



Role of clothing in both accelerating and impeding dermal absorption of airborne SVOCs

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2
3 1 **Title:** Role of clothing in both accelerating and impeding dermal absorption of airborne
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5 2 SVOCs
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25 **Running title:** Clothing enhances dermal uptake of airborne SVOCs

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28 financial interests.

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6 32 **Abstract**

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8 33 To assess the influence of clothing on dermal uptake of SVOCs, we measured uptake of
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10 34 selected airborne phthalates for an individual wearing clean clothes or air-exposed clothes
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12 35 and compared these results with dermal uptake for bare-skinned individuals under
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14 36 otherwise identical experimental conditions. Using a breathing hood to isolate dermal from
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16 37 inhalation uptake, we measured urinary metabolites of diethylphthalate (DEP) and di-n-
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18 38 butylphthalate (DnBP) from an individual exposed to known concentrations of these
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20 39 compounds for 6 hours in an experimental chamber. The individual wore either clean
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22 40 (fresh) cotton clothes or cotton clothes that had been exposed to the same chamber air
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24 41 concentrations for 9 days. For a 6-hour exposure, the net amounts of DEP and DnBP
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26 42 absorbed when wearing fresh clothes were respectively 0.017 and 0.007 $\mu\text{g}/\text{kg}/(\mu\text{g}/\text{m}^3)$;
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28 43 for exposed clothes the results were 0.178 and 0.261 $\mu\text{g}/\text{kg}/(\mu\text{g}/\text{m}^3)$ (values normalized
29
30 44 by air concentration and body mass). When compared against the average results for bare-
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32 45 skinned participants, clean clothes were protective, while exposed clothes increased
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34 46 dermal uptake for DEP and DnBP by factors of 3.3 and 6.5 respectively. Even for non-
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36 47 occupational environments, wearing clothing that has adsorbed/absorbed indoor air
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38 48 pollutants can increase dermal uptake of SVOCs by substantial amounts relative to bare
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40 49 skin.

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49 50 **Introduction**

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51 51 Dermal absorption of organic compounds directly from air has been observed for some
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53 52 volatile and semi-volatile compounds. In reviews by Rehal et al.¹ and Rauma et al.² a
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55 53 handful of volatile organic compounds (VOCs) have been observed to have dermal uptakes
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3 54 that are substantial compared to inhalation intakes. For example, Piotrowski^{3,4} found that
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5 55 nitrobenzene and phenol doses via dermal absorption were about 50% those due to
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8 56 absorption by inhalation. Weschler and Nazaroff^{5,6} argued that the dermal absorption dose
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11 57 from air could also compare with or exceed the dose due to inhalation for semi-volatile
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13 58 organic compounds (SVOCs) that meet specific criteria under steady-state conditions. In a
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15 59 refinement of that model for non-steady-state conditions, Gong et al.⁷ showed that timing of
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18 60 exposure can significantly influence dose due to resistance and accumulation within the
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21 61 dermis. In a test of the hypothesis that the dermal dose of SVOCs from air could be
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23 62 significant, Weschler et al.⁸ showed that dermal absorption was approximately equal to
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25 63 inhalation dose for six bare-skinned male participants exposed to diethylphthalate (DEP)
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28 64 and di n-butylphthalate (DnBP) for six hours in a chamber.
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30 65
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32 66 A few studies have evaluated how clothing may influence dermal uptake of organic
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35 67 compounds from air or by transfer from treated fabrics. Piotrowski³ found that clothing
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37 68 reduced dermal uptake of airborne nitrobenzene by about 20-30% but had no observable
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40 69 effect on phenol absorption.⁴ Organics that have been applied to clothing can be also
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42 70 absorbed. Blum et al.⁹ observed metabolites of a flame retardant in the urine of children
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45 71 who had worn clothing treated with this flame retardant. Similarly, subjects wearing
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47 72 permethrin-impregnated battle dress uniforms absorbed this insecticide as evidenced by
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49 73 urinary metabolites¹⁰⁻¹².

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54 75 We hypothesize that sorption to clothing acts either to reduce or to increase dermal
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56 76 uptake, depending on the extent to which the clothing has equilibrated with room air
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3 77 contaminants prior to wearing. For some compounds, the boundary layer of air adjacent to
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5 78 the skin presents greater resistance to transport than does the stratum corneum and viable
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8 79 epidermis.⁶ For such compounds uptake is sensitive to the magnitude of the boundary layer
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10 80 permeability⁷ and could be altered significantly by sorption to fabrics, especially for
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13 81 compounds with high air-fabric partition coefficients.
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18 83 For fabrics that are initially clean, adsorption to fabric fibers should decrease fabric
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20 84 permeability, and lower overall dermal uptake, by reducing diffusional flux through the
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22 85 fabric. With continued exposure, fabric permeability would increase as fabric surfaces
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24 86 equilibrate with SVOCs. Fabrics that are exposed to building air for extended periods (e.g.
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26 87 hanging up in a closet) may absorb a substantial quantity of SVOCs, or even reach
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28 88 equilibrium, prior to wearing. For these clothes, we predict that dermal uptake will be
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30 89 higher than uptake to bare skin.
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37 91 Our objective is to test this hypothesis with two compounds that have been predicted, and
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39 92 recently shown, to exhibit low dermal uptake resistance relative to mass-transfer
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41 93 resistance through the layer of air adjacent to body surfaces. In this study, we measure
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43 94 urinary concentrations and total excretion of DEP and DnBP metabolites during and after a
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45 95 participant is exposed for 6 hours to known air concentrations of DEP and DnBP for 2
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47 96 conditions: i) wearing freshly cleaned cotton clothing; ii) wearing previously clean cotton
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49 97 clothing that had been exposed to the phthalates for at least one week. Inhalation uptake is
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51 98 controlled with a breathing hood. Results are compared against results from six individuals
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3 99 who wore only shorts but were subjected to nearly identical conditions (results reported in
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6 100 Weschler et al.⁸).

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10 102 **Methods**

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13 103 The experiments reported here were integrated into the dermal uptake experiments
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15 104 reported by Weschler et al.⁸ and nearly all procedures, conditions and analytical methods
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17 105 are therefore identical. The clothed individual was exposed to phthalates in the same
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19 106 chamber at the same time as bare-skinned participants during two of the chamber
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21 107 exposure intervals, specifically Wednesday of the 1st week and Tuesday of the 2nd week.
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25 109 *Exposure chamber*

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28 110 The 55 m³ chamber housed two mixing fans, desks and chairs. The air exchange rate was
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30 111 maintained at 0.7 1/h and the temperature was controlled at 30°C. The relative humidity
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32 112 was not controlled and ranged from 20 to 35% during the experiments. A breathing hood
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34 113 (Amron International, Vista, CA, #8890 Oxygen Treatment Hood) was used so that the
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36 114 participant in the clothing experiments could breathe clean air from outside the chamber,
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38 115 thus allowing for the separation of dermal from inhalation dose. See Figure S.1 for an image
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40 116 of the participant wearing test clothing and the hood while seated in the experimental
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42 117 chamber. Air concentrations of DEP and DnBP were maintained by continuous emission
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44 118 from aluminum panels (total area of 12 m²) coated with Latex paint. The paint had been
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46 119 formulated with 1% DEP and 10% DnBP (by weight), and was used to deliver these
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48 120 phthalates into chamber air at a relatively constant emission rate.^{8,13}
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3 122 *Clothing and clothing preparation*
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6 123 Clothing was purchased from two different clothing stores in Rolla, Missouri, USA. Each set
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8 124 included a cotton undershirt, a pair of cotton jeans, a long-sleeved cotton tee shirt, cotton
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10 125 underwear and cotton socks. Details such as size, style and manufacturer can be found in
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12 126 Table S.1.
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18 128 Two sets of clothing were prepared by washing all pieces at the same time in a standard
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20 129 clothes washer using unscented detergent. They were then dried in an electric dryer on the
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22 130 “medium” setting and each set was packaged separately in two layers of clean aluminum
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24 131 foil until use. During the first 6-hour exposure period, one set was worn directly from its
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26 132 package and is denoted “fresh”. Another set of clothing was exposed to chamber air for 9
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28 133 days and denoted as “exposed”. This exposure took place in the same chamber, under the
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30 134 same conditions and at the same time as bare-skin dermal uptake experiments occurred;
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32 135 the latter are described in Weschler et al.⁸ The clothing was hung inside-out in the path of
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34 136 fans to improve transfer of phthalates from air to the clothing. The air concentration was
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36 137 measured during days 2 and 3 of the 9 day clothing-exposure interval. During these
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38 138 periods, the average concentrations for DEP were 250 and 233 $\mu\text{g}/\text{m}^3$ and that of DnBP
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40 139 was 123 and 114 $\mu\text{g}/\text{m}^3$.
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49 141 *Preparation of participant*
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51 142 Because there were a limited number of breathing hoods available in the exposure
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53 143 chamber, it was only possible to study one clothed participant. The participant was a 48
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55 144 year old Caucasian male, 192 cm tall weighing 91 kg. The participant followed the same
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3 145 restricted diet and restricted use of personal care products protocol described in Weschler
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6 146 et al.⁸ These restrictions were intended to reduce background metabolites of DEP and
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8 147 DnBP in the participants' urine. In brief, for 12 hours prior to exposure and 54 hours after
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10 148 exposure began, the participant only ate Swedish dried bread and ate thick-rinded fruit
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13 149 such as oranges, bananas and melons. He drank only tap water or tea made from tap water.
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15 150 The participant showered without soaps or detergents 24 hours prior to the experiment
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18 151 and showered without soaps again 48 hours after the beginning of an exposure. The
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20 152 research protocol was approved by the Capital Region of Denmark Committee for Research
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22 153 Ethics. The participant provided informed consent before participation and consented to
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25 154 publication of his photo.
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30 156 *Description of exposure periods*

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32 157 The participant participated in two exposure experiments. The first took place on a
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34 158 Wednesday coincident with the 2nd set of exposure experiments during the first week
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37 159 described in Weschler et al.⁸ The participant collected two urine samples on the morning of
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39 160 the experiment. Immediately before entering the chamber the participant collected a urine
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41 161 sample, changed into the "fresh" set of experimental clothes, donned a breathing hood and
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43 162 entered the chamber at 11:00. The participant sat at a desk for most of the 6-hour exposure
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45 163 period and left the chamber once briefly to collect a urine sample. At 17:00, the participant
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47 164 left the chamber and changed into his normal clothing. Following this, the participant
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49 165 maintained the restricted diet and personal product restrictions and collected all urine for
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52 166 48 hours. The second exposure experiment took place on a Tuesday coincident with the 3rd
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55 167 set of exposure experiments during the second week described in Weschler et al.⁸ The
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3 168 procedure was identical to the first experiment except that the participant changed into the
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6 169 “exposed” set of clothes before entering the chamber at 10:00 (leaving at 16:00).
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11 171 *Analysis of air and urine*
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13 172 Air concentrations of phthalates were determined by first collecting 6 L samples of air with
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15 173 Tenax-TA filled thermal desorption tubes, and analyzing by thermal desorption followed by
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17 174 gas chromatography using a mass selective detector. Phthalates were quantified using
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20 175 original standards. The concentrations in air and other conditions are tabulated in
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23 176 Weschler et al.⁸
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27 178 Urine samples were weighed on the day of collection and stored in a freezer until they were
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30 179 shipped overnight to the Institute for Prevention and Occupational Medicine of the German
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32 180 Social Accident Insurance in Bochum, Germany. Urine samples were analyzed for mono-
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34 181 ethyl phthalate (MEP), a metabolite of DEP, as well as mono-n-butyl phthalate (MnBP) and
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37 182 3OH-mono-n-butyl phthalate (3OH-MnBP), metabolites of DnBP. The concentrations of
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40 183 these metabolites were determined by two-dimensional high performance liquid
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42 184 chromatography coupled to tandem mass spectrometry (LC/LC-MS/MS) using internal
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44 185 isotope-labeled standards after enzymatic deconjugation of the phthalate metabolites from
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46 186 the glucuronidated form following methods published by Koch et al.^{14,15} Other details of
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49 187 analytical methods can be found in Weschler et al.⁸
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54 189 *Calculations*
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3 190 Calculated total uptake of DEP or DnBP during the 6 hour exposure period was based on
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6 191 methods described by Koch et al.¹⁵⁻¹⁷ and outlined in Weschler et al.⁸ Metabolite
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8 192 concentrations were converted to mass excreted and then converted to parent molecule
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11 193 uptake using predetermined metabolic conversion factors. In the “fresh clothes”
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13 194 experiment there was very low overall dermal uptake of DEP and DnBP (see Results and
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15 195 Discussion). To better quantify uncertainty in this case, background uptake has been
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18 196 determined in a somewhat different manner than in Weschler et al.⁸ For the present
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20 197 participant, little residual uptake from the 6-hour experiment remained, relative to
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23 198 background uptake, for the last four urinations of the fresh clothes experiment (collected
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25 199 from 40.5 to 50.0 hours after exiting the chamber). Therefore, the average dose rate (total
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27 200 dose/elapsed time) from these samples was subtracted from the dose rate calculated for
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30 201 each post-exposure sample for both fresh and exposed clothes experiments. This was
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32 202 multiplied by the sample time interval, and the result from each interval summed, to
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35 203 determine the background-corrected total dose. Dermal uptake was also corrected for DEP
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37 204 and DnBP measured in the breathing hood air ($40.7 \mu\text{g}/\text{m}^3$ and $5.7 \mu\text{g}/\text{m}^3$, respectively)
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39 205 using a breathing rate of $0.7 \text{ m}^3/\text{h}$. Dermal uptake was then normalized by the air
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42 206 concentration during the 6 hours in the chamber and the participant’s weight. Also
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44 207 reported is the average flux for the 6-h exposure, corrected for background uptake and
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47 208 hood air inhalation. Exposed surface area is taken as 2.06 m^2 , estimated by equation 7A-7
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49 209 of the Exposure Factors Handbook¹⁸ and corrected for the area of the head (6.6% of total).
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51
52 210 To compare the rate of uptake among exposure conditions and between phthalates, we
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54 211 calculated a normalized metabolite excretion rate. First we calculated the slope of net
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56 212 metabolite vs time from initial sample (after exposure begins) to the last sample that
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3 213 includes no more than 75% of total net metabolite excreted. This slope divided by the total
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6 214 mass excreted was defined as the normalized metabolite excretion rate. See Table S.2 for
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8 215 additional details regarding the calculation methods.
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11 12 13 217 **Results and Discussion**

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15 218 Major quantitative results are shown in Table 1. There is a striking difference between
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18 219 results for fresh and exposed clothing experiments. The net metabolites excreted over the
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20 220 54 hour period after initiation of exposure are far higher for exposed clothes than for fresh
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22 221 clothes, indicating that parent compound uptake is much higher for exposed than for fresh
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25 222 clothes. When corrected for background uptake rate and inhalation from the breathing
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27 223 hood and normalized by body mass and air concentration, wearing exposed clothes
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29 224 resulted in DEP and DnBP uptakes that were 11 and 36 times greater, respectively, than
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31 225 when wearing fresh clothes. The mass of metabolites excreted over the first 24 hours by
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33 226 the volunteer wearing exposed clothes (3.6 mg MEP; 2.1 mg MnBP) approaches that due to
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35 227 application of a 2% DEP/DnBP cream over most of the skin of subjects as reported by
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37 228 Janjua et al.¹⁹ (MEP range 2.5-85 mg; MnBP range 3.6-18 mg).
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44 230 The ratio of the normalized uptake of DEP/DnBP was very different for the two scenarios.
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46 231 For exposed clothing, normalized uptake of DEP is somewhat smaller than for DnBP
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48 232 (DEP/DnBP = 0.7), but for fresh clothing it is much higher (DEP/DnBP = 2.3). This suggests
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50 233 that fresh clothes retard uptake of DnBP more than DEP and/or that exposed clothes
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52 234 enhance uptake of DnBP relative to DEP. Both mechanisms are consistent with a higher
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54 235 cloth/air partition coefficient for DnBP, which has a higher molecular weight than DEP.
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6 237 Normalized dermal uptake for both fresh and exposed clothes differ substantially from
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8 238 uptake to bare skin as described in Weschler et al.⁸ For comparison, results for bare skin
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10 239 and clothing experiments are shown in Figure 1. For exposed clothes, normalized uptake of
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12 240 DEP and DnBP are 3.3 and 6.5 times greater, respectively, than the average of bare skin
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14 241 results and 1.9 and 3.9 times higher than the highest uptake observed in the bare skin
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16 242 experiments. For fresh clothes, uptake is 3.2 and 5.6 times lower than the average for bare
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18 243 skin experiments. Based on a t-test, the probability, p , of the results stemming from random
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20 244 variation was $<10^{-4}$ for exposed clothes and <0.017 for fresh clothes. These findings are
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22 245 consistent with the hypothesis that fresh clothes retard uptake and exposed clothes
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24 246 increase uptake compared with bare skin.
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32 248 A comparison of the results, accounting for participant age, is also enlightening. Weschler
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34 249 et al.⁸ observed a striking relationship between dermal uptake and age. Shown in Figure 2a
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36 250 and 2b are plots of net amounts of MEP and MnBP excreted, from the time exposure began
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38 251 until the end of urine sampling, for the two clothing experiments (worn by a 48 year-old
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40 252 participant) and bare skin results from the 47 year-old participant reported by Weschler et
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42 253 al.⁸ For both phthalates, wearing exposed clothing increased the excretion rate and net
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44 254 excretion by a large margin. Wearing fresh clothes significantly reduced excretion rate and
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46 255 net excretion.
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54 257 In Figure 3, normalized clothing results for uptake of parent compounds are plotted against
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56 258 age along with the normalized results for all six bare-skinned participants. The clothing
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3 259 results are clearly “off the line”; the exposed clothes result in much higher uptake and fresh
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6 260 clothes much lower uptake than for bare skin. Again, using the 47 year-old participant from
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8 261 Weschler et al.⁸ as the best comparator, we observe that exposed clothes resulted in 2.3
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10 262 and 3.4 times more uptake of DEP and DnBP respectively. Fresh clothes resulted in 4.5 and
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12 263 11 times lower uptake.

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18 265 The excretion rate of metabolites differs among conditions and between phthalate
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20 266 metabolites. The difference between MnBP and MEP is apparent for exposed clothes in
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22 267 Figure 2, with MEP rising faster than MnBP. The normalized excretion rate for both
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25 268 conditions studied in this research and for the six bare skin participants is shown in Figure
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27 269 4. To make the comparison more clear, the bare skin results for participants wearing
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30 270 hoods are grouped with the clothed results. Qualitatively, clothed results are similar to
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32 271 bare skin results: the normalized excretion rate of MEP is higher than for MnBP. For both
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35 272 MEP and MnBP, the excretion rate is higher for exposed clothes than for fresh clothes. This
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37 273 is consistent with the hypothesis that fresh clothes act as a barrier and delay transport
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40 274 from air to skin. The difference is more pronounced for MnBP than for MEP, possibly due to
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42 275 stronger sorption of DnBP to clothing.

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47 277 The results support the hypotheses that 1) fresh clothes are protective, reducing uptake of
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49 278 DEP and DnBP compared with bare-skinned participants and 2) exposed clothes increase
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51 279 uptake. Although only one participant was tested (in two exposure periods), we believe the
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54 280 results are compelling, especially when compared with the narrow range of results for six
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56 281 bare-skinned participants. All results are significantly different from the six bare-skinned
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3 282 participant results. When compared by age, the difference is even more apparent (Figure
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6 283 3). However, replication of these results with a larger number of participants will be
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8 284 valuable.
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13 286 For both DEP and DnBP, the dose while wearing fresh clothes is small and could have come
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15 287 from a combination of penetration through clothing and absorption by bare skin. The
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17 288 participant in this study was not completely clothed: the hands were bare. We can estimate
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19 289 the absorbed dose by hands assuming that hands are 4.7% of an average adult male's total
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21 290 surface area.¹⁸ We will use participant 2, the bare skinned participant closest in age to the
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23 291 clothed participant, for comparison and will assume that the shorts worn by participant 2
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25 292 covered approximately 5% of his total surface area. Correcting for the reduced total
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27 293 exposed area due to hood (3.9% of total surface area) and shorts, the normalized dermal
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29 294 uptake due to exposed hands for participant 2 would be approximately 0.004 and 0.003
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31 295 $\mu\text{g}/\text{kg} / (\mu\text{g}/\text{m}^3)$ for DEP and DnBP respectively. These values can be compared with 0.017
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33 296 and 0.007 $\mu\text{g}/\text{kg} / (\mu\text{g}/\text{m}^3)$ for DEP and DnBP for the fresh clothes experiment. Hence, for
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35 297 DnBP, uptake by bare hands could represent a substantial fraction of total uptake from the
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37 298 fresh clothing experiment. It is also interesting to note that the estimated DEP uptake by
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39 299 bare hands accounts for only 25% of the observed uptake; therefore, penetration through
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41 300 clothing may account for much of the uptake.
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51 302 It is perhaps intuitive that fresh clothes should impede transfer from air to skin of airborne
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53 303 contaminants. Clothing has been designed to protect workers from pesticide spray and
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55 304 industrial toxic gases. A recent paper reported on the ability of "every-day" clothing²⁰ to
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3 305 reduce in vitro dermal penetration of chlorpyrifos from solution. But early human subject
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6 306 studies of VOCs showed little influence of clothing on dermal absorption of nitrobenzene
7
8 307 and phenol.^{3,4} This could be because sorption to cloth is weak for these low molecular
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10 308 weight compounds. Higher molecular weight, low volatility organic compounds are known
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12 309 to exhibit substantial air-fabric partitioning.^{21,22} As volatility decreases, partitioning from
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14 310 air to fabric increases and we would anticipate that retardation of transport across fabric
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16 311 would also increase. Indeed, in this research we observed a lower normalized uptake of
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18 312 DnBP relative to DEP, consistent with the roughly 25 times lower vapor pressure of DnBP.
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24 314 Given the complicated geometry of fabric and skin, and the potential for air movement
25
26 315 through and under fabric, the data cannot be used to test more detailed models, to generate
27
28 316 exposure estimates or to identify compound/fabric combinations that would be most
29
30 317 protective or hazardous. Qualitatively, the transport of SVOCs into and out of clothing may
31
32 318 be described well by a model of transport of contaminants through porous media.²³ In the
33
34 319 context of this model, both advection and diffusion of contaminants through fabric would
35
36 320 be retarded by sorption. Key parameters influencing transport and dermal uptake are
37
38 321 likely to include the geometry and permeability of the fabric, how closely clothing fits, the
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40 322 air-to-cloth partition coefficient, the dermal permeability of the contaminant, the elapsed
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42 323 time the cloth is exposed to contaminated air after washing and the elapsed time clothing is
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44 324 worn.
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54 326 **Geometry and permeability of fabric.** Transport of air and moisture through fabric has
55
56 327 been extensively measured and modeled.²⁴⁻²⁷ Hydraulic permeability lumps geometric
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3 328 complexity of fabric into a parameter that characterizes fabric resistance to advective flux;
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6 329 hydraulic permeability is defined as the volume flux of air due to a specific pressure
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8 330 difference across the fabric in units of $\text{cm}^3/(\text{cm}^2 \text{ s})$ (usually at a pressure difference equal
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10
11 331 to 125 Pa). Hydraulic permeability can range over several orders of magnitude: very low
12
13 332 ($<0.1 \text{ cm}^3/(\text{cm}^2 \text{ s})$) for dense or sealed materials and very high for loosely woven thin
14
15 333 fabrics ($>300 \text{ cm}^3/(\text{cm}^2 \text{ s})$). SVOC transport is likely to be more influenced by advection in
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17
18 334 loosely woven materials with a high hydraulic permeability; for tightly woven materials,
19
20 335 diffusive transport is expected to dominate. For intermediate materials, the relative
21
22
23 336 contributions of diffusion and advection will be influenced by pressure gradients, wind and
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25 337 movement.

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27 338
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29
30 339 **How close clothing fits.** Some sorbed SVOCs may transfer from cloth to skin by contact.
31
32 340 However, since most of the surface area available for adsorption in a woven fabric is
33
34 341 internal, only a small fraction of the sorbed SVOC is likely a consequence of transfer by
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36
37 342 direct contact with the outer fabric fibers. Instead, we believe that the more important
38
39 343 mechanism is desorption from fiber surfaces and diffusion across a thin air gap to skin. For
40
41
42 344 diffusion across a quiescent air gap, flux is proportional to the reciprocal of the air gap
43
44 345 distance. The air gap distance was not measured in this study but we estimate it ranged
45
46 346 from $<0.1 \text{ cm}$ to 0.5 cm . By comparison, a typical bare-skin concentration boundary layer is
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48
49 347 about 0.2 to 0.4 cm , which can be estimated by dividing the gas diffusivity of the SVOC
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51 348 ($0.056 \text{ cm}^2/\text{s}$)²⁸ by an air-to-skin deposition velocity (0.14 - 0.28 cm/s)²⁹. Therefore, the
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53
54 349 initial flux from fabric to skin could be smaller, or more than 4 times greater, than from
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56 350 bulk air when wearing equilibrated clothing. Notably, this estimate overlaps the observed
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3 351 ratio of uptake for exposed clothing to the average for bare skin (3.3 for DEP and 6.5 for
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6 352 DnBP).
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10 354 **Air-to-cloth partitioning.** The sorptive capacity of fabric will influence how it reduces or
11
12 355 enhances transport from air to skin. There is recognition that adsorption and desorption of
13
14 356 indoor-relevant gases on fabrics for tobacco smoke products³⁰⁻³² and pesticides²¹ can
15
16 357 influence exposure. Several studies have shown that dry-cleaning solvents³³⁻³⁶ and moth
17
18 358 repellants³⁷ can sorb to clothing and subsequently desorb, increasing indoor
19
20 359 concentrations. Specialty fabrics have been developed that sorb or react with chemical
21
22 360 warfare agents or pesticides to protect the wearer.³⁸ However, we have only identified two
23
24 361 papers^{22,39} that report equilibrium partition coefficients for an indoor air contaminant and
25
26 362 commonly worn fabrics. In one paper²² the investigators measured equilibrium partition
27
28 363 coefficients for airborne free-base methamphetamine and fabrics including cotton and
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30 364 polyester. The partition coefficients were high enough that mouthing of these fabrics was
31
32 365 predicted to be the primary route of exposure for toddlers, similar to the observation by
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34 366 Gurunathan et al.²¹ for chlorpyrifos and plush toys.
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44 368 **Dermal permeability.** We anticipate that the compounds that are most likely to exhibit
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46 369 enhanced dermal uptake from exposed clothes are those that have high dermal
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48 370 permeability coupled with gas-to-fabric partition coefficients in an intermediate range (not
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50 371 too high, not too low). Uptake of compounds with low dermal permeability is limited by
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52 372 resistance across skin; modest changes in mass-transfer conditions external to skin will
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54 373 likely have little impact on overall uptake. Dermal permeability of compounds typical of
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3 374 indoor air have been estimated by Weschler and Nazaroff.⁶ They identified more than 30
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5 375 common indoor pollutants that are predicted to have high dermal uptake relative to
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8 376 inhalation uptake. If a compound has too high a gas-to-fabric partition coefficient, this will
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10 377 retard transfer from the fabric to skin. On the other hand, if a compound has too small a
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12 378 gas-to-fabric partition coefficient, then exposed clothes have sorbed very little of the
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14 379 compound and there will be concomitantly little enhancement. It is in the intermediate
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16 380 range of gas-fabric partitioning that sorption to clothes prior to wear will have the greatest
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18 381 enhancement on uptake.
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25 383 **Elapsed time clothing sorbs contaminants and time clothing is worn.** It takes time for
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27 384 fabric to adsorb airborne contaminants and approach equilibrium with gas phase
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29 385 concentrations. It also takes time for contaminants to desorb and transfer to skin. As an
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31 386 example, consider a tight-fitting shirt that has been washed, stored in the presence of a
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33 387 contaminant (in air) and then worn. If we assume that the characteristic time for the fabric
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35 388 to equilibrate (τ_e) is independent of the air concentration, then we can qualitatively
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37 389 compare this time to the actual time stored in the presence of a contaminant (t_s) or the time
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39 390 clothing is worn after storage (t_w). *Scenario 1: $t_s < \tau_e$.* For this scenario, the fabric has not
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41 391 equilibrated with the contaminant concentration in the air and may in fact continue to sorb
42
43 392 contaminants even while worn. Regardless of the chemical, enhanced flux from cloth to
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45 393 skin will be limited. *Scenario 2: $t_s \geq \tau_e$ and $t_w < \tau_e$.* For this scenario the fabric is well
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47 394 equilibrated with a contaminant before wearing, but the time worn is short relative to the
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49 395 time it takes for the contaminant to reach a new steady-state. During the time worn, flux to
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51 396 skin will be enhanced but the mass adsorbed to fabric will not change substantially. This
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3 397 scenario could be represented by a shirt worn for a short time that had adsorbed a
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6 398 relatively more volatile, low partition coefficient chemical; it could also be represented by a
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8 399 shirt worn for a longer period of time that had adsorbed a less volatile, higher partition
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10 400 coefficient chemical. *Scenario 3*: $t_s \geq \tau_e$ and $t_w > \tau_e$. Here, the shirt has been worn long enough
11 401 that a substantial fraction of the contaminant has desorbed. While the initial flux to skin
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13 402 may be high, the time-averaged flux will be lower than for *Scenario 2* (all else being equal).
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20 404 Since DnBP is anticipated to have a higher partition coefficient than DEP, we would
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22 405 anticipate that τ_e would be greater for DnBP than DEP. We observe a normalized dermal
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24 406 uptake that is higher for DnBP than DEP from exposed clothes. If the 6-hour period that the
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26
27 407 participant wears the exposed clothing is similar or longer than τ_e for DEP, then it may fall
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30 408 under *Scenario 3*, while DnBP falls under *Scenario 2*.
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35 410 **Conclusions**

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37 411 Clothing acts as a barrier to exposure, but also as a reservoir for recently adsorbed
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39 412 chemicals; the latter can increase dermal uptake. Not only are people subjected to airborne
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41 413 SVOCs while at home, they are also exposed to “home pollutants” outside of their residence
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43 414 when they wear clothing that has been stored in the presence of various SVOCs at home.
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45 415 Given the very large increase in the normalized dermal uptake of DEP and DnBP observed
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48 416 for exposed fabric in this study, we believe clothing-mediated dermal uptake is an under-
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51 417 recognized exposure pathway that could be a substantial or even a dominant exposure
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54 418 route for many chemicals. This is of potential importance in occupational as well as non-
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57 419 occupational settings.
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7 539 **Table 1.** Net amount of metabolites excreted during the time period from entering the chamber
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9 540 until 54 hours later, as well as parent compound uptake calculated from the metabolite levels;
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11 541 details regarding the calculation of the listed values are presented in Supporting Information.

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3 544 **Figure legends.**
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5 545 **Figure 1.** Normalized mass of DEP and DnBP absorbed for fresh and exposed clothes
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8 546 experiments. Also shown for comparison are results from the 6 bare-skinned participants
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10 547 (boxplot) reported in Weschler et al.⁸ The line within the box represents the median; the
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12 548 bottom and top of the box, the 25th and 75th percentiles; the upper and lower whiskers, the
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14 549 10th and 90th percentiles.
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18 550 **Figure 2.** Net amount of MEP (2a) and MnBP (2b) excreted from beginning of exposure
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20 551 until last urine sample. Results for fresh and exposed clothes are compared against the bare
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22 552 skin results of the closest aged participant in Weschler et al.⁸
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25 553 **Figure 3.** Normalized dermal uptake of DEP and DnBP versus age. Shown are results from
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27 554 this research (clothes) and results for six bare skin participants reported by Weschler et
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29 555 al.⁸
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32 556 **Figure 4.** Normalized metabolite excretion rate for MEP and MnBP. Shown are results from
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34 557 this research (clothes) and results for six bare-skinned participants reported by Weschler
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36 558 et al.⁸ The line within the box represents the median; the bottom and top of the box, the
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38 559 25th and 75th percentiles; the upper and lower whiskers, the 10th and 90th percentiles.
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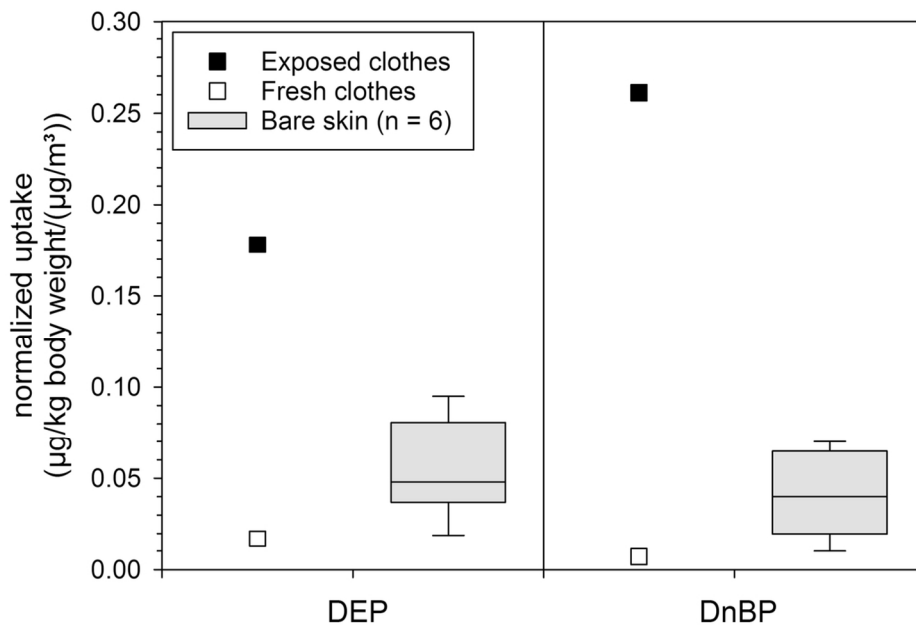


Figure 1. Normalized mass of DEP and DnBP absorbed for fresh and exposed clothes experiments. Also shown for comparison are results from the 6 bare-skinned subjects (boxplot) reported in Weschler et al.⁸ The line within the box represents the median; the bottom and top of the box, the 25th and 75th percentiles; the upper and lower whiskers, the 10th and 90th percentiles.

114x80mm (300 x 300 DPI)

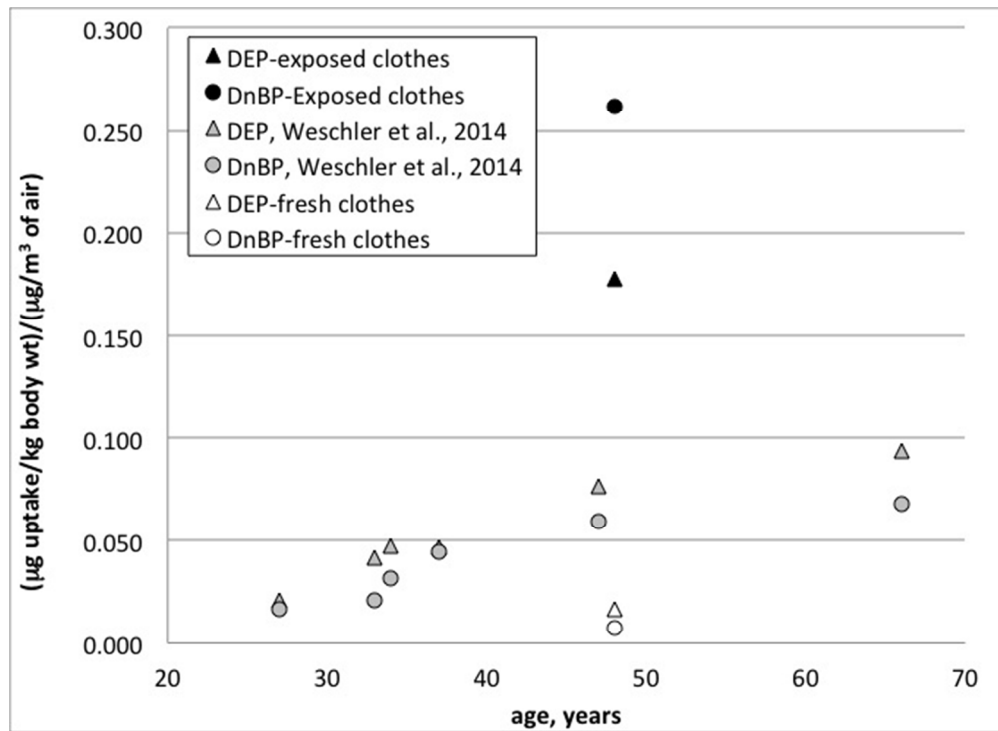


Figure 3. Normalized dermal uptake of DEP and DnBP versus age. Shown are results from this research (clothes) and results for six bare skin subjects reported by Weschler et al.⁸
241x175mm (72 x 72 DPI)

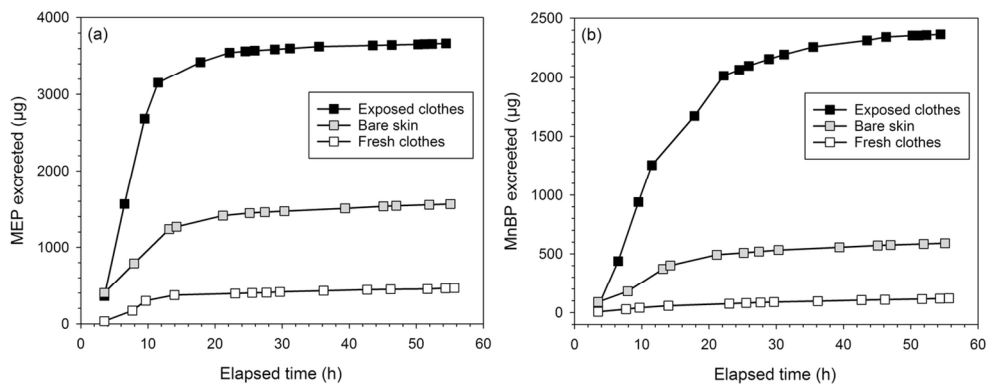
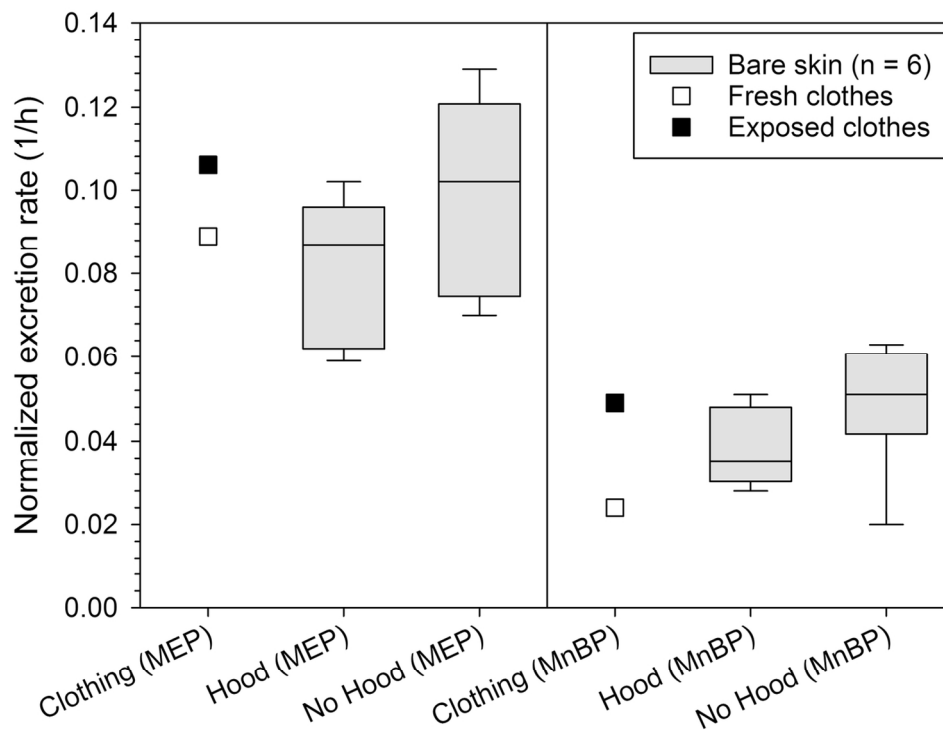


Figure 2. Net amount of MEP (2a) and MnBP (2b) excreted from beginning of exposure until last urine sample. Results for fresh and exposed clothes are compared against the bare skin results of the closest aged subject in Weschler et al.⁸
 121x50mm (300 x 300 DPI)



Normalized metabolite excretion rate for MEP and MnBP. Shown are results from this research (clothes) and results for six bare-skinned subjects reported by Weschler et al.⁸ The line within the box represents the median; the bottom and top of the box, the 25th and 75th percentiles; the upper and lower whiskers, the 10th and 90th percentiles.
129x108mm (300 x 300 DPI)

Only

Table 1. Net amount of metabolites excreted during the time period from entering the chamber until 54 hours later, as well as parent compound uptake calculated from the metabolite levels; details regarding the calculation of the listed values are presented in Supporting Information.

	Metabolites excreted (µg)			Total uptake parent (µg)		Background corrected uptake parent (µg)		Dermal only uptake parent (corrected for concentration in hood) (µg)		Normalized dermal uptake (µg/kg/(µg/m ³))		Average flux based on 6 hour exposure (µg /m ² /h)	
	MEP	MnBP	3OH-MnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP
Fresh clothing	466	121	7.7	634	176	522	98	352	74	0.017	0.007	28	6
Exposed clothing	3666	2367	136	4995	3432	4882	3355	4712	3331	0.178	0.261	381	270

Supplemental Material

Role of clothing in both increasing and decreasing dermal absorption of airborne SVOCs

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Table S1. Clothing characteristics determined from product packaging or labels

Clothing	Composition	Size	Manufacturer	Description
Undershirt	100% cotton	M/M, 38-40" (97-102 cm)	Gildan	Short sleeve, crew neck, color: white Estimated cloth area = 0.91 m ²
Underwear	100% cotton with elastic band	L/G, 36-38" (91-97 cm)	Hanes	Boxer style briefs, color: grey Estimated cloth area = 0.24 m ²
Shirt	100% cotton	M	Gildan	Long sleeve tee-shirt, crew neck, color: dark green Estimated cloth area = 1.03 m ²
Pants	100% cotton	36" (91 cm) waist 36" (91 cm) inseam	Wrangler	Jeans, slim fit, color: dark blue Estimated cloth area = 1.10 m ²
Socks	85% cotton 12% polyester 1% elastic 1% nylon 1% spandex	12W-15	Starter	Tube socks that rise ~20 cm above ankle, color: white Estimated cloth area (pair) = 0.07 m ²

**Figure S1.** Male subject shown wearing full set of test clothing and the breathing hood while seated in the test chamber.

Table S2. Net amount of metabolites excreted during the time period from entering the chamber until 54 hours later, as well as parent compound uptake calculated from the metabolite levels. This Table is identical to Table 1, but is included here with detailed explanations of calculation methods.

	Metabolites excreted (μg) ¹			Total uptake parent (μg) ²		Background corrected uptake parent (μg) ³		Dermal only uptake parent (corrected for concentration in hood) (μg) ⁴		Normalized dermal uptake ($\mu\text{g}/\text{kg}/(\mu\text{g}/\text{m}^3)$) ⁵		Average flux ($\mu\text{g}/\text{m}^2/\text{h}$) ⁶	
	MEP	MnBP	3OH-MnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP	DEP	DnBP
Fresh clothing	466	121	7.7	634	176	522	98	352	74	0.017	0.007	28	6
Exposed clothing	3666	2367	136	4995	3432	4882	3355	4712	3331	0.178	0.261	381	270

1. The mass of metabolites excreted is determined by multiplying the concentration of each metabolite by the volume of urine collected for each sample and summing over all samples collected during the 54 hour period after the exposure started.

2. Total parent uptake is calculated by converting mass from metabolite to parent and using a metabolic conversion factor.

Compound	Abbreviation	CAS-no.	Molecular weight (g/mol)	Metabolic conversion factor
Diethylphthalate	DEP	84-66-2	222.24	NA
Di-n-butylphthalate	DnBP	84-74-2	278.34	NA
Monoethylphthalate	MEP	2306-33-4	194.18	0.84
Mono-n-butylphthalate	MnBP	131-70-4	222.24	0.84
3OH-mono-n-butylphthalate	3OH-MnBP	57074-43-8	238.24	0.07

$$\text{DEP} = [(\text{MEP} / 194.18) * 222.24] / 0.84$$

$$\text{DnBP} = [(\text{MnBP} / 222.24) * 278.34] + [(3\text{OH-MnBP} / 238.24) * 278.34] / (0.84 + 0.07)$$

3. Background-corrected uptake of the parent compound is determined by subtracting out the background concentration of metabolites, integrating the resulting mass, then applying the conversion described in (2) above. Background is defined as the pre-exposure urine concentration.

4. Dermal uptake of parent compounds is calculated by subtracting from background-corrected uptake the inhaled mass of DEP and DnBP based on concentrations in breathing air of the hood (40.7 and $5.7 \mu\text{g}/\text{m}^3$, respectively). Inhalation rate is assumed to be $0.7 \text{ m}^3/\text{h}$. Therefore, the mass subtracted is 170 and $24 \mu\text{g}$ for DEP and DnBP respectively.

5. Normalized uptake is calculated by dividing the dermal uptake by average exposure air concentration and the subject body mass. Average air concentrations during the fresh clothing experiment were $230 \mu\text{g}/\text{m}^3$ DEP and 113

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3 $\mu\text{g}/\text{m}^3$ DnBP. Average air concentrations during the exposed clothing experiment were 291 $\mu\text{g}/\text{m}^3$ DEP and 140
4 $\mu\text{g}/\text{m}^3$ DnBP.
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7 6. The average flux is estimated from the “Dermal only” corrected parent compound uptake, divided by exposed
8 surface area of the participant and the exposure period (6 hours). Exposed surface area is taken as 2.06 m^2 , estimated
9 by equation 7A-7 of the Exposure Factors Handbook¹⁸ and corrected for the area of the head (6.6% of total).
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