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Modeling Dermal Exposure—An Illustration for Spray Painting Applications

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This article presents a conceptual, mathematical model of dermal exposure resulting from aerosol deposition on human forearm hair. The model is applicable to exposure scenarios where dermal deposition is governed by aerosol impaction, interception, and diffusion mechanisms. The model employs filtration theory, single fiber efficiency equations, and a modified potential airflow approximation. The results are extended, using previously published results, for application to dermal deposition on the forearm during spray painting. The average ($N = 8$) predicted dermal deposition of 1,6-hexamethylene diisocyanate as collected on a 10-cm² tape strip is 108.9 (± 70.3) pmol, whereas field measurements indicated an average of 168.6 (± 82.0) pmol per strip. The corresponding measured average dermal flux was 3.63 pg/cm²s (± 1.34); the prediction was 2.24 pg/cm²sec (± 1.25). The study calls attention to the importance of body hair both for modeling and measuring dermal exposures.

Keywords aerosols, dermal exposure, impaction, isocyanates, mathematical modeling

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INTRODUCTION

Increasing attention is being given to dermal exposure assessment and the skin as an important route of entry into the body for toxic compounds.⁽¹⁾ Several studies report methods for exposure assessment^(2,3) and modeling for some of the data collected.⁽⁴⁾ In addition, more general conceptual models of the dermal exposure process have also been explored,⁽⁵⁾ some of which are specific to spray painting.⁽⁶⁾ Although some gases are directly absorbed by the skin (e.g., hydrogen cyanide), particle deposition is likely to be a more important source of dermal exposure for airborne contaminants.

Mechanisms of aerosol deposition include gravitational settling, electrostatic forces, inertial impaction, interception, and diffusion. In an important and often neglected publication, Asset and Pury⁽⁷⁾ demonstrated that the hairs on the body act as effective sites for the inertial impaction of airborne particles. They noted virtually no deposition on the hairless (volar) side of the human forearm but significant deposition on the dorsal side covered with hair.

Model Development

The following model is based on the assumption that particle deposition on human body hair is governed by impaction, interception, and diffusive transport mechanisms. The following definitions (with dimensions) are used with the understanding that a specific particle size is being considered (i.e., monodisperse); for a polydisperse aerosol summation over all sizes is required.

F = the particle flux to the skin = $M/(L^2T)$

J = the particle flux in the air = $M/(L^2T)$

C = particle concentration in air = M/L^3

U = air velocity = L/T

η = efficiency of deposition

Given a uniform concentration in the freestream the following relationship holds:

$$F = \eta J = \eta C U \quad (1)$$

The basic dermal deposition model treats the forearm hair as a filter of thickness h , which is on the order of 0.5 cm and defines the deposition efficiency per unit area according to fiber filter theory⁽⁸⁾ as:

$$\eta = 1 - \exp(-\gamma h) \quad (2)$$

Here, γ , the fractional efficiency per unit thickness of the hair bed, is the product of the overall single hair efficiency (ω), the effective hair length per unit volume (L), and the hair diameter (d_f).

$$\gamma = \omega L d_f \quad (3)$$

The effective hair length per unit volume is the product of the average length of a hair fiber (l_f) and the areal fiber density λ_f , (hairs per cm²), divided by the thickness of the hair bed.

$$L = \frac{l_f \lambda_f}{h} \quad (4)$$

Whereas solidity is often used as a parameter in fiber filter theory, the hair length per unit volume is used here. This is intuitively reasonable because particles are collected all along the length of the fiber throughout the depth of the hair bed (filter). See Hinds⁽⁸⁾ for details of this derivation. The overall single hair fiber efficiency is approximated as the sum of single fiber efficiencies due to the mechanisms of impaction, interception, and diffusion. Due to the horizontal orientation of the forearm with respect to the airflow, gravitational deposition is considered negligible on the dorsal surface of the forearm for the applications explored here.

Single fiber efficiency for impaction is determined by correlations developed by Wessel and Righi⁽⁹⁾ and a potential flow approximation for airflow around the forearm. Inertial impaction is characterized by the Stokes number and the particle Reynolds number.

$$Stk = \frac{\rho_p d_p^2 U_0}{18\mu R} \quad (5)$$

$$Re_0 = \frac{\rho_g d_p U_0}{\mu} \quad (6)$$

When non-Stokes drag is significant and the particle Reynolds number is less than 1000, the Stokes number is corrected to an "effective Stokes number" as follows:

$$Stk_e = Stk \cdot \psi(Re_0) \quad \text{where} \quad \psi(Re_0) = \frac{3[\sqrt{c}Re_0^{1/3} - \tan^{-1}(\sqrt{c}Re_0^{1/3})]}{c^{3/2}Re_0} \quad (7)$$

and $c = 0.158$. According to Wessel and Righi⁽⁹⁾ the single hair efficiency per unit length due to impaction is then given by either of the following empirical correlations:

for $0.125 \leq Stk_e \leq 0.5$ use:

$$\omega_I = 0.01978749(\ln[8Stk_e]) + 0.513645(Stk_e - 0.125) - 0.0482858(Stk_e - 0.125)^2$$

for $Stk_e > 0.5$ use:

$$\omega_I = \left[1 + \frac{1.54424}{(Stk_e - 0.125)} - \frac{0.538013}{(Stk_e - 0.125)^2} + \frac{0.2020116}{Stk_e - 0.125} \right]^{-1} \quad (8)$$

To complete the deposition model, an estimate of the air velocity (U_0) approaching the hair fibers is needed. A simple approximation is employed based on a modified potential flow solution for airflow around the forearm. In this model, the forearm is treated as a circular cylinder of radius a , in the cylindrical coordinate system due to symmetry in the (r, θ)

plane (Figure 1). The radial air velocity, normal to the cylinder (forearm) predicted by potential flow theory,⁽¹⁰⁾ is:

$$U_0 = v_r = U_\infty \cos \theta \left(1 - \left[\frac{a}{r} \right]^2 \right) \quad (9)$$

Here, U_∞ is the undisturbed air velocity upstream of the arm. Assuming deposition is primarily on the length of the hair fiber parallel to the forearm surface at the outer edge of the hair bed, the radial velocity is:

$$v_r = U_\infty \cos \theta \left(1 - \left[\frac{a}{a+h} \right]^2 \right) \quad (10)$$

To get an average radial velocity over one half of the upstream surface of the forearm, the following integration is required:

$$\bar{v}_r = \frac{2}{\pi} U_\infty \int_{\pi}^{\pi/2} \left(1 - \left[\frac{a}{a+h} \right]^2 \right) \cos \theta \, d\theta \quad (11)$$

with the assumption that the forearm is a cylinder of diameter 7.5 cm this is:

$$\bar{v}_r = \frac{2}{\pi} U_\infty \left(1 - \left[\frac{3.75}{4.25} \right]^2 \right) \approx 0.15 U_\infty \quad (12)$$

Figure 1 illustrates the geometry for modeling the airflow field around the forearm in the hair bed.

The single fiber efficiency due to interception is:⁽⁸⁾

$$\omega_R = \frac{1}{2Ku} \left[2(1+R) \ln(1+R) - (1+R) + \frac{1}{1+R} \right] \quad (13)$$

where

$$R = \frac{d_p}{d_f} \quad Ku = 2 \quad (14)$$

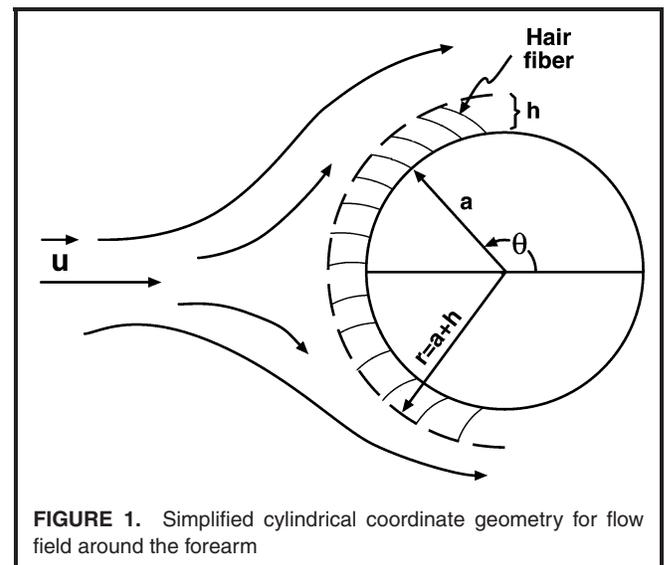


FIGURE 1. Simplified cylindrical coordinate geometry for flow field around the forearm

The maximum single fiber efficiency due to interception is $1+R$, and the Kuwabara number, Ku , is set at 2 based on low solidity (<0.005) of the hair bed.

Additional deposition mechanisms for diffusion and an augmented diffusion-interception term are included following Hinds:⁽⁸⁾

$$\omega_D = 2(Pe)^{-2/3}$$

$$Pe = \frac{d_f U_0}{D} \quad (15)$$

where Pe is the Peclet number and D is the particle diffusion coefficient. The single fiber efficiency due to interception augmented by diffusion when $Pe > 100$ is:

$$\omega_{DR} = \frac{1.24R^{2/3}}{\sqrt{KuPe}} \quad (16)$$

The model for efficiency for a single size particle is thus:

$$\eta = 1 - \exp(-\omega \lambda_f l_f d_f) \quad (17)$$

$$\omega = \omega_I + \omega_R + \omega_D + \omega_{DR} \quad (18)$$

The single fiber efficiency for impaction is calculated using the following equations for Stokes and Reynolds numbers:

$$Stk = \frac{\rho_p d_p^2 \bar{v}_r}{9\mu d_f} \quad (19)$$

$$Re_0 = \frac{\rho_g d_p \bar{v}_r}{\mu} \quad (20)$$

The objective of this work was to first evaluate the model above by comparing the predictions with the original data published in Asset and Pury.⁽⁷⁾ This exercise produced reasonably encouraging results and the model was subsequently expanded for application to spray painting tasks. A small-scale field study was conducted to examine this expanded model.⁽¹¹⁾

Comparison to Asset and Pury's Data

In the original study by Asset and Pury,⁽⁷⁾ human volunteers placed their forearms in a wind tunnel test section (1 ft²) with air velocities of either 2 mph or 5 mph (0.89–2.24 m/sec). A triphenyl phosphate aerosol was produced with two smoke generators. Particle size was determined with a cascade impactor, and concentrations were determined with isokinetic sampling. It is unclear whether the isokinetic sampling was also used with the impactor. A colorimetric chemical analysis method was used to determine the amount of triphenyl phosphate recovered from the skin after an alcohol wash with a special receptacle. The authors reported the efficiency of recovery of triphenyl phosphate from the skin as 100% ±8.

The particle size distribution used in the wind tunnel tests was reported as having a mass median diameter of 6.5 μm with 95% of the mass below sizes that varied between 12 μm and 17 μm. This suggests a geometric standard deviation (GSD) between 1.4 and 1.8 if one assumes a lognormal size distribution. There was no information in the original work on the forearm diameters or the length or density of forearm

hair on the volunteers. To account for this a range of values for each of these variables was used for the model inputs. These were estimated as 5–10 cm for the forearm diameter, 1–2 cm for forearm hair length, and 13–23 hairs per cm² as the areal fiber density. The range of forearm hair diameters was estimated at 38–62 μm. Thirty-two different combinations of these variables were run for each of the two velocities examined by Asset and Pury.⁽⁷⁾ The model equations listed above were entered into a computer code and the particle size distribution was represented as 100 discrete 1-μm intervals from 0–100 μm. For each interval the efficiency was calculated using the midpoint of the interval. The mass deposition for the given size range was then calculated by multiplying the size-specific efficiency by the mass fraction for that interval. This was summed to give the overall mass-deposition efficiency for comparison with the data of Asset and Pury.

Asset and Pury⁽⁷⁾ reported average deposition efficiency on the hairy side of the forearm as 0.17% ± 0.22 at a wind tunnel air velocity of 0.89 m/sec (2 mph). The data were from 11 samples taken on six different individuals. Six of the 11 samples reported no detectable mass. Their measured efficiencies ranged from 0.0% to 0.65%. At a wind speed of 2.23 m/sec (5 mph) the average deposition efficiency was 0.90% ± 0.15 based on eight samples from four individuals. The measured values ranged from 0.39% to 1.9%. Model predictions for dermal deposition at the 0.89 m/sec speed ranged from 0.2% to 1.9%. At 2.23 m/sec the predicted range of deposition efficiency was 0.3–2.9%. In general there was reasonable agreement between the predicted and measured deposition efficiency ranges.

METHODS

The field study was conducted in an autobody repair shop using isocyanate-based paint (1,6-hexamethylene diisocyanate, HDI) in a semidowndraft, enclosed spray booth. This booth had a cross sectional area of 10.9 m². The booth airflow of 2.9 m³/sec came in through a 2.74 m by 3.35 m filter bed located in the ceiling. This was determined by velocity traverse with a rotating vane anemometer at the filter face, resulting in an average booth air velocity of 0.27 m/sec.

The primary objects painted during the study were cars and vans, that were oriented so that the worker spent much of his time spraying perpendicular to the flow of air through the booth. The spray gun was a conventional compressed air spray gun (DeVilbiss Plus gun; DeVilbiss, Maumee, OH) operated at 60 psig. All of the spraying was done by the same right-handed worker who was sampled for dermal deposition of HDI on the right forearm and hand using a modification of the tape strip method, which has been developed and used to measure dermal exposure to HDI,⁽¹¹⁾ multifunctional acrylates,⁽¹²⁾ and naphthalene as a marker for jet fuel exposure.⁽¹³⁾ The worker's forearm radius (a) was 4.5 cm.

In this study, HDI deposited on skin was collected by removal of stratum corneum with Cover-Roll BSN medical

adhesive tape (Beiersdorf AG, Hamburg, Germany). Three successive tape strips (10 cm²) were applied to various locations on the exposed hands and forearms directly following spray application of paint. The HDI on the tape-strips was derivatized with 1-(2-methoxyphenyl) piperazine and quantified as the urea derivative by liquid chromatography/mass spectrometry. The average recovery of HDI from tape, based on controlled experiments in which solutions containing known amounts of HDI were spiked onto tape, was 99.3% (95% confidence interval [CI] 97.1–102%).⁽¹¹⁾ The amount of HDI collected from each site on the skin was determined by summing the levels collected with the three successive tape strips.

To apply the model described above to spray painting tasks, it is necessary to obtain estimates of the particle flux to the forearm. This requires both the concentration and size distribution of the aerosol as well as the air velocity. The concentration used was the breathing zone concentration, which was measured for the worker during each task evaluated. The concentration was adjusted to a time-weighted average for the spraying period under the assumption that exposure during the nonspraying period was 0.

The size distribution of the overspray was estimated based on published data⁽¹⁴⁾ as having a mass median diameter of 14.7–19.2 μm and a GSD of 1.7. The larger mass median diameter (MMD) is associated with spraying perpendicular to the airflow, whereas the smaller size was measured during back-to-flow orientations. The value of 19.2 μm is selected here as the most representative estimate, but some sensitivity analyses are conducted with the smaller diameter as well. The remaining pieces of information required include the areal fiber density set at 18 fibers/cm² (from ICRP data),⁽¹⁵⁾ the length of forearm hair measured at 1.74 cm, and the diameter of the

forearm hair measured at 49.6 μm (±11.7). Table I summarizes the information for input to the model.

RESULTS

The equations above were translated into a FORTRAN program (Digital Visual Fortran version 6.0). Given the input parameters described above, predictions of dermal deposition for five different field-spraying conditions were generated. Eight corresponding field samples were obtained, four from the dorsal surface of the right hand and four from the dorsal surface of the forearm. The results of the measured and predicted values are shown in Table II as the mass of HDI on the 10-cm² tape strip. The predicted values also show a range that was determined by using the smaller MMD and also by using hair diameters equal to the measured mean plus or minus one standard deviation. Figure 2 shows the corresponding plot of the measured vs. predicted amounts. The error bars in the model predictions reflect the range of uncertainty based on the MMD and hair fiber diameters. The experimental errors were estimated to be ±10% based on variability in the standard curve and the extraction efficiency of HDI from tape. The average value of the eight measurements was 168.6 pmol per tape, whereas the corresponding average prediction was 108.0 pmol per tape, an underestimation of approximately 36%.

DISCUSSION

The model of dermal exposure presented here does a reasonable job of estimating the average dermal deposition of HDI on the forearm and hand for the processes examined. Whereas there is significant task-to-task variability in the prediction, there is a distinct underestimation bias consistent with the model limitations. Although there are many sources of uncertainty in the model, the use of the breathing zone concentration may be an important factor in this

TABLE I. Model Input Parameters that Remained Constant for All Processes Sampled

Parameter	Input Value	Method of Assignment
l_f —forearm hair length	1.74 cm (±0.08)	Measured
d_f —diameter of forearm hair	49.6 μm (±11.7)	Measured
λ_f —forearm areal fiber density	18 per cm ²	Reference 15
a —forearm radius	4.5 cm	Measured
h —thickness of hair bed	0.5 cm	Estimated
MW—molecular weight, HDI	168.7 g/mole	Reference value
MMAD of overspray	19.2 μm	Reference 14
GSD of overspray	1.7	Reference 14
U—spray booth air velocity	0.27 m/sec	Measured

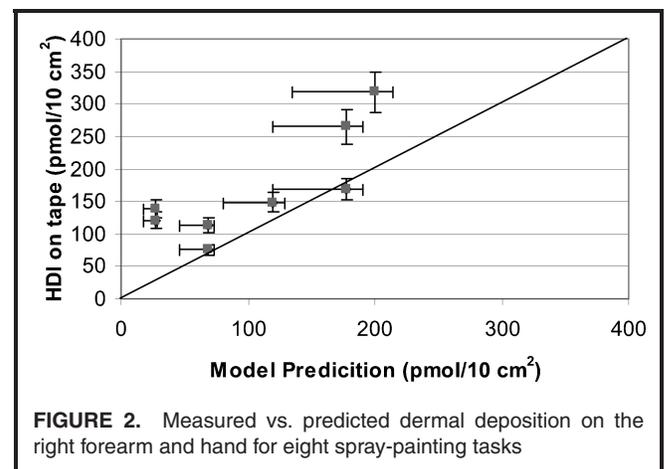


FIGURE 2. Measured vs. predicted dermal deposition on the right forearm and hand for eight spray-painting tasks

TABLE II. Measured and Predicted Dermal Deposition of HDI on the Right Hand and Forearm with the Corresponding Breathing-Zone Concentration

	Sample 3 1/28/05	Sample 1 2/18/05	Sample 2 2/18/05	Sample 7 2/18/05	Sample 8 2/18/05	Sample 14 2/18/05	Sample 19 2/18/05	Sample 20 2/18/05
Sample Location	Hand	Hand	Forearm	Hand	Forearm	Forearm	Hand	Forearm
HDI measured on tape strip pmol/tape	318.3	138.4	121	75.2	113	149	265	169
HDI prediction and range pmol/tape	199.8 135–215	26.7 18–28	26.7 18–28	68.2 46–73	68.2 46–73	119.7 81–121	177.5 120–191	177.5 120–191
Sample duration (min)	18	12	12	13.5	13.5	10	18.5	18.5
Spray time (min)	15	10.6	10.6	12.3	12.3	9.2	16.6	16.6
HDI breathing-zone concentration during spraying mg/m ³	0.0132	0.0025	0.0025	0.0055	0.0055	0.0129	0.0106	0.0106

underestimation. It is likely that the hand and arm experience a greater concentration due to their proximity to the overspray cloud and the back and forth motion through it. Splatter is also an additional source of error that may lead to the negative bias.

There is uncertainty in determining the size distribution of the overspray. Almost all methods in the literature use some type of aspirating sampler (e.g., impactor, filter cassette) to measure the size of paint overspray particles. These sampling techniques are biased and miss the larger sizes or undersample them rather severely. In addition, the evaporation of solvents complicates the determination of particle size. The assumption that all of the airborne mass of isocyanate is in the particle phase may lead to some overestimation of deposition if in fact a significant portion of the HDI is vapor. There is also an uncertainty associated with the measurement method because some of the HDI deposited on the hairs and skin may penetrate into the skin, react, or be only partially captured by the tape stripping process.

The model is rather simplistic both in the level of detail to the airflow in the hair bed and in the assumption that efficiency is exclusively due to the normal velocity component through the fiber bed. The forearm is, of course, not a filter because the airflow cannot go through the skin. The tangential air velocity around the forearm will carry the particles that are not initially collected through the hair bed, and additional collection will occur. The model in its present form does not account for this and, therefore, this may be a significant source of the underestimation. The model does not include deposition from electrostatics nor gravity, which also may contribute to the underestimation.

Despite these limitations, the approach is encouraging and further development is under way. The importance of the hair as a major determinant in dermal deposition indicates that sampling methods that fail to account for it may result in biased estimates of dermal exposure. The tape strip approach is promising in that it accounts for this important deposition mechanism. There are, of course, spills and splashes that the model will not account for although these can sometimes be identified as outliers. It should also be noted that diffusion

and the diffusion interaction term contributed little to overall deposition for the conditions run here.

CONCLUSIONS

The original work of Asset and Pury⁽⁷⁾ indicates that the basic mechanism of inertial impaction on the body hairs is an important determinant of dermal exposure to airborne contaminants. This information was used to develop a simple conceptual model of dermal deposition based on treating the forearm hair as a fibrous filter and applying basic single fiber efficiency equations. The model agreed reasonably well with the data of Asset and Pury. The model was subsequently extended to spray painting operations and examined with a small-scale field study. The average predicted dermal deposition was 64% of the average measured value. The simple model used here may have the potential to provide dermal exposure estimates for spray painting applications, and further research is currently under way to improve its predictive capability. The importance of the body hair to the deposition process suggests that sampling methods need to account for this mechanism—something the tape strip approach can do reasonably well.

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