

Original Article

Prediction of Dermal Exposure to Chemical Substances Using a Fluorescence Method within the SysDEA Project

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Abstract

Dermal exposure is an important exposure route for occupational exposure and risk assessment. A fluorescence method has been developed to quantify occupational dermal exposure based on a visualization technique, using Tinopal SWN as a fluorescent tracer. The method was developed within the framework of a large experimental study, the SysDEA project. In SysDEA, dermal exposure was measured with different methods for 10 simulated exposure situations by sampling powder and liquid formulations containing Tinopal SWN on coveralls and patches and subsequently chemically analysing them. For the fluorescence method, photographs of exposed volunteers who performed the experiments were taken inside a room which consisted of an optimized arrangement of several UV irradiating tube light brackets, reflective and non-reflective backgrounds for maximum light diffusion and a camera. Image processing analysis software processed these photographs to obtain corresponding light intensity in terms of summed pixel values. To be able to estimate the amount of Tinopal SWN, 25% of the measured data from the SysDEA experiments were used to calibrate by correlating the summed pixel values from the photographs to actual measured exposure values using a second order regression model. For spraying both high and low viscosity liquids, showing uniformly distributed exposure patterns, strong Pearson correlation coefficients ($R > 0.77$) were observed. In contrast, the correlations were either inconsistently poor ($R = -0.17$ to 0.28 for pouring, rolling high viscosity liquid, manually

What's important about this paper?

Dermal exposure is an important exposure route for many chemicals in diverse occupations, but there is a critical need for precise methods to determine occupational dermal exposure. Fluorescent tracers are one method by which to directly measure dermal exposure. This study evaluated the relation between light intensity obtained from a fluorescent tracer photographed under UV light with exposure to that tracer which was chemically analysed. The method is a promising tool to qualitatively or semi-quantitatively assess dermal exposure without the need of chemical analysis of the compound.

handling objects immersed in low viscosity liquid and handling objects contaminated with powder), moderate ($R = 0.73$ for dumping of powder), or strong ($R = 0.83$ and 0.77 for rolling low viscosity liquid and manually handling objects immersed in high viscosity liquid). A model for spraying was developed and calibrated using 25% of the available experimental data for spraying and validated using the remaining 75%. Under given experimental conditions, the fluorescence method shows promising results and can be used for the quantification of dermal exposure for different body parts (excluding hands) for spraying-like scenarios that have a more uniform exposure pattern, but more research is needed for exposure scenarios with less uniform exposure patterns. For the estimation of exposure levels, the surface loading limit should be lower than $1.5 \mu\text{g}/\text{cm}^2$ (a lower limit could not be quantified based on experiments conducted in this study) on a large surface, like a coverall, which should be ideally perpendicular to the camera.

Keywords: dermal exposure assessment; experiments; fluorescence; workplace exposure

Introduction

Dermal exposure can be defined as the process of contact between a chemical or biological entity (or an agent) and human skin (ISO, 2011). The relevance of dermal exposure to various chemicals is well recognized as it occurs in a wide variety of occupations, which span over both agriculture and manufacturing sectors (Schneider *et al.*, 2000; Semple, 2004). The workers in these sectors regularly encounter contamination of the skin by transport processes such as emission, transfer, or deposition (Schneider *et al.*, 2000) and/or cause splashes, spilling, dripping, and aerosols of various products (Behroozy, 2013), making dermal exposure a relevant exposure route.

To assess the dermal exposure in such occupational settings, there are three methods, which are generally employed. Interception methods theoretically capture all contaminant *en route* to the body (potential dermal exposure) that could occur without any exposure reducing methods being applied (WHO, 2014) by means of sampling with, for instance, coveralls, clothing, patches, or gloves. Removal methods estimate the amount of contaminant that has reached the skin by means of, for instance, wiping, tape stripping, washing, or rinsing, either directly or by penetration or permeation through the (protective) clothing, and has not been redistributed,

absorbed by the skin or removed otherwise. The mass in direct contact with the (bare) skin that is available for absorption is also called actual dermal exposure (OECD, 1997).

In situ methods involve direct assessment of the agent or a fluorescent tracer, for example, by image acquisition and processing systems, without the need of sampling and chemical analysis. Fluorescent techniques exploit the visual properties of surrogate fluorescent compounds (tracers) that are, for instance, added to a product and made visible by means of UV light. When combined with imaging, they make quantification of dermal exposure patterns possible (Fenske *et al.*, 1986; Fenske, 1988; Fenske, 1993). Fluorescent techniques have already been employed as a qualitative method for dermal exposure assessment (Franklin *et al.*, 1981; Fenske *et al.*, 1986; Roff, 1994; Bierman *et al.*, 1998) as well as for educational purposes (Foss *et al.*, 2002).

As there is a principal need for more precise methodology regarding the determination of occupational dermal exposure, a project called 'Systematic analysis of Dermal Exposure to hazardous chemical Agents at the workplace (SysDEA)' was launched with an overall aim to generate scientific knowledge to improve and standardize measurement methods for dermal exposure to chemicals at the workplace (Kasiotis *et al.*, 2020). As

part of SysDEA, a fluorescence method was developed in this study, which aimed to quantify the amount of potential dermal exposure by measuring fluorescence. The identified capabilities and limitations of the method provide further insight in its applicability domain. The results from the chemical analysis of samples which were collected in [Kasiotis et al. \(2020\)](#) were used to calibrate and validate the estimates of the fluorescence method.

Method

Approach fluorescence method

The fluorescence method consists of

- (i) UV room in which volunteers exposed to a fluorescent tracer (Tinopal SWN; CAS 91-44-1; 7-diethylamino-4-methylcoumarin) are photographed, and
- (ii) image processing software, that processes and analyses the photographs to estimate exposure based on the obtained light intensity of the tracer.

UV room set-up

For this study, the volunteers were photographed inside a UV room (before and after each experiment) in which six UV rays irradiating double tube light brackets were placed on a construction of aluminum pipes (see [Fig. 1a](#)). The brackets could be freely moved. The optimal set-up with regard to light diffusion was determined by varying with the number and movement of brackets, placement of a reflective background on the

floor or ceiling, distance from the brackets to the volunteer and height of the camera. All brackets were placed in a semi-circle, facing the spot where volunteers stood in front of a non-reflective black background screen. A reflective surface was laid on the floor over which the volunteer stood to increase the light diffusion on the lower parts of the body. A camera (either Canon EOS 700D for determining the set-up, and photographing of contaminated plates resulting from additional experiments or a Nikon D90 for photographing each volunteer after each measurement) was placed on a tripod which was set at 95 cm height. The degree of light diffusion of the set-up was tested using a cross of white paper patches attached on the background screen reflecting the height of an average person in Greece (~175 cm) ([Fig. 1b](#)). The light intensity for the set-up, as shown in [Fig. 1b](#), was found to be evenly distributed (0.94–1) for the middle and low lying patches, but lower (0.64–0.82) for the high lying patches and was used to optimize the set-up of the UV room.

Software development

When a photograph of a volunteer was taken, the camera produced two files (the photograph itself and a RAW image file containing more information). The image processing software reads the RAW photograph with an open source tool (DCRaw) and converts the camera output into a ppm-file, thereby reducing the influence of picture processing from the camera itself. The ppm file is a large data matrix in which each value

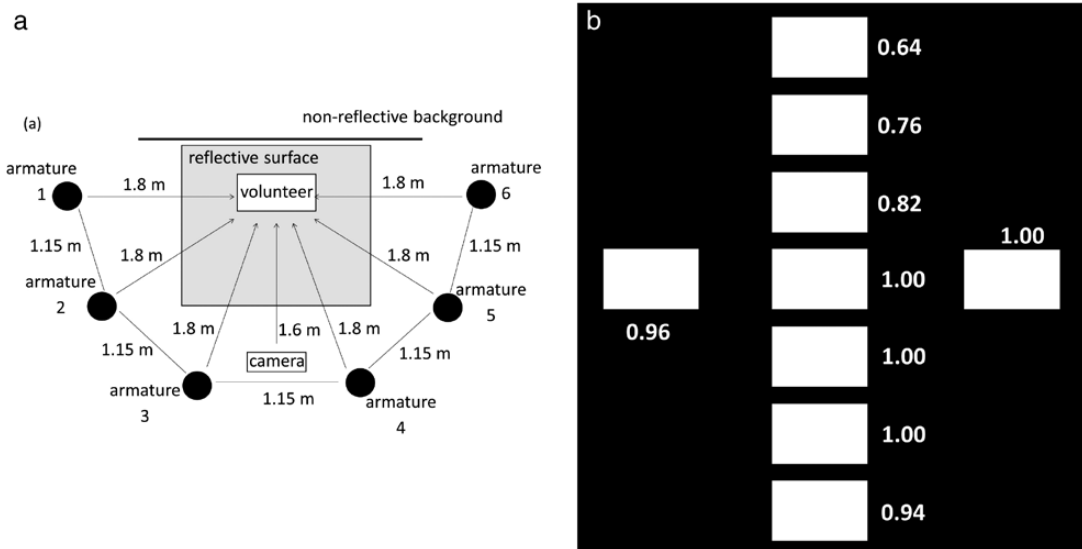


Figure 1. (a) Schematic representation of the set-up of the UV lights and brackets inside the UV room. (b) Diffusion of the light on the white paper patches attached on the background screen.

represents one optical sensor of the camera. Once the camera output is converted to a ppm file, an 'annotation tool' is used to indicate the location of 13 body parts (i.e. head, upper left arm, lower left arm, upper right arm, lower right arm, front torso, back torso, left hand, right hand, upper left leg, lower left leg, upper right leg, and lower right leg) on the photograph, which is currently done manually by the assessor. Contextual information such as experiment ID, volunteer number, and exposure situation, can also be added for each body part with the annotation tool. This information is then used as input for the second part of the software program that obtains the light intensity. User interface of the annotation tool is shown in Supplementary information (Fig. S1 [available at *Annals of Occupational Hygiene* online]).

By default, the camera was set on ISO 3200, F/8. For each volunteer, three consecutive photos were taken with a shutter time of 1/80th, 1/20th, and 1/5th second. Photographs with a shutter time of 1/20th second were processed by the software first. However, if this does not yield a satisfactory photograph, a different shutter time (i.e. 1/80th or 1/5th second) can be used, depending on whether the light intensity in the photograph is too low or too high to analyse. This ensures that photographs are not overexposed for quantification. In this case, a correction for the default shutter time of 1/20th second is applied. Most consumer brand cameras use a Bayer filter on the light sensitive chip that has three color filters arranged in a grid pattern, which relates to the RGB (red, green, and blue) channels. Since Tinopal SWN emits at 430–436 nm, which falls within the blue range, the software only uses the information from the blue channel. The blue channel on consumer brand cameras is sensitive for blue light of a much broader range, resulting in some background noise. To correct for background noise, the pixel value, as estimated for the background, is subtracted from all other annotations. After the blue channel image is corrected for camera settings and background, the tool automatically sums all pixel values for each of the 13 annotated body parts. This summed pixel value is the light intensity as emitted by Tinopal SWN for a given body part.

Determination of the applicability domain

Experiments were conducted to investigate the applicability domain of the fluorescence method by

determining its quantification limit and angular dependency relative to the loaded surface.

Quantification limit

To determine the quantification limit of the fluorescence method, three experiments were conducted. For the first quantification experiment, low viscosity liquid formulations with varying concentrations of Tinopal SWN were prepared (see Table 1), of which 15 µl drops were deposited on spots of pre-determined size of both 5.3 and 2.3 cm² on high-pressure laminate (HPL) plates. HPL plates consist of a non-fluorescent material that does not absorb any liquid. The spot size was determined by circling a bottle cap and measuring the diameter. The formulation was spread out using a needle to make sure that the total area of the spot was, as much as possible, evenly loaded. Three spots with same concentration were applied in a row. Thus, nine rows with nine different concentrations were obtained. The HPL plate was placed in a upright position at chest height in the same position as a person would stand and was photographed under UV light using the same experimental set-up as shown in Fig. 2a directly after applying the Tinopal SWN (without drying). The photograph was subsequently annotated (Fig. 2b), after which the light intensity of each spot was obtained by the software.

For the second quantification experiment, several patches of Tyvek material (SprayGuard, Indutex SA), which is the same material as used during the experiments described later, were cut in sizes of 30 × 30 cm². Either the high or the low viscosity formulation of Tinopal SWN (2 g/l concentration) was sprayed using a nebulizer (creating a fine mist) on a patch once, twice, four times, seven times, or 10 times, resulting in a total of 15 patches for each type of liquid. A schematic is shown in Supplementary data Fig. S2 (available at *Annals of Occupational Hygiene* online). The Tyvek patches were located at chest height in the same position as a person would stand and were individually photographed under UV light using the same experimental set-up as shown in Fig. 1a. The photograph was subsequently annotated, after which the light intensity of each patch was determined by the software. The patches were then chemically analysed (Kasiotis *et al.*, 2020) to determine the amount of Tinopal SWN on each patch.

Table 1. Concentrations of Tinopal SWN per 20 ml of liquid formulation.

Concentration of Tinopal SWN (g/L)	4	2	1	0.5	0.25	0.125	0.0625	0.0312	0.0156
Mass of Tinopal SWN added (g)	0.08	0.04	0.02	0.01	0.005	0.0025	0.00125	0.000625	0.000312

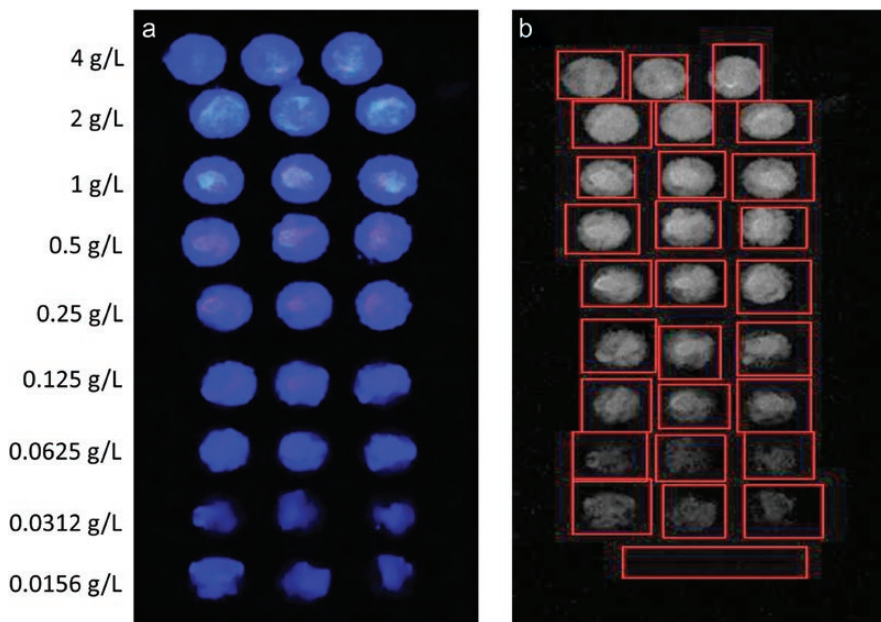


Figure 2. (a) Photograph of UV illuminated dried spots of Tinopal SWN formulations with varying concentrations and same surface area of 5.3 cm² on HPL plate. (b) Associated annotations in the software.

In the third quantification experiment, 40 petri dishes (surface area of 10.7 cm² each) were covered with cotton layer (from cotton coveralls) and weighed on an analytical balance. They were loaded with varying amounts of grinded Tinopal SWN powder with a sieve size of 50 µm and were weighed again to determine the amount of Tinopal SWN on each petri dish. All samples were located at chest height in the same position as a person would stand and were photographed under UV light using the same experimental set-up as shown in Fig. 1a. The photograph was subsequently annotated, after which the light intensity of each sample was determined by the software.

Angular dependency

To avoid any loss in the fluorescence intensity, a loaded surface should ideally be perpendicular to the incident ray from the camera. However, in reality, the surface to observe can be at an angle other than 90°, for example, due to the non-planar form of the body and wrinkles/creases on the coverall worn by a volunteer and. This may lead to reduction in the registered intensity. To investigate this effect, spots of 10 µl of Tinopal SWN liquid formulation with 0.25 g/l concentration (=2.5 µg of Tinopal SWN per spot) were applied on the Tyvek surface and pasted on a flat curved surface of a bucket (schematic is shown in Supplementary data Fig. S3

[available at *Annals of Occupational Hygiene* online]). The horizontal distance between each spot in a row was calculated so that each spot would be under an angle that was 10° more relative to the camera as the previous spot. Photos were taken under similar circumstances as the previous described experiments. The same procedure was repeated for the other two rows. Subsequently, the light intensity of each spot was determined by the software.

Volunteer recruitment and safety procedures

Medical ethical approval was obtained for the study by the Ethical Committee of BPI. Volunteers were recruited to participate in the SysDEA study based on experience of previous participation in field trials, physical capabilities, and willingness to perform the tasks in the study. An occupational physician and a safety supervisor officer were present during recruitment of the volunteers and the experiments. Personal protective measures in the form of UV-goggles were provided to all personnel in the UV room when photographs were taken, and the room was only operative when photographs were taken to keep UV exposure to a minimum. More details with regards to recruitment and training of the volunteers is described in Kasiotis *et al.* (2020).

Calibration and validation of the fluorescence method

As mentioned earlier, the present fluorescence method is a part of the SysDEA project, in which well-designed and reproducible real life exposure simulating experiments were performed (Kasiotis *et al.*, 2020). Ten different dermal exposure situations were studied, which were related to six tasks (pouring low and high viscosity liquids, rolling low and high viscosity liquids, spraying low and high viscosity liquids, manually handling objects immersed in low and high viscosity liquid, dumping powder and handling objects contaminated with powder). Tinopal SWN was used as a test substance, either in liquid formulations with low or high viscosity or pure powder. A total of 320 experiments were performed (10 exposure situations $\times$ 2 combinations of dermal exposure measurement methods $\times$ 4 volunteers $\times$ 4 repetitions per volunteer) using whole body dosimetry (coverall) and patches for body exposure measurement, gloves and hand wash for hand exposure measurement and headband and head wipe for head exposure measurement. For the body exposure measurements, 10 patches have been mounted on different body sections (upper and lower legs, upper arms and forearms, front and back of the torso), or for the whole body dosimetry the coveralls were divided into 10 segments representing these body sections. In addition, before and after each experiment, three photographs (each with a different shutter time as described earlier) of the volunteer were taken under UV light each from the front and from the back of the volunteers using the same set-up as shown in Fig. 1a. The 4000 collected samples were chemically analyzed to determine the amount of Tinopal SWN (mass in μg on each dosimeter). The entire experimental design as well as the description of the chemical analysis of the samples can be found in Kasiotis *et al.* (2020).

To be able to quantify the amount of dermal exposure based on the obtained light intensity, expressed as summed pixel values per body part, the amounts of Tinopal SWN from chemical analysis were used to calibrate the output of the fluorescence method using a second order regression model. However, for the hands, no differentiation in light intensity could be obtained by the software. The measured amounts of Tinopal SWN on the hands of the volunteers were high (Kasiotis *et al.*, 2020) and led to a saturation of the light intensity from Tinopal SWN on the hands. Therefore, the calibration was limited to body exposure (i.e. excluding the hands).

The exposure data for all body parts (excluding hands) was combined for each exposure situation (i.e.

pouring, rolling, spraying LV, HV liquids, etc.) to obtain one dataset per exposure situation. For each exposure situation, 25% of the data was randomly selected using the '=RAND()' formula and 'SORT' function in MS Excel. For each exposure situation, the correlation between the amount of Tinopal SWN (as measured by chemical analysis) and the summed pixel values (as obtained by the tool) was determined through Pearson coefficient of correlation (R) based on the selected 25% of the data. Based on the relatively high correlation coefficients for both LV and HV liquids and more uniform exposure patterns, spraying was picked as an example for which second order fit regression model(s) were derived. This model quantifies the amount of Tinopal SWN on the basis of obtained light intensity in case of spraying. The fit model was validated using the remaining 75% of spraying relevant dataset. It should be noted that more uniform exposure patterns, in the case of spraying, allowed the use of a larger pool of data. Pooling of all data with high(er) correlation coefficients was not considered appropriate for other exposure situations due to the differences in exposure patterns and differences in fluorescence between the liquid and solid formulations.

Results

Applicability domain

Quantification limit

When applied as spots (quantification experiment 1), the concentrations of Tinopal SWN in the formulation are compared to the corresponding summed pixel values in Fig. 3. The lowest concentration that was used in this setup was 0.015 g/L, that is, the lowest possible detectable concentration within the tested experimental conditions. For the spots of 5.3 cm^2 , the summed pixel values increased linearly with the concentration of Tinopal SWN in the formulation up to 0.5 g/L, which is equal to 7.5 μg of Tinopal SWN ($= 0.5 \text{ g/L} \times 15 \mu\text{L}$) and a surface loading limit of 1.4 $\mu\text{g}/\text{cm}^2$ ($= 7.5 \mu\text{g}/5.3 \text{ cm}^2$). For the spots of 2.3 cm^2 , the linear increase in the summed pixel values is up to a surface loading limit of 1.6 $\mu\text{g}/\text{cm}^2$ ($= 0.25 \text{ g/L} \times 15 \mu\text{L} / 2.3 \text{ cm}^2$) which is almost comparable to the previous surface loading limit for 5.3 cm^2 spot size and was also observed to remain almost constant (around 1.5 $\mu\text{g}/\text{cm}^2$) even with further decrease in spot size (not shown here). Beyond this upper detection limit of 1.5 $\mu\text{g}/\text{cm}^2$, the detection capacity of the method starts to saturate and obtained summed pixel values lead to inaccurate derivation of amount of Tinopal SWN.

When sprayed (quantification experiment 2), the amounts of Tinopal SWN and the summed pixel values are higher for high viscosity liquid compared to low viscosity liquid (Fig. 4a). The spray samples have an almost uniformly distributed Tinopal SWN. Contrary to quantification experiment 1, the summed pixel values increase up to the maximum extent of 490 μg of Tinopal SWN which corresponds to a surface loading of 0.54 $\mu\text{g}/\text{cm}^2$ ($= 490 \mu\text{g}/900 \text{ cm}^2$) without any saturation or upper detection limit. However, this value is below the upper level found in quantification experiment 1.

In case of the experiments with Tinopal SWN powder, instead of an upper detection limit, a lower detection limit of around 2 mg Tinopal SWN per 10.7 cm^2

(corresponding to a surface loading of 0.18 mg/cm^2) was observed (see Fig. 4b). In other words, up to a surface loading of 0.18 mg/cm^2 , the light intensity of the powder is almost negligible. Moreover, the light intensity emitted by Tinopal SWN in powder form is considerably lower than that of Tinopal SWN in liquid formulations as considerably higher amounts of Tinopal SWN powder (in mg) result in considerably lower summed pixel values. For instance, for the tested maximum amount of 10.5 mg of Tinopal SWN powder (resulting in a surface loading of $\sim 1 \text{ mg}/\text{cm}^2$), a summed pixel value of 4×10^7 is obtained. This is even lower than the minimum summed pixel value of 1×10^9 as obtained for spraying 24.5 μg of Tinopal SWN as part

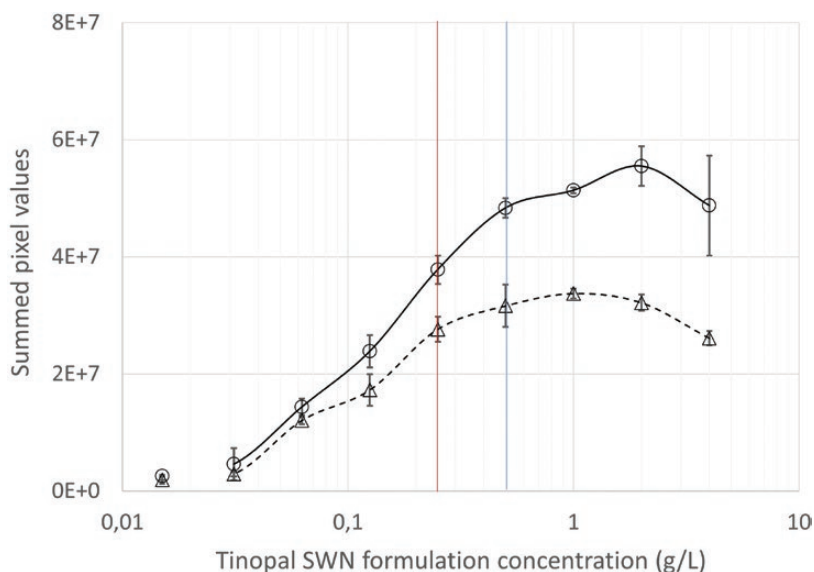


Figure 3. Variation of summed pixel values of the applied spots of low viscosity liquid with increasing concentrations of Tinopal SWN for two different spot sizes; the concentration levels of 0.25 and 0.5 g/l are highlighted in red and blue vertical lines.

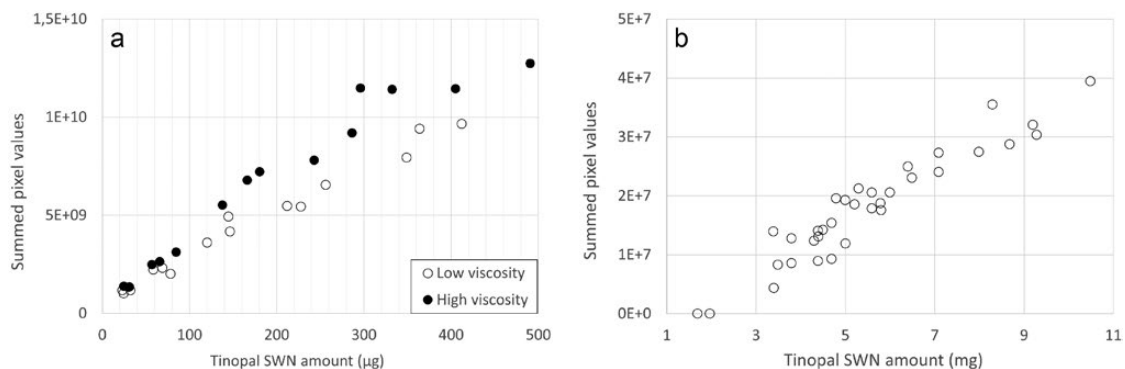


Figure 4. Variation of summed pixel values with an increase in the amount of Tinopal SWN on (a) Tyvek patches after spraying low and viscosity liquid formulations. (b) Petri dishes loaded with grinden Tinopal SWN powder.

of a low viscosity liquid, resulting in a much lower surface loading of 0.023 µg/cm².

Angular dependency

A small 10° change in the relative angle (from 90°) was observed to produce an average reduction of 4% in the summed pixel values. When the relative angle changed by 50°, the summed pixel values further reduced by 67%. The reduction in the observed summed pixel values of surfaces with a relative angle other than 90° is due to the foreshortening of surfaces angled to the camera.

Model calibration

The summed pixel values are compared to the amount of Tinopal SWN for the respective 10 exposure situations (see Fig. 5a–j). For each exposure situation, a Pearson correlation coefficient (*R*) is provided. The correlation coefficients are below 0.28 for 5 out of 10 exposure situations, that is, pouring LV liquid (Fig. 5a), pouring HV liquid (Fig. 5b), rolling HV liquid (Fig. 5d), manually handling objects immersed in LV liquid (Fig. 5e) and handling objects contaminated with powder (Fig. 5h), indicating no to low correlations between measured amounts of Tinopal SWN and summed pixel values for these exposure situations. For the remaining five exposure situations, the correlation coefficients are greater than 0.7, which indicates strong correlations. However, strong correlation coefficients, for both low and high viscosity liquids, are observed only for spraying.

To investigate the performance of a predictive model the data of the two spraying exposure situations were combined, thus excluding the effect of the viscosity of the liquid and have a larger dataset available (see Fig. 6), based on which a second-order statistical fit model was derived (equation 1).

$$\text{Exposure mass } (\mu\text{g}) = -8 \times 10^{-18} \cdot \left\{ \sum (\text{pixel value}) \right\}^2 + 2 \times 10^{-7} \cdot \sum (\text{pixel value}) \quad (1)$$

This model can be used to estimate the loaded amount of Tinopal SWN on each body part (excluding hands) for spraying activities.

Model validation

In Fig. 7a, the estimated amounts of Tinopal SWN (using equation 1) are compared with the corresponding chemically analysed amounts of Tinopal SWN for the remaining 75% of the data for spraying both low and high viscosity liquids. The correlation coefficient is relatively

strong (*R* = 0.77) for the remaining 75% dataset and its distribution around 1:1 line is rather uniform.

The measured and estimated amounts of Tinopal SWN on large surface areas (i.e. parts of coveralls) are compared in Fig. 7b. The estimated median amounts of Tinopal SWN are almost similar to the measured median amounts of Tinopal SWN (factor of difference ≈1) for both low and high viscosity liquids. For both types of liquid, the measured and estimated Tinopal SWN loadings are lognormally distributed, and the 25th and 75th percentiles of the distribution of the measured amounts of Tinopal SWN lie within the respective interquartile ranges (IQR) of the estimated amounts. As shown in Fig. 7c, for smaller surface areas (i.e. patches), the estimated median amounts of Tinopal SWN are much lower than the measured median amounts of Tinopal SWN, with a factor of difference ≈5 for both types of liquid. For patch samples, the estimated IQR fails to cover both 25th and 75th percentiles of the measured Tinopal SWN amount for both types of liquid, indicating that there is a mismatch between the estimated and measured amounts of Tinopal SWN. It should be noted that the amounts of Tinopal SWN on the patches are based on the surface area of the patch itself, which is not extrapolated to reflect the surface of the corresponding body part as a whole.

When the distribution of the surface loading of Tinopal SWN on different parts of the coverall are compared in Fig. 8, the fluorescence method estimates an almost uniform exposure pattern which is similar to the measured levels in Kasiotis *et al.*, (2020). The highest median surface loading of ~0.06 µg/cm² is estimated for the lower right arm, while the lowest median surface loading of ~0.003 µg/cm² is estimated for the head. The estimated surface loading distribution covers the entire range of the measured surface loading distribution for each body part (except the head). For 6 out of 10 body parts, that is, upper right leg, upper left leg, upper left arm, upper right arm, back torso, and front torso, both 25th and 75th percentiles of the measured surface loading distributions are within the respective estimated IQR, and the estimated median surface loading is slightly higher by an average factor of 1.17. For the four remaining body parts, that is, lower right arm, lower left arm, lower right leg and lower left leg, the measured median surface loadings lie within the respective estimated IQR, and the estimated median surface loading is slightly lower by an average factor of 1.24. In case of head exposure, the measured median surface loading lies beyond the estimated IQR but within the estimated distribution range (i.e. within the bounds of minimum and maximum

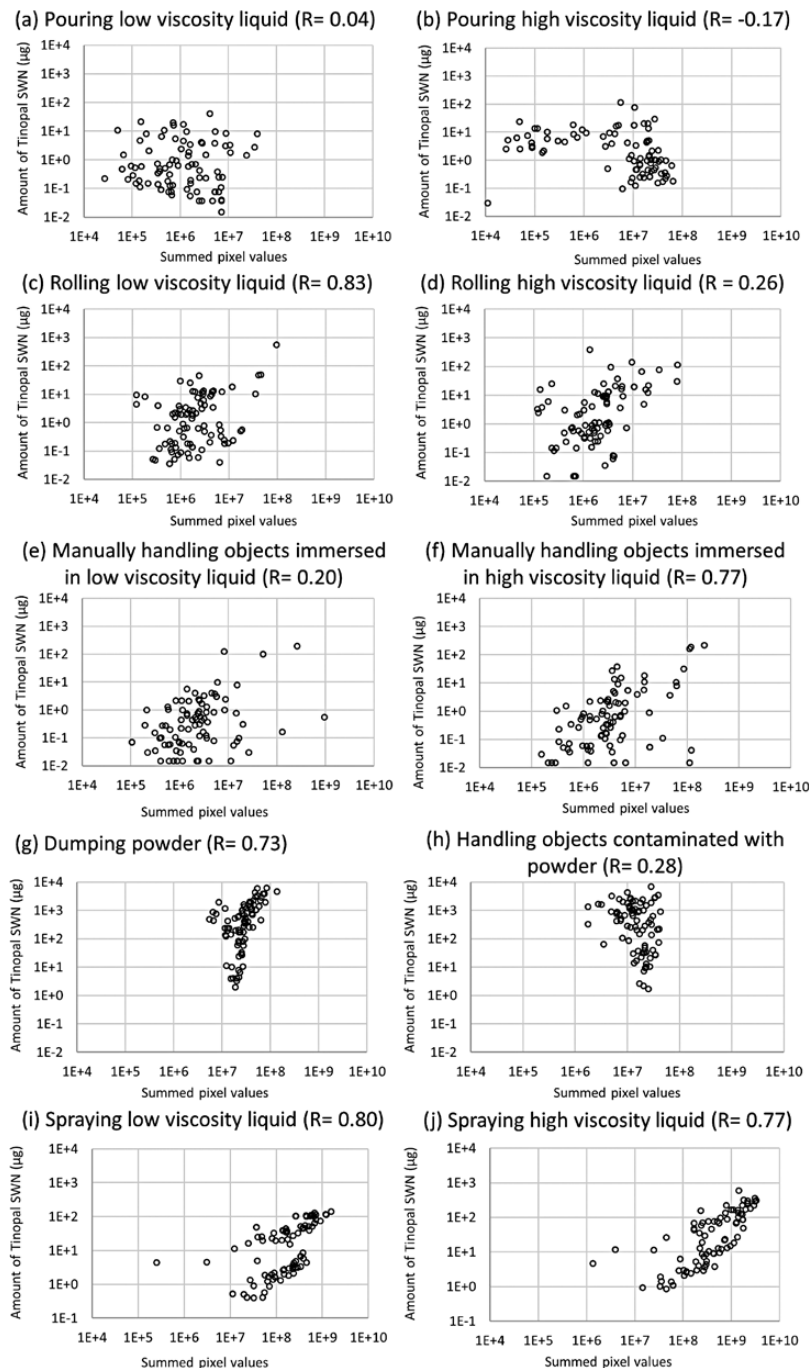


Figure 5. Amount of Tinopal SWN measured by the chemical analysis and corresponding summed pixel values as obtained by the fluorescence method after (a) pouring low and (b) high viscosity liquid, (c) rolling low and (d) high viscosity liquid, (e) manually handling objects immersed in low and (f) high viscosity liquid, (g) dumping powder, (h) handling objects contaminated with powder, and (i) spraying low and (j) high viscosity liquid.

estimated values) and the estimated median surface loading is lower by a factor of 7.15. The results show that the amount of Tinopal SWN and the related

surface loading on individual body parts can be estimated for spray activities on large surfaces like (parts of) coveralls.

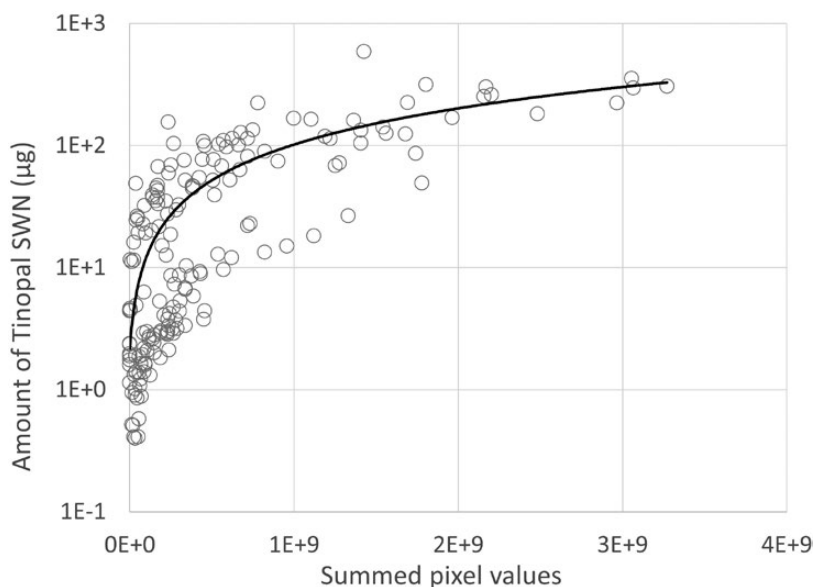


Figure 6. Amount of Tinopal SWN measured by the chemical analysis and corresponding summed pixel values as obtained by the fluorescence method for spraying both low and high viscosity liquids.

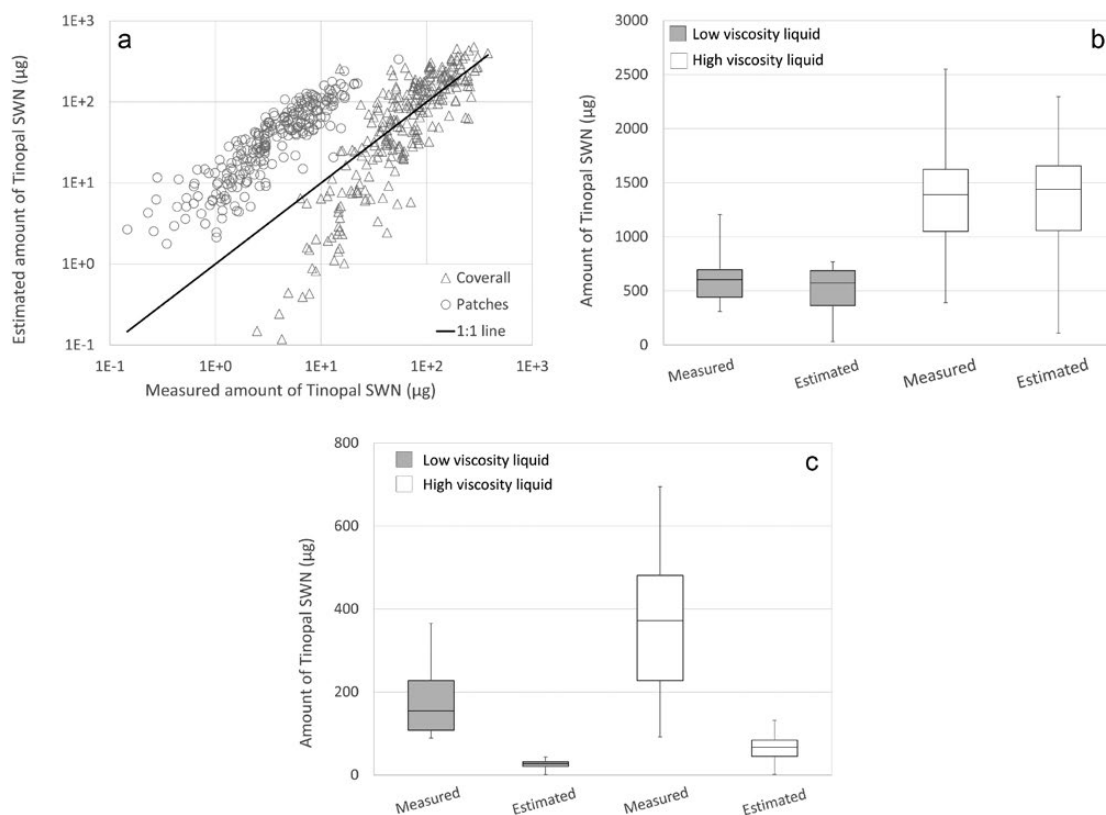


Figure 7. Comparison between the measured (from chemical analysis) and estimated (from fluorescence method) Tinopal SWN amounts after spraying liquids for (a) experiments with both coveralls and patches as sampling matrices, (b) only coveralls, and (c) only patches.

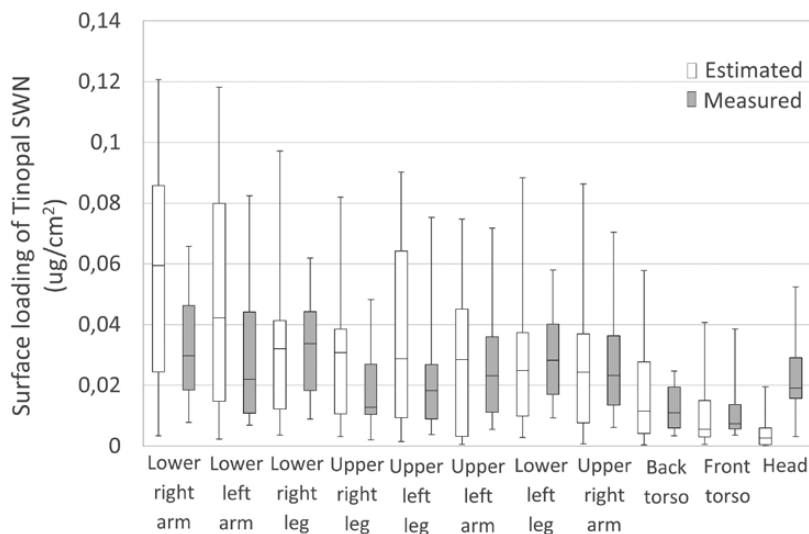


Figure 8. Estimated surface loading distributions of Tinopal SWN on coverall and headband for different body parts after spraying in the decreasing order of estimated median values and their comparison with measured distributions.

Discussion

The current study aimed to develop a fluorescence method capable of estimating dermal exposure on different body parts based on obtained light intensity from photographs of workers exposed to a fluorescent tracer. This study showed that the light intensity emitted by the tracer Tinopal SWN can be detected and the exposed tracer amount can be quantified with this method up to a surface loading limit of 1.5 µg/cm².

Although this was not quantified during the experiments, it is expected that for a given planar surface, with an increase in the surface loading, Tinopal SWN starts stacking. This may result in a 3-dimensional structure of Tinopal SWN on the planar surface. Since the fluorescence method processes 2-dimensional photographs and lacks the depth perception of the stacked Tinopal SWN, it consequently considers the projected 2-dimensional image of the Tinopal SWN as present on the surface. Thus, the magnitude of the corresponding summed pixel value does not increase with increasing layers of Tinopal SWN present on the surface. This phenomenon of stacking is plausible for high amounts of Tinopal SWN on hands, large droplets or splashes that result in localized exposure, and probably even more for situations involving powders.

Kasiotis *et al.* (2020) reported that spraying liquids resulted in a more uniform distribution of Tinopal SWN over the body compared to tasks like rolling, pouring and handling immersed object, during which, in general,

only part of the body is exposed due to spills and/or splashes and it tends to stack relatively high amounts of Tinopal SWN on relatively small surface areas (i.e. high surface loading). In case of spraying, it is generally expected that the surface loading will not reach the quantification limit of the present fluorescence method. In fact, under the given experimental conditions, Kasiotis *et al.* (2020) observed 0.085 µg/cm² to be the highest surface loading of Tinopal SWN on a part of the coverall after spraying. While these experiments were standardized and exposure durations were relatively short, it is expected that in real life settings the surface loading can be higher. Nevertheless, it is still assumed that the quantification limit of 1.5 µg/cm² holds true for spraying too when higher surface loadings are occurring.

Moreover, we obtained no to low correlations between measured levels of Tinopal SWN and obtained summed pixel values for exposure situations such as pouring of both low and high viscosity liquids, rolling of high viscosity liquid, and handling of objects immersed in low viscosity liquid. The tendency towards stacking and, thus, high surface loading results in the light intensity detection saturation of the fluorescence method, which may explain these low correlations. However, rolling of low viscosity liquid and handling objects immersed in high viscosity liquid resulted in stronger correlations. We, therefore, need to investigate the effect of the liquid viscosity on the obtained pixel values to justify the inconsistency in the correlation factors. For the

situations involving powders (i.e. dumping and handling contaminated objects), we observed:

- a lower quantification limit of 0.18 mg/cm² (without any observation of an upper limit),
- poor light intensity of Tinopal SWN powder compared to Tinopal SWN dissolved in a liquid formulation and strong correlation factor of 0.8 (for powder dumping) and low correlation factor of 0.28 (for handling contaminated objects) between measured amounts of Tinopal SWN and obtained summed pixel values.

These three observations can be considered interdependent by assuming the observed light intensity to be proportional to the available surface area of Tinopal SWN on which UV rays can interact and Tinopal SWN particles tend to agglomerate in powder form resulting in a smaller relative surface area to emit light. The correlation difference between dumping powder and handling contaminated objects cannot directly be explained based on the currently available data. It is possible that for dumping powders, an evenly distributed pattern of exposure explains the better correlation like for spraying, where handling contaminated objects might result in more stacking of particles on the exposed surface.

For the aforementioned situations, which either result in exposure patterns with high surface loading or involve powders, a quantitative estimation of dermal exposure levels is not possible with the current experimental setup. The fluorescence method can still provide qualitative information about the exposure patterns related to different exposure scenarios, for instance more uniformly distributed exposure on the whole body versus concentrated exposure on certain parts of the body. This can be an added value in addition to the more established dermal measurement methods by means of interception or removal, that generate exposure data through chemical analysis of collected samples without being able to pinpoint the exact location of this exposure. The fluorescence method, with Tinopal SWN as a tracer, which has no evident toxicity, can also be used for training purposes by visualizing workers the exposure patterns resulting from their work methods in a qualitative way. The visual information gathered by using the fluorescence method in a pilot study can also be used to determine the optimal measurement strategy to apply during a measurement campaign, for instance, to determine the best method for the measurements (e.g. patches or coveralls) or to determine the placement of patches.

In the present study, a similar methodology as used in different studies in the past (namely translation of pixel values to exposure values using software) is applied

(Fenske *et al.*, 1986; Roff, 1994; Bierman *et al.*, 1998; Brouwer *et al.*, 1999; Schneider *et al.*, 2000; Wheeler and Warren, 2002; Galea *et al.*, 2014). When comparing the current study with previous studies, it can be noted that each study set-up has its advantages and disadvantages. For example, the calibration curve as described in Roff (1997) was generated by applying amounts of Tinopal directly on the skin of the forearm of a volunteer, and thus the effect of the characteristics of the skin on the measured intensity could be studied. By applying Tinopal on different types (smooth and rough [Tyvek]) of surfaces, in the present study the effects of surfaces, stacking and amount could be studied. When comparing the set-ups, the FIVES system has the advantage that it better captures the angles of the body due to the set-up of the lighting and the combination of a diffuse light source and a point source, which results in better information about the orientation of contaminated surfaces in relation to the camera in comparison to the set-up as used in the present study. However, the FIVES system is stationary and difficult to employ in for example work-site visits, while the set-up of the current study is relatively mobile, and easily constructed in other locations. Another notable difference between the studies is that the current study focused on uniform, non-fluorescent and mainly non-reflective surfaces (cotton and Tyvek patches and coveralls), which already proved to be challenging. Expanding the applicability domain of this fluorescence technique to the skin (and especially the hands), as done by Roff *et al.* (1997), Fenske *et al.* (1997), and Brouwer *et al.* (1999), will be increasingly challenging due to the additional factors that would have to be taken into account, like skin pigmentation and the possible influence of perspiration and sebaceous fluids, as identified by Roff (1997) and Fenske *et al.* (1997).

Despite the promising results of this study, at least for the spraying scenarios the scope still remains limited. The dermal exposure patterns related to everyday exposure scenarios like spills or splashes may reach localized parts of the body, which result in higher exposure amounts per surface area. Such high amounts lie beyond the current quantification limit of the method. Thus, efforts are needed to improve its estimation accuracy and applicability domain. In terms of future efforts, the image quality of the UV-illuminated photographs could perhaps be improved by using a monochrome light sensor with a narrow bandpass filter, which narrows the range of light waves captured by the camera closer to the 430–436 nm emitted by Tinopal SWN, as has been used in the previous studies as well (Fenske *et al.*, 1986; Roff, 1994; Bierman *et al.*, 1998). This may reduce the background noise or other lighting interference

and thus decrease the need for correction, which in turn reduces the uncertainty in the estimates. Another possibility can be the development of functionality in the software that allows the method to identify the fraction of the body surface that is contaminated based on the observed light intensity. This would be more of a semi-quantitative evaluation, but would generate important information on exposure patterns that, for instance, can be used to design relevant control strategy. In addition, the analyzed surface ideally needs to be perpendicular to the camera, which in practice almost never is the case as a person is always 3-dimensional, and the coveralls used as sampling matrix tend to introduce even more folds, etc. One way to overcome this may be to remove the coverall from the person and place it on a flat surface, but this would involve more work and may result in transfer of the tracer from one place on the matrix to another when handling the matrix. The relation to the contours of the body and other parts of the body (hands, head) can also get lost. The lack of depth perception may be improved by using a point light source in conjunction with a diffuse lighting source. The difference in measured intensity between a point source lighting and a diffuse light source can be used to correct for surface curvature, as is shown by application of the FIVES method (Roff, 1994). Alternatively, newer methods can be applied for 3-dimensional imaging, such as the Xbox Kinect camera, which is capable of making 3-dimensional scans of persons.

Furthermore, the current model was calibrated only on Tinopal SWN as a fluorescent tracer. The type of effect that different tracers may have on the model outcomes could be explored. Additionally, for the method to be applicable in practice, it is important to investigate how representative deposition of the tracer is for the hazardous chemical(s) of interest in the formulation at hand (e.g. paint), and thus determine their quantitative relationship, as has been done with pesticides in the agriculture sector (Roff, 1994). It may also be important to investigate if the transfer rate of the tracer and the hazardous chemical(s) is consistent.

After these improvements, the fluorescence method can be expected to be applicable for exposure scenarios with non-uniform exposure patterns caused by droplets, spills, and/or splashes, for exposure scenarios involving powders, as well as to estimate hand exposure.

Conclusion

The fluorescence method, as developed within the SysDEA project, shows promising results and can be used for the quantification of dermal exposure on

different body parts (excluding the hands) for the spraying-like scenarios that have a more uniform exposure pattern. For other scenarios, the method can provide valuable qualitative information.

Further research on the improvement of the quality of UV-illuminated photographs, depth perception or three dimensional scanning of the tracer as present on the surface and the effect of different tracers may improve the estimation accuracy and applicability domain of the this fluorescence method for assessment of potential dermal exposure.

Supplementary data

Supplementary data are available at *Annals of Work Exposures and Health* online.

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Conflict of interest

The authors declare no conflict of interest relating to the material presented in this article. Its contents, including any opinions and/or conclusions expressed, are solely those of the authors.

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