available to support this. Discrete surface areas to be wiped, preferably using templates, are essential to meet this sampling condition. The study by Campbell *et al.* (2000) showed that no single wetting agent for wipe media has the best performance, however 1-propanol showed the best overall sampling efficiency for different pesticide compounds.

Studies indicate that many sampling parameters affect sampling efficiency, but the differences in study protocols regarding sampling methodology result in very small databases. Investigators should try to adopt protocols that have been used or try to harmonize protocols to provide comparable data

which enable a thorough evaluation of key parameters. This would be a first step in the development of validated methods which provide accurate and reliable data on the sampling performance of removal techniques.

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