Conceptual model for assessment of dermal exposure

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Abstract
Dermal exposure, primarily to pesticides, has been measured for almost half a century. Compared with exposure by inhalation, limited progress has been made towards standardisation of methods of measurement and development of biologically relevant exposure measures. It is suggested that the absence of a consistent terminology and a theoretical model has been an important cause of this lack of progress. Therefore, a consistent terminology based on a multicompartment model for assessment of dermal exposure is proposed that describes the transport of contaminant mass from the source of the hazardous substance to the surface of the skin. Six compartments and two barriers together with eight mass transport processes are described. With the model structure, examples are given of what some existing methods actually measure and where there are limited, or no, methods for measuring the relevant mass in a compartment or transport of mass. The importance of measuring the concentration of contaminant and not mass per area in the skin contaminant layer is stressed, as it is the concentration difference between the skin contamination layer and the perfused tissue that drives uptake. Methods for measuring uptake are currently not available. Measurement of mass, concentration, and the transport processes must be based on a theoretical model. Standardisation of methods of measurement of dermal exposure is strongly recommended.
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Exposure to hazardous substances most commonly occurs either by inhalation, ingestion, dermal contact, or some combination of these routes. Occupational hygiene has traditionally focused on exposure by inhalation because it was almost invariably considered to be the most important pathway. Many methods have been developed to measure exposure levels from inhalation and there is a clear understanding of how such levels should be interpreted to help reduce risk. The situation is less clear for the dermal route of exposure. Practical methods of measurement have been developed to assess dermal exposure[1] and proposals have been made to develop dermal exposure limits in an analogous way to those for inhalational exposure.[2 3] However, there has been criticism of the existing methods of measurement of dermal exposure because they determine the mass of contaminant either depositing on the skin or retained on the skin at the end of the exposure period.[1 4]

Hazardous substances on the dermal surface will be taken up continuously into the body through the stratum corneum and the epidermis towards the dermis where they or their dermal metabolites will be removed by the blood flow. The transport process is driven by the concentration gradient between the dermal surface and the perfused tissue. The risk arising from dermal exposure is thus firstly related to the time dependent concentration of a substance on the dermal surface rather than the mass of material on that surface at any given time. Mass is nevertheless important when there is little material available for uptake.

Contamination of the skin may arise in many different ways. It is possible for hazardous substances to land on or be absorbed into the skin directly from the air. They may be transferred to the skin from contact with contaminated surfaces or by submersion of part of the body into the substance. Also, the contaminant may be lost from the skin, either by evaporation or some other mechanisms such as washing or abrasion, without being taken up into the body. Finally, the presence of clothing or protective garments may modify the rate at which hazardous substances come into contact with the skin. All of these processes are important to consider when making an assessment of dermal exposure and a complete understanding of these complex processes will help in developing an appropriate control strategy.

In this paper we have attempted to produce a consistent terminology for assessment of dermal exposure. The terminology is based on a conceptual model of the processes leading to exposure (from the source of a hazardous substance to the surface of the skin). We have also defined several terms related to exposure, which provide a valid basis for investigating the risks posed by dermal exposure. It has not been our intention to consider the process of uptake into the body or the derivation of dose estimates from dermal exposure, although others have developed models that could be used in such contexts.[5 6]

Conceptual model of dermal exposure
A consistent terminology has to be based on a coherent and systematic description of dermal exposure scenarios. A multicompartment

model is an appropriate basis for a terminology as it comprises distinct physical objects or compartments connected by mass transport processes. Models of this type are concerned with what happens—for example, where fingers get contaminated by touching a surface—and not why it happens—for example, particle adhesion.

COMPARTMENTS

All compartments are assumed to be well and instantaneously mixed. As a result the concentration in a compartment is described by the amount of mass and distribution volume of the
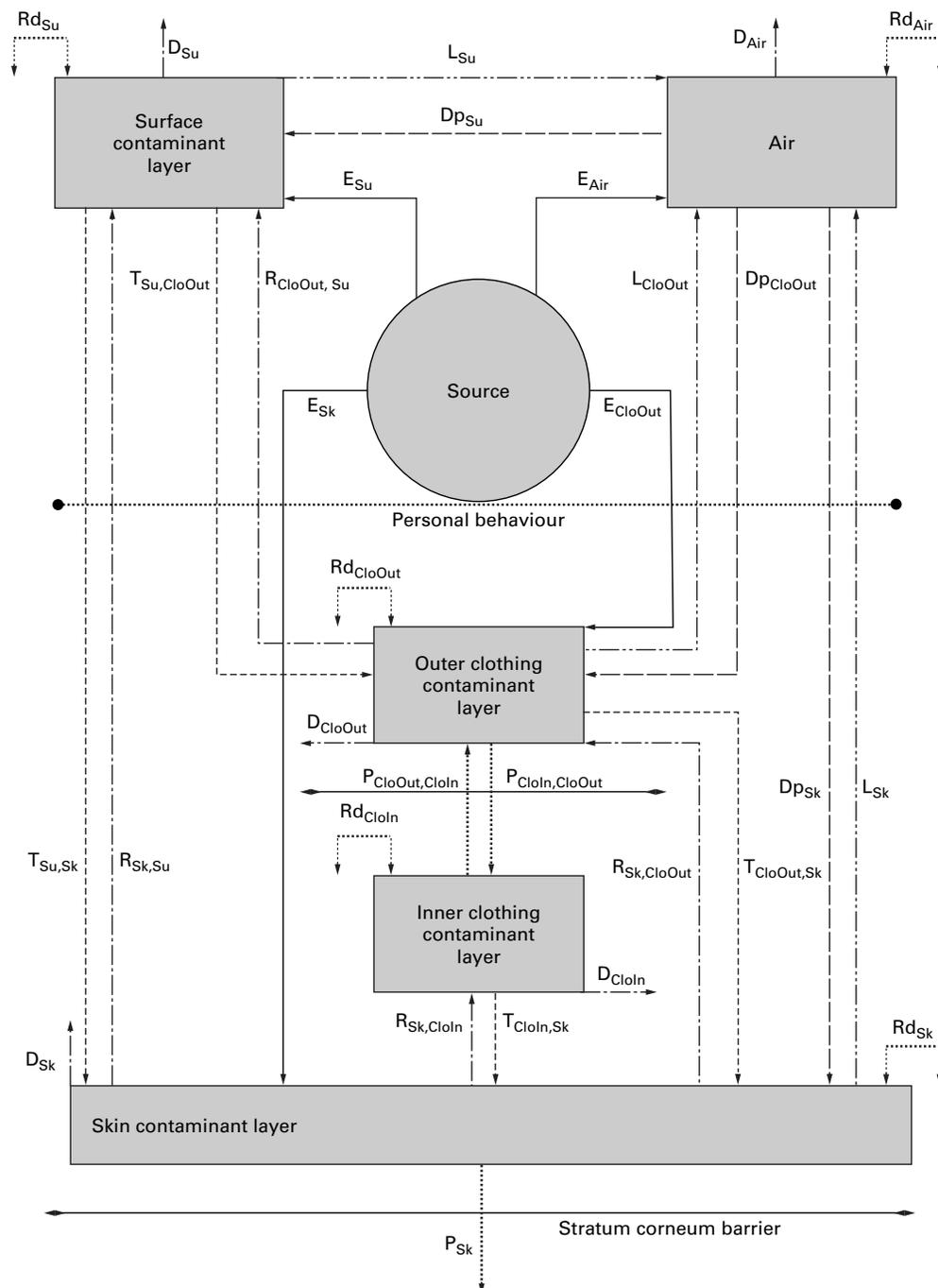
compartment. Six principle compartments are being distinguished in the model (figure):

Source (S)

Processes or activities, from which a mass is being introduced into any of the compartments, will be considered as sources.

Air

The air compartment contains vapours and dispersed particles, which are assumed to be homogeneously distributed in the compartment. The total mass of a given substance in the air compartment is well defined and can, in

Overview of the conceptual model, compartments and rate constants. E=emission (——); Dp=deposition (———);
L=resuspension or evaporation (—..—..—); T=transfer (- - -); R=removal (—·—); Rd=redistribution (·········);
D=decontamination (-·----·-); P=penetration and permeation (·········).

principle, be measured. The compartment volume is given by the size of room or other boundaries, either physical or virtual.

Surface contaminant layer (Su)

Contaminants on a surface form a layer, which delineates the compartment called the surface contaminant layer. The compartment is assumed to be homogeneous. In principle, all substances belonging to the surface contaminant layer can be identified and thus mass in this compartment can be assessed. The compartment volume is given by the three dimensional volume of this layer. For many practical purposes, a two dimensional representation of this layer will suffice.

Outer and inner clothing contaminant layer (CloOut, CloIn)

Solid or liquid contaminants at the boundary between outside and the surface of the outer clothing are modelled as the compartment called outer contaminant layer. The fabric separates this compartment from the inner clothing contaminant layer. For simplicity of the model the fabric is described as a barrier (mass transfer rate limiting) having the property to retain mass ($M_{Reservoir}$). If the mass of the hazardous substance in the outer clothing contaminant layer compartment is M_{CloOut} then some of this material will be transported through the clothing to the compartment called inner clothing contaminant layer. The mass in the inner clothing compartment is $M_{CloIn}=M_{CloOut}-M_{Reservoir}$. For many practical purposes a two dimensional representation of these compartments will suffice.

Skin contaminant layer (Sk)

On the skin, contaminants, sweat, skin oil, and barrier cream (if applied) form a layer. This layer constitutes the skin contaminant layer. The compartment is assumed to be homogeneous. In principle, all substances belonging to the skin contaminant layer can be identified and thus the mass in this compartment can be identified and measured. The compartment

volume is given by the three dimensional volume of this layer. The conventional two dimensional representation of this layer is an oversimplification, which has contributed to the confusion about the principles involved in the choice of measurement and interpretation of dermal contamination in terms of dermal uptake.

MASS TRANSPORT PROCESSES

The mass transport from the source to the compartments and sinks in the system is shown in the figure. Below the horizontal dotted line in the figure a person's movement begins to influence the transport processes. Two units are used to measure transport of mass; $g.s^{-1}$ and $g.event^{-1}$. For processes called events, it is important to describe the number of events within a reference period, typically 8 hours. The mass transport can be divided into eight distinct processes as described later and in table 1.

Emission (E)—Emission (E) is the transport of substances into the air, onto surfaces, outer clothing, and the skin contaminant layer from all primary sources. Evaporation of liquids or emission of droplets or particles into the air gives rise to emission of contaminant mass to the air. For aerosols we restrict this emission pathway to those with aerodynamic diameter <100 μm so that sedimentation is relatively unimportant. Emission to the different surfaces in the model can arise from splashing, spilling, immersion, and impaction of large particles. Splashing is the emission of large droplets the trajectory of which towards the surfaces is unaffected by air movement, whereas spilling is the event by which a liquid or powder is spilled on a surface. Immersion is an event whereby a part of the body is submerged into a liquid or a powder. Impaction is the process by which large particles are generated at the source and ejected from that source to impact onto surfaces. The emission rate is given as either $g.s^{-1}$ or as $g.event^{-1}$.

Deposition (Dp)—Deposition (Dp) is the transport of substances from the air to surfaces,

Table 1 Compartment descriptors

Compartment	Definition of metric	Symbol	Relation	Units
Source	Mass of hazardous substance available for emission	M_s		g
	Concentration of a hazardous substance in the source	C_s		$g.g^{-1}$, $g.m^{-3}$
Air	Mass of substance in the air compartment	M_{Air}		g
	Volume of the air compartment	V_{Air}		m^3
	Concentration of hazardous substance in the air	C_{Air}		g/m^3
Surface contaminant layer	Mass of a hazardous substance in the surface contaminant layer	M_{Su}		g
	Concentration of a hazardous substance on the surface	C_{Su}	$M_{Su}/(M_{Su}+M_{Other})$	$g.kg^{-1}$
	Area of surface which is contaminated with hazardous substance	A_{su}		cm^2
Outer clothing contaminant layer	Mass of hazardous substance in the outer clothing contaminant layer compartment	M_{CloOut}		g
	Concentration of a hazardous substance in the outer clothing compartment	C_{CloOut}	$M_{CloOut}/(M_{CloOut}+M_{OtherOut})$	$g.kg^{-1}$
	Area of the outer clothing which is contaminated with hazardous substance	A_{CloOut}		cm^2
Inner clothing contaminant layer	Mass of hazardous substance in the inner clothing contaminant layer compartment	M_{CloIn}		g
	Concentration of a hazardous substance in the inner clothing compartment	C_{CloIn}	$M_{CloIn}/(M_{CloIn}+M_{OtherIn})$	$g.kg^{-1}$
	Area of the inner clothing which is contaminated with hazardous substance	A_{CloIn}		cm^2
Skin contaminant layer	Mass of hazardous substance on the skin surface	M_{Sk}		g
	Concentration of hazardous substance in the skin contaminant layer	C_{Sk}	$M_{Sk}/(M_{Sk}+M_{Other})$	$g.kg^{-1}$
	Area of the skin which is contaminated with hazardous substance	A_{Sk}		cm^2

M_{Other} = mass of all other substances in a particular compartment.

outer clothing, and the skin contaminant layer. Deposition can be of mass as either solid, liquid, or vapour. The time dependent mass deposition rate $Dp(t)$, in units of $g.s^{-1}$, can be represented by the deposition velocity $v(t)$ in units of $cm.s^{-1}$ as follows:

$$Dp(t) = v(t) . C(t) . A_X \qquad (1)$$

where $C(t)$ is concentration in air outside the boundary layer and A_x is the area of the two dimensional representation of the compartment, where X is either Su for surface, Clo for clothing, or Sk for skin).

Resuspension or evaporation (L)—Resuspension or evaporation (L) is the transport of substances from surfaces, outer clothing, and the skin contaminant layer to the air, as partic (resuspension), vapours, or both. Evaporation is a continuous process driven by diffusion. Mechanical forces cause resuspension. If resuspension is caused by a single mechanical impact then the transport of mass is conveniently modelled as a transfer of mass per single event. If resuspension is caused by a sudden flow of air along a surface, there is a short initial peak of resuspended mass followed by a decay of resuspended mass per time.[7] Thus resuspension can, in general, be meaured as a transport of mass per single event.

Transfer (T)—Transfer (T) is the transport of substances by direct contact between surface, skin, and outer and inner clothing contaminant layers in a direction towards the worker. Transfer from the surface to the skin contaminant layer is event based and takes place from a small area of actual contact. That area could be considerably smaller than the surface area of the body part involved in the contact. The actual surface area of the skin that will be contaminated, as well as the efficiency of mass transfer, will depend on the actual contact—for example, single pressure, or movements of the skin along the surface.

Removal (R)—Removal (R) is the transport of substances by direct contact between skin, inner and outer clothing, and surface contaminant layer in a direction away from a worker. Removal thus is defined as an event based transport in the opposite direction of transfer.

Redistribution (Rd)— If contaminants in the air, on the surface, clothing, or skin are not homogeneously distributed, or different parts of the body such as palm, neck, and trunk need to be distinguished, the compartments can be subdivided initially into subcompartments. Redistribution then is the transport of substances from a subcompartment to another subcompartment of the same type. Redistribution of contaminants from one part of the skin contaminant layer to another can occur as a result of touching the face with contaminated fingers. Also, fabric wetting can redistribute liquid contaminants.

Decontamination (D)—Decontamination (D) is the deliberate transport of contamination from the system—for example, ventilation of room air, cleaning of room surfaces and outer clothing, or washing material off the skin. The air compartment is decontaminated by the combined effect of natural and mechanical ventilation. Cleaning of taminated surfaces, chang-

ing of clothing, and cleaning of skin all result in permanent loss of mass from the system and thus are decontamination processes. By contrast, brushing dust off clothing transports particulate mass to the room air and thus is resuspension, not decontamination.

Penetration and permeation (P)—Penetration and permeation (P) both involve transport of substances through a rate limiting barrier—such as clothing or the stratum corneum. Penetration is transport caused by external pressure, capillary penetration, and evaporation-condensation. Permeation always involves diffusion.

Transport of contaminants through permeable clothing occurs by aerosol penetration and liquid transport. External air pressure can be considered to be the driving force for penetration of aerosols through fabric,[8] whereas the mechanisms of liquid transport are capillary penetration, pressure penetration, impact penetration, and evaporation-condensation.[9] Mass transport through non-permeable clothing is a diffusion process driven by concentration.

The rate of mass transport through the stratum corneum $P_{Sk}(t)$ can be represented by a permeability coefficient $K(t)$, the concentration difference over the stratum corneum $C_{Sk}(t)$, and skin contaminant layer area $A(t)$ as:

$$P_{Sk}(t) = K(t) . C_{Sk}(t) . A(t) \qquad (2)$$

The area of the skin contaminant layer can be time dependent, which for example is the case for a drop that dries while on the skin. Roed *et al*[10] have shown that very small particles may penetrate into the stratum corneum but their fate is not known.

Concentration

The model describes transport of mass of a given substance, which is a conserved quantity provided we neglect chemical reactions. However, several transport processes are driven by the compartment concentration. For the air compartment concentration is readily obtained from mass because an air compartment can be defined so that it has a constant volume. For the surface and skin contaminant layers, volume is defined by the total amount of material present (table 2). If the skin contaminant layer compartment only contains liquids the concentration C_{Sk} of a hazardous substance of mass M_{Sk} is given by the ratio

$$C_{sk} = M_{Sk}/M_{Sk} + M_{Sk,other} \qquad (3)$$

where $M_{Sk, other}$ is the mass of all other liquid substances. This concentration can be readily transformed into the molar concentration, which is more relevant for the skin contaminant layer. As a first approximation and for substances of low volatility C_{Sk} can be considered to be the concentration in the bulk liquid.

The concept of concentration is more complicated for particles as they are discrete entities and solid substances must dissolve to diffuse through the stratum corneum. For soluble or leachable substances it is the concentration in the wet layer around the individual particle that has to be used for C_{Sk} in equation 2. This may mean that uptake will be limited by the rate of dissolution rather than diffusion through the stratum corneum. Fur-

Table 2 Mass transport process descriptions

Process	Definition	Symbol	Units
Emission	Mass of hazardous substance emitted into air from primary sources per unit time	E_{Air}	$g.s^{-1}$
	Mass of hazardous substance emitted to surface contaminant layer by splashing, spilling and ejection of particles from primary sources per unit time per event	E_{Su}	$g.s^{-1}$ $g.event^{-1}$
	Mass of hazardous substance emitted to the outer clothing contaminant layer for a particular worker by splashing, spilling and ejection of particles from primary sources per unit time per event	E_{CloOut}	$g.s^{-1}$ $g.event^{-1}$
	Mass of hazardous substance emitted to skin contaminant layer for a particular worker by splashing, spilling and ejection of particles from primary sources per unit time per event	E_{Sk}	$g.s^{-1}$ $g.event^{-1}$
Deposition	Mass of hazardous substance deposited from air compartment to the surfaces per unit time	Dp_{Su}	$g.s^{-1}$
	Mass of hazardous substance deposited from the air compartment to the outer clothing contaminant layer for a particular worker per unit time	Dp_{CloOut}	$g.s^{-1}$
	Mass of hazardous substance deposited from the air compartment to the skin contaminant layer for a particular worker per unit time	Dp_{Sk}	$g.s^{-1}$
Resuspension or evaporation	Mass of hazardous substance lost from the surface contaminant layer to the air compartment by evaporation per unit time or by resuspension or evaporation per event	L_{Su}	$g.s^{-1}$ $g.event^{-1}$
	Mass of hazardous substance lost from the outer clothing contaminant layer of a particular worker to the air compartment by evaporation per unit time or by resuspension or evaporation per event	L_{CloOut}	$g.s^{-1}$ $g.event^{-1}$
	Mass of hazardous substance lost from the skin contaminant layer of a particular worker to the air compartment by evaporation per unit time or by resuspension or evaporation per event	L_{Sk}	$g.s^{-1}$ $g\ event^{-1}$
Transfer	Mass of hazardous substance transferred from the surface contaminant layer to the skin contaminant layer for a particular worker by direct contact per event	$T_{Su,Sk}$	$g.event^{-1}$
	Mass of hazardous substance transferred from the surface contaminant layer to the outer clothing contaminant layer for a particular worker by direct contact per event	$T_{Su,CloOut}$	$g.event^{-1}$
	Mass of hazardous substance transferred from the inner clothing contaminant layer to the skin contaminant layer for a particular worker by direct contact per event	$T_{CloIn,Sk}$	$g.event^{-1}$
	Mass of hazardous substance transferred from the outer clothing contaminant layer to the skin contaminant layer for a particular worker by direct contact per event	$T_{CloOut,Sk}$	$g.event^{-1}$
Removal	Mass of hazardous substance removed from the skin contaminant layer of a particular worker and transferred to the surface contaminant layer by direct contact per event	$R_{Sk,Su}$	$g.event^{-1}$
	Mass of hazardous substance removed from the skin contaminant layer and transferred to the inner clothing contaminant layer of a particular worker by direct contact per event	$R_{Sk,CloIn}$	$g.event^{-1}$
	Mass of hazardous substance removed from the skin contaminant layer and transferred to the outer clothing contaminant layer of a particular worker by direct contact per event	$R_{Sk,CloOut}$	$g.event^{-1}$
	Mass of hazardous substance removed from the outer clothing contaminant layer of a particular worker and transferred to the surface contaminant layer by direct contact per event	$R_{CloOut,Su}$	$g.event^{-1}$
Redistribution	Mass of hazardous substance transferred from one part of air compartment to other part per event	Rd_{Air}	$g.event^{-1}$
	Mass of hazardous substance transferred from one part of contaminant layer to other part per event	Rd_{Su}	$g.event^{-1}$
	Mass of hazardous substance transferred from one part of inner clothing contaminant layer to other part per event	Rd_{CloIn}	$g.event^{-1}$
	Mass of hazardous substance transferred from one part of outer clothing contaminant layer to other part per event	Rd_{CloOut}	$g.event^{-1}$
	Mass of hazardous substance transferred from one part of skin contaminant layer to other part per event	Rd_{Sk}	$g.event^{-1}$
Decontamination	Mass of hazardous substance removed from the air compartment by ventilation per unit of time	D_{Air}	$g.s^{-1}$
	Mass of hazardous substance removed from the surface contaminant layer by deliberate decontamination per event	D_{Su}	$g.event^{-1}$
	Mass of hazardous substance removed from the outer clothing contaminant layer by deliberate decontamination per event	D_{CloOut}	$g.event^{-1}$
	Mass of hazardous substance removed from the inner clothing contaminant layer by deliberate decontamination per event	D_{CloIn}	$g.event^{-1}$
	Mass of hazardous substance removed from the skin contaminant layer by deliberate decontamination per event	D_{Sk}	$g.event^{-1}$
	Mass of hazardous substance transported through stratum corneum	P_{Sk}	$g.s^{-1}$
	Mass of hazardous substance transported from the outer clothing contaminant layer to the inner clothing contaminant layer per unit time	$P_{CloIn,CloOut}$	$g.s^{-1}$
Penetration and permeation	Mass of hazardous substance transported from the inner clothing contaminant layer to the outer clothing contaminant layer per unit time	$P_{CloOut,CloIn}$	$g.s^{-1}$

Air=air compartment; Su=surface contaminant layer; Sk=skin contaminant layer; CloOut=outer clothing contaminant layer; CloIn=inner clothing contaminant layer.

thermore, if uniform deposition takes place a monolayer of particles may be built up until the entire surface is covered. As a multilayer forms, the additional mass will have less and less influence on uptake and may be likely to dislodge and fall off. For this reason the mass of particles in the skin contaminant layer may only have limited relevance to uptake.

Measurement methods of skin and surface contamination

Depending on the perspective from which the exposure scenario is investigated a range of measurement approaches have been developed from the 1950s onwards.[12] Table 3 lists methods in common use for skin and surface contamination. Several of these methods are used to assess both compartment mass—for example, total mass in surface layer—and mass transport processes—for example, dislodgeable mass. A clear distinction is not always made and this can create confusion about determination and interpretation of sampling efficiencies. As an example, measurement methods should aim to have 100% sampling efficiency where they intend to assess compartment mass. On the other hand, a 100% sampling efficiency for a wipe test to measure the transfer to skin upon

contact with a surface is not necessarily desirable.[1] The amount that can be transported from surfaces to the skin or outer clothing contaminant compartments depends on the type of surface, the contaminant, and on the forces acting on the contaminant layer, rather than how much mass is present in the compartment. Measurement of transport must therefore be based on a model. Several routes may be followed. The transport process from surfaces could be simulated with, for example, a standardised instrumental sampling method or standardised events. Transport can also be measured during actual field conditions for an exposed population, with the result summarised in the form of a distribution of mass transport. Such reported distributions have been used for risk assessment—for example, transfer from soil giving the results as soil to skin adherence.[32]

Deposition from air is more predictable and Schneider and Stokholm[33] have proposed a theoretical model dependent on particle size for the transport of airborne dust onto the ocular surface, in relative deposition velocities. This could be called the ocular deposition fraction, by analogy with the inhalable fraction. Roed *et al*[10] have given experimental data for

Table 3 Measurement methods (for a review see Carson et al[11])

Method	Principle of sampling	Sampling area definition	Measured compartment mass*	Measured transport process†	Reference
UV Fluorescence	In situ	0.1–2 m²	M_{Sk}, M_{Su}, M_{CloOut}, M_{CloIn}	All processes	13–17
Portable x ray fluorescence monitor	In situ	Instrument defined	M_{Su}	All processes related to the surface contaminant layer	18
Wet wipe	Manual wiping	None or template	M_{Su}, M_{Sk}	$T_{Su,Sk}$, $T_{Su,CloOut}$, $R_{Sk,Su}$, $R_{Sk,CloIn}$, $R_{Sk,CloOut}$	19
Wet wipe	Mechanised wiping	Instrument defined	M_{Su}	$T_{Su,Sk}$, $T_{Su,CloOut}$	20
Gelatine foil	Surface dust lifting	1:1 transfer of dust layer	M_{Su}	$T_{Su,Sk}$, $T_{Su,CloOut}$	21
Fixed pressure dislodgeable residue sampler	Mechanical transfer in situ	10–20 cm × length sampled	M_{Su}	$T_{Su,Sk}$, $T_{Su,CloOut}$	22
Dislodgeable foliar residue sampling	Surface removal	Punches 0.1–2.5 cm², total 100 cm²	M_{Su}	$T_{Su,Sk}$, $T_{Su,CloOut}$	23
Adhesive tape	Skin stripping	1:1 transfer of dust layer	M_{Sk}	Dp_{Sk}, $R_{Sk,Su}$, $R_{Sk,CloIn}$, $R_{Sk,CloOut}$	24
Hand wash	Wash with water or alcohol	Total hand surface	M_{Sk}	$R_{Sk,Su}$, $R_{Sk,CloIn}$, $R_{Sk,CloOut}$, D_{Sk}	25
Patch	Passive	Sampler defined	M_{Sk}	$T_{Su,Sk}$, $T_{CloIn,Sk}$, $T_{CloOut,Sk}$, Dp_{Sk}	26
Whole body	Passive	Body parts	M_{Sk}	$T_{Su,Sk}$, $T_{CloIn,Sk}$, $T_{CloOut,Sk}$, Dp_{Sk}	26
SMAIR	Resuspension by air jet	Instrument defined	—	L_{Su}	27
STEPP	Resuspension by impact	Instrument defined	—	L_{Su}	28
Microvacuuming	Resuspension by suction	Instrument defined	M_{Su}	L_{Su}	29
FLEC	Evaporation by airflow	Instrument defined	—	L_{Su}	30
TEWL	Evaporation	Instrument defined	—	L_{Sk}	31

*Definition of symbols as for table 1.
†Mass transport process descriptions as for table 2.

mean deposition velocities on arms and other body parts for people seated in a test room, which likewise could form a basis for defining other dermal deposition fractions.

There is a close analogy to requirements for sampling inhalable airborne particles. Historically, fractions with biologically relevant particle size have been based on experimental data that reflect mean deposition efficiencies in the airways. An alternative approach has been to define the fraction as that obtained by a given sampling instrument. Initially there were different proposals for the size fractions, and samplers were not available which could measure according to these criteria. However, better experimental data on deposition efficiencies has resulted in the adoption of an international standard for the definition of the respirable, thoracic, and inhalable fractions.[34] Recent development has resulted in samplers, which have aspiration efficiencies that match the definition of the fractions.[35]

In the following section some strategies for measuring with the model compartment mass and mass transport processes will be discussed, highlighting several problems in conventional approaches to measurement and possible routes forward.

COMPARTMENTS

The mass in a compartment at a given time can in principle be measured by sampling all of the contaminant present at that time in the compartment. A strategy often used in monitoring is to estimate the entire compartment mass by sampling a small proportion of the whole.[26 36]

Air

Methods for measurement of concentration of hazardous substances in the air compartment are well established relative to estimation of inhalation risk. For aerosols specifications are available for sampling fractions of biologically relevant inhalable size,[34] but methods for measuring the concentration of non-inhalable particles, which are relevant for estimating surface deposition, have not yet been developed.

Surface contaminant layer

For surfaces, several in situ methods are available, but they can only measure a limited range of substances, and never total mass. Portable x ray fluorescence analysis is one example of this type of analysis.[18] Methods based on removal— such as adhesive tape sampling—can have a high sampling efficiency. The efficiency in field use is usually estimated with consecutive sampling of the same area. However, this is based on a circular argument, as only the contaminant that the tape can actually remove will ever be sampled.[20]

Clothing contaminant layers

In the model clothing was divided into an outer and inner contaminant layer, with a buffer to represent the mass residing inside the clothing which does not come into contact with surfaces or skin, respectively. Measurement of the entire mass in the clothing would in principle be no problem and if this were done for non-permeable fabrics theoretically the outer contaminant layer would be measured. To our knowledge a method which measures the inner contaminant layer for permeable layers is not available.

Skin contaminant layer

The skin rinse method[25] has been used to assess the mass in the skin contaminant layer compartment. The method typically recovers 40%–90% of contaminants spiked onto the skin. Recoveries can be measured experimentally. However, spiking experiments have inherent problems as it is unclear whether this result should be interpreted as a 40%–90% sampling efficiency for the skin contaminant layer or if it reflects partitioning between this compartment and the stratum corneum or perfused tissue.

Measurement methods based on UV fluorescence of tracer compounds mixed with the contaminant at source[13–17] indirectly measure the mass of the contaminant in the skin contaminant layer. However, fluorescent tracers have the ability to bind with the cell proteins in the stratum corneum and the method is not able to dif-

ferentiate between the skin contaminant layer and the stratum corneum.

MASS TRANSPORT PROCESSES

Transport can in principle be measured with a mass balance calculation for the change in mass in all relevant compartments per event or per time. Transport can more directly be determined by methods which standardise a set of external factors that are considered to be important determinants of the transport process (table 3).

Deposition

Adhesive tape can measure the particle deposition unaffected by loss. This has been done for particle deposition onto the face,[37] but the method does not work for parts of the body that get into contact with other surfaces. Charcoal cloth has been used to measure skin deposition of volatile compounds. A problem is that the method does not differentiate between vapours from air and liquid splashes.[38 39]

Resuspension or evaporation

Resuspension is usually best described as a series of single events. If events are repeated, say at frequency γ, and the ratio γ/T tends to infinity, where T is a suitable reference time interval, resuspension can be treated as a continuous process. In this case it is customary to define a resuspension rate R by the equation where the mass of substance lost from the surface per unit time (L) (table 2) is:

$$L(g\ s^{-1}) = R(s^{-1})\ .\ M_{Sk}(g) \qquad (4)$$

This definition assumes that R is independent of M_{Sk} (see also discussion for transfer).

An approach to assess resuspension based on events would be to collect all dust resuspended as the result of a standardised mechanical impact. Kildesø *et al*[28] have developed the surface total emission potential of particles (STEPP) tester, which simulates resuspension caused by a walking person from a carpet. Resuspension caused by air movements is simulated in the SMAIR tester.[27] In this instrument a well defined air jet is directed at a surface contaminant layer and the resuspended particles collected on a filter. Vacuuming techniques could be used to simulate resuspension caused by strong air velocities, provided the nozzle does not touch the surface.

Evaporation is driven by diffusion and thus it is simpler to specify external factors. Wolkoff[30] has developed a field and laboratory emission cell (FLEC), which in principle is a small air compartment that is placed on a surface. A well defined flow of conditioned air is passed through the compartment and the exhaust air is analysed for evaporated substances. The diffusive transport of water vapour from the skin (transepidermal water loss) can be measured simply with instruments.[31] A similar principle could be used for other vapours lost from the skin.

Transfer

The increase in mass over time in the skin contaminant layer is the measured net balance between transport to and from the compartment and thus is affected by deposition, resuspension or evaporation, removal, and uptake. Surrogate skin methods such as pads, gloves, and coveralls are based on the assumption that they are estimates of the transport of mass to the skin contaminant layer from surfaces. However, the materials used do not have the roughness, stickiness, and other properties of human skin and so may not meet this assumption.

Dislodgeable foliar residue, a procedure first described by Iwata *et al*,[23] determines the pesticide remaining on foliage after spraying. It involves destructive sampling of foliage by punching or removing leaves to provide a sample leaf area of about 100 cm^2 and subsequent, partly mechanical partly chemical, removal of pesticide residue by shaking the leaves in water with some drops of surfactant. This process is intended to mimic transfer to skin. Then pesticide is extracted from the water and analysed chemically. Sometimes an organic solvent is used in the first step, but this then measures the mass in the compartment rather than the dislodgeable foliar residue. Both methods, however, have been used to indicate the strength to transfer by direct contact from the source, which may be relevant to predict exposure by contact. A more general form of dislodgeable foliar residue is transferable residue (TR). If TR is measured simultaneously with an increase per event of mass per area M_{Sk}/A_{Sk} in the skin contamination layer, a dermal transfer coefficient (DTC) can be defined as[1 40]:

$$\text{DTC (cm}^2.\ \text{event}^{-1}) = M_{Sk}(g.\text{event}^{-1})/A_{Sk}\text{TR}(g.\text{cm}^{-2}) \qquad (5)$$

For this equation to be useful it has to be assumed that TR is independent of M_{Sk}/A_{Sk}. However, Brouwer *et al*[41] have shown that TR depends non-linearly on the mass per area in the surface contaminant layer, M_{Su}/A_{Su}. A better approximation would be to assume a functional relation:

$$\text{TR}=\text{TR}(M_{Su}/A_{Su}) \qquad (6)$$

determined by non-linear regression. In reality a stochastic relation must be expected, and thus for given M_{Su}/A_{Su}, TR has a given distribution with variables (mean and variance) being functions of M_{Su}/A_{Su}. It is to be expected that liquid contaminants can be assessed with the simple relation, grease or other paste-like contaminants an intermediate relation, and particulate contaminants with the most complex relation. Transfer of particles will depend on contaminant properties—such as particle size, distribution, shape, and humidity or cohesion. In all cases it must be remembered that TR will depend on the method used for its measurement.

There are several versions of samplers designed to exert a constant force on a defined area in a wiping action. These methods have potential for simulating the transfer of mass and are non-destructive. One such monitor is a fixed pressure dislodgable residue sampler—such as a polyurethane foam roller.[22] Ross *et al*[42] used standardised movements (aerobic dance routines following an instructor and music) to measure transfer of pesticides from fogged carpets onto the skin.

Transfer from the inner clothing contaminant layer to the skin contaminant layer is

usually estimated with patches or underwear. No other method seems to have been developed specifically for measuring this transfer.

Removal
Dissipation of contaminants from the clothing compartment has been the subject of many studies, however, the studies were focused on the transport of mass from the clothing compartment to the skin compartment—that is, the opposite direction to removal. Yang and Li[43] reported a frictional transport of pesticides of 1%–5% from contaminated clothing to underwear. As mechanical or frictional transport may be an important mechanism of removal, these data indicate the range of mass transport from clothing in the opposite direction. Removal from skin to surface has been studied by Brouwer *et al*.[41] They reported a mean removal efficiency of 38% during a single pressure contact of a contaminated hand with an uncontaminated flat surface.

Redistribution
Information on the area exposed and the redistribution of contaminant could be obtained by observation. However, the method of choice is the fluorescent tracer technique with a quantitative image analysis system.[13–17] These methods have high spatial resolution.

Decontamination
Decontamination of the air compartment is the removal of mass from the air compartment by either local or general ventilation. Methods to assess the efficiency of ventilation in reducing the concentration of gaseous and particulate contaminants are well developed.[44] Decontamination of surfaces can be assessed by measuring the mass removed—for example, the quantity of dust in a vacuum cleaner filter bag—or by the decrease in mass of the surface contaminant layer. A measurement strategy for decontamination of surface dust in offices has been described by Schneider *et al*[21] and a standard procedure to assess the efficiency of handwashing has been developed by Fenske and Lu.[45]

Penetration and permeation
Penetration and permeation stand for transport of mass through either the clothing barrier or the stratum corneum. These mass transport pathways have been studied in great detail relative to the effectiveness of protective clothing[46] and percutaneous penetration.[47] In both cases a test cell design is used to measure permeation. However, several different methods are used in the assessment of percutaneuos penetration ranging from in vitro models with static diffusion cells and flow through diffusion cells with animal or human skin (viable and non-viable).

General discussion and conclusions
In this paper we propose a conceptual model of both the important pathways leading to dermal exposure and the intermediate compartments where the hazardous substances may reside. This model has allowed us to define consistent terminology, which should form the basis for improved comparability between studies of dermal exposure or surface contamination. Consistent use of the model will ensure that most appropriate variables are measured in any situation. The model has been constructed at a conceptual level and omits much of the detail, which is evident in real situations. It is possible to extend the model as a series of interlinked compartments for the skin, surfaces, etc. In this way compartments for hands, arms, torso, etc could represent the skin surface; or if greater resolution is required, fingers, palms, etc could represent the skin surface. In this way the existing model encompasses sufficient detail to enable terminology to be developed, and it could be generalised for any particular need.

The model can cope with special exposure scenarios—such as immersion of body parts in liquid or powder. In this case the liquid or powder constitutes the skin contaminant layer compartment and has infinite volume.

For interpretation of contaminated substances derived from soil and paste-like substances in the skin contaminant layer a refinement of the model is necessary. The skin contaminant layer could be subdivided into an outer layer and an inner layer in intimate contact with the stratum corneum. The possibility exists that it is the supply of hazardous substance from the outer layer to the inner layer, which limits the rate of uptake. This identifies a need to refine existing methods of measuring mass in the skin contamination layer compartment.

We have not considered the stratum corneum in much detail, as it is generally not available for surface sampling methods. Skin stripping with adhesive tape is a possible exception. Consecutive stripping allows semiquantitative depth profiling, but if the skin contaminant layer is not empty, the mass in this compartment and in the stratum corneum will be partially mixed up. The finite thickness of the stratum corneum constitutes a buffer capacity and introduces a lag time for the transport process to reach equilibrium after a step change in concentration.[24] The lag time may vary from a few minutes up to days and its significance for risk assessment would be determined by duration of exposure compared with lag time.

The inventory of existing methods for measurement of skin and surface contamination (table 3) is only illustrative. Compilation of an exhaustive inventory and discussion relative to the model is beyond the scope of the present paper. However, with the model structure, limited or non-existing methods for measuring relevant compartment mass or transport processes have been identified:

● The model stresses the importance of measuring concentration in the skin contaminant layer but methods relevant for uptake are lacking

● The mass of particles in the skin contaminant layer may only have limited relevance for uptake. Particles are less likely to result in uptake than liquids and solubility of particles is an important qualifier which should be

specified along with results of measurements of the mass and composition of the skin contaminant layer

● A clear distinction must be made between mass in a compartment and transport of mass. Direct measurement of mass transport must be based on an appropriate theoretical model.

We envision that our paper will stimulate discussions and help in the development of more appropriate methods for the assessment and control of dermal exposure.

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