



Critical Review

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# **Clothing-mediated exposure to chemicals and particles**

Dusan Licina, Glenn C. Morrison, Gabriel Beko, Charles J. Weschler, and William W. Nazaroff Environ. Sci. Technol., **Just Accepted Manuscript** • DOI: 10.1021/acs.est.9b00272 • Publication Date (Web): 29 Apr 2019 **Downloaded from http://pubs.acs.org on April 30, 2019**

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# **Clothing-mediated exposures to chemicals and particles**

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## **Abstract**

 A growing body of evidence identifies clothing as an important mediator of human exposure to chemicals and particles, which may have public health significance. This paper reviews and critically assesses the state of knowledge regarding how clothing, during wear, influences exposure to molecular chemicals, abiotic particles, and biotic particles, including microbes and allergens. The underlying processes that govern the acquisition, retention and transmission of clothing-associated contaminants and the consequences of these for subsequent exposures are explored. Chemicals of concern have been identified in clothing, including byproducts of their manufacture and chemicals that adhere to clothing during use and care. Analogously, clothing acts as a reservoir for biotic and abiotic particles acquired from occupational and environmental sources. Evidence suggests that while clothing can be protective by acting as a physical or chemical barrier, clothing-mediated exposures can be substantial in certain circumstances and may have adverse health consequences. This complex process is influenced by the type and history of the clothing, the nature of the contaminant and by wear, care and storage practices. Future research efforts are warranted to better quantify, predict and control clothing-related exposures.

TOC Art:



## **1. Introduction**

 Diverse chemicals, particles and microbes are found on clothing. Some are present at the time clothing is purchased, and some are acquired during the care, storage, and use of garments. People spend most of their lives in intimate contact with clothing. They are exposed to the species found on and in their clothing via inhalation, ingestion and dermal absorption (Figure 1). More specifically, humans inhale species that desorb or are released from their clothing, ingest clothing-associated chemicals and particles when clothing materials enter their mouths, and acquire species on their skin from the clothing they wear. Once in the lungs, in the 45 gastrointestinal system or on the skin, chemicals from clothing may be absorbed into the body.<sup>1</sup> As we show in this review, the resulting exposures are influenced by factors inherent to clothing, such as fiber type, weave, morphology, dyeing process, color and chemical treatment (including incorporation of flame retardants, stain repellants, and anti-wrinkle agents). Exposures are also influenced by external factors such as washing, drying, storage, and usage patterns. Clothing-







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 **Figure 1.** Non-dietary routes of human exposure for contaminants of concern: Inhalation, ingestion and dermal absorption.

## **2. Chemical exposures**

## **2.1 Evidence of clothing-associated exposure to chemicals**

2.1.1 Clothing-associated chemicals in skin, blood and urine

Human exposure and uptake of organic compounds by means of transfer from treated fabrics has

- 83 been investigated for several decades. For example, in the late 1970s, Blum et al.<sup>7</sup> reported
- finding metabolites of the flame retardant tris(2,3-dibromopropyl)phosphate (tris) in the urine of
- children who had worn clothing treated with this chemical. Radiolabeled tris in treated and dried
- cloth was shown to penetrate clipped skin of rabbits. Moistening the cloth with simulated sweat
- 87 did not increase absorption.<sup>8</sup> Earlier, Armstrong et al.<sup>9</sup> and Brown<sup>10</sup> reported instances of infant
- poisoning attributable to use of phenolic disinfectants in improperly laundered hospital fabrics.

 Recently, forestry workers wearing permethrin-treated, tick-proof pants were shown to have significantly elevated levels of a permethrin metabolite in their urine.<sup>11</sup> Moreover, absorption of ethylene oxide (a fumigant), glyphosate (an herbicide), malathion (an insecticide) and benzothiazole (used as dye, biocide, herbicide, and fungicide) from fabric into skin or a skin-93 mimicking membrane has been demonstrated in studies using an *in vitro* diffusion cell.<sup>12−14</sup> Measurements of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) in the stratum corneum, epidermis and subcutis of eight volunteers as well as in a variety of new fabric swatches showed that some textiles are contaminated and can be an important source of exposure 97 to these chemicals.<sup>15</sup> The PCDD/F species were shown to diffuse through the stratum corneum into the deeper layers of the skin. Stratum corneum concentrations were substantially higher after wearing contaminated shirts rather than uncontaminated shirts. Skin contamination was heterogeneous, both among individuals and among sites on the same individual. However, when identical, homogeneously contaminated T-shirts were used in a companion study, relatively little 102 spatial and interpersonal variability was observed.<sup>16</sup> Uptake from polyester was found to be an order-of-magnitude lower than from cotton. Wearing fabrics that were previously worn enhanced transfer. Residual sweat and lipid compounds may have served as transfer vehicles, or possibly weakened the binding interaction between the fabric and PCDD/F. Heavy perspiration during intense physical activity also increased the migration rate of a textile dye, Dianix®, onto the skin 107 of volunteer subjects, while contact time was found to be less important.<sup>17</sup> Clothing can act as a means of transporting pollutants from one environment to another. This phenomenon has been studied in the context of health concerns related to "para-occupational"

- exposures. Certain hazardous chemicals, such as lead, beryllium, polychlorinated biphenyls
- (PCB) and pesticides, can be transferred from a work site to the worker's home via clothing and

 thereby contribute to elevated levels in the blood and urine, or even to direct adverse health effects.<sup>18</sup>−<sup>20</sup> For example, women who laundered agricultural work clothes had up to 42% higher serum levels of dichlorodiphenyltrichloroethane (DDT) and hexachlorobenzene compared to 115 women who did not.<sup>21,22</sup> Similarly, women living in homes in which agricultural workers wore their work clothes had higher levels of most of the organochlorine pesticides that were being 117 used.<sup>22</sup> Multivariate analyses by Park et al.<sup>23</sup> indicated an association between serum levels of polybrominated diphenyl ethers in California firefighters and the storage and cleaning practices used for protective gear. The authors suggested that these flame retardants can be transported to fire stations via fireborne dust on soiled turnout gear and that good housekeeping practices can reduce subsequent exposure. (See also §3.1.4.)

### 122 2.1.2 Influence of clothing on dermal uptake of airborne chemicals

 Until recently, the influence of clothing on dermal uptake of airborne organic compounds received relatively little attention. Initial studies examined a few chemicals, primarily volatile 125 organic compounds in occupational settings. Piotrowski<sup>24</sup> found that clothing reduced dermal uptake of airborne nitrobenzene by about 20-30%, but that clothing had no observable effect on 127 phenol absorption.<sup>25</sup> Recent efforts have addressed dermal exposures to semivolatile organic 128 compounds common in everyday indoor settings. For example, Morrison et al.<sup>26</sup> measured the uptake of two airborne phthalates, diethyl phthalate (DEP) and di-n-butyl phthalate (DnBP), by an individual wearing either clean clothes or clothes previously air-exposed in a chamber with elevated phthalate concentrations. When compared with dermal uptake for bare-skinned individuals under otherwise identical experimental conditions,<sup>27</sup> clean clothes decreased transdermal uptake by factors of 3–6, whereas previously exposed clothes increased dermal uptake by factors of 3 and 6 for DEP and DnBP, respectively. Analogous results were obtained





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 **Figure 2.** Dynamic urinary excretion rates due to dermal uptake for nicotine and two urinary metabolites (summed) after exposing participants, who were wearing breathing hoods, to 151 airborne nicotine.<sup>29</sup> (a) Freshly laundered clothing, unexposed to nicotine, is protective; this is analogous to a tree at the beginning of a rainstorm that protects a person from getting wet. (b) Clothing that has been previously exposed to airborne nicotine dramatically increases urinary excretion rate for days after wearing the clothing, just as a standing under tree after a long rainstorm is most certain to get the person wet. This exposure occurs while wearing the clothes in the environment containing the contaminant (third tree from left) and can continue after leaving this environment.

2.1.3 Health effects as evidence of exposure

Studies of health effects related to hazardous substances in textiles further suggest clothing-

- associated exposures. These works have mainly focused on dermatitis caused by textile dyes and
- finishing resins.<sup>36</sup>−<sup>41</sup> A limited literature also exists on carcinogenic, mutagenic and reprotoxic
- substances in textile articles. These effects have been suggested for certain dyes, especially azo
- dyes<sup>42</sup>−<sup>45</sup> and for some antibacterial agents, such as triclosan.46,47 Brominated flame retardants,
- phthalates and degradation products of highly fluorinated polymeric water repellents and stain

 repellents, which can be present in textile articles, have been associated with reproductive and developmental toxicity.<sup>48</sup>−<sup>52</sup> Evidence of direct health effects of such clothing-related exposures is lacking. Comprehensive reviews of textile-related health studies can be found in the Swedish 169 Chemicals Agency's report<sup>3</sup> and in the opinion statement of the German Federal Institute for Risk Assessment.<sup>2</sup>

#### **2.2 Occurrence, persistence and accumulation of chemicals in clothing**

The chemicals present in clothing are a mix of those present at the time of purchase (possibly

attenuating with time) and those acquired post-purchase. This mix changes with cleaning

practices, storage and wear.

#### 2.2.1 Chemicals present at time of purchase

 Most of the chemicals that have been measured in clothing at the time of purchase are a consequence of manufacturing processes (e.g., dyeing, bleaching, finishing) or have been deliberately added and are intended to be retained during the life of the garment. The latter group, referred to as "auxiliaries," includes antiwrinkling resins, flame retardants, antimicrobial agents, pesticides, surfactants and other coating chemicals. Dyeing involves the largest range of 181 chemicals, with an estimated 800 dyes currently in use.<sup>2</sup> A move towards more environmentally 182 benign textile dyeing is altering the mix of chemicals used in dyeing.<sup>53</sup> Some chemicals in clothing fabrics are present as a consequence of packaging, transport, storage and other processes that occur between manufacture and purchase.

Chemicals that have been identified on newly purchased clothing include trace elements such as

heavy metals;54−62 residual aromatic amines associated with certain azo dyes;63,64 quinoline and

substituted quinolines;65−69 alkylphenol ethoxylates, alkylphenols, bisphenols and



2.2.2 Chemicals acquired post-purchase

 Chemicals present in air, especially indoor air, can also be present on clothing exposed to that air.30,88−95 A commonly encountered example of chemical uptake from air occurs when clothing is exposed to environmental tobacco smoke (ETS). Up to a milligram of nicotine can be sorbed 206 by a square meter of cotton fabric during just a few hours of exposure.<sup>96</sup> Odors derived from ETS constituents can linger on clothing for hours to days. More generally, how much or how little of a chemical is transferred from air to clothing depends on several factors. One key factor is the 209 partition coefficient between clothing and air  $(K_{ca})$  for the fabric in question. As a rule of thumb, the more an airborne chemical resembles the chemical nature of the fabric that constitutes the



 chemical as well as with the cleaning practices, including the detergent or dry-cleaning solvent 231 that is employed. Gong et al.<sup>31</sup> found that the efficiency with which machine washing removed

fraction of a chemical that is removed from clothing during cleaning varies with the nature of the

232 phthalates from cotton jeans increased with the octanol/water partition coefficient  $(K_{ow})$  of the

 phthalate. During dry-cleaning, clothing can retain chemicals from cleaning solvents that 234 subsequently contribute to personal exposures.<sup>102,103</sup> During laundering, clothing may acquire 235 scents (e.g., synthetic musks<sup>2</sup>) and other detergent constituents (e.g., alkylphenol ethoxylates<sup>104</sup>). Following the wash cycle, clothing is either air-dried or mechanically dried. When air-dried, the clothing can sorb chemicals from the air in which it is dried. When mechanically dried, some chemicals can be thermally desorbed while other chemicals (e.g., fabric softeners introduced using 'dryer sheets') may be sorbed by the clothing. Laundering also results in chemicals being transferred among the differing items that are washed or dried together. Cross-contamination of 241 fabrics during laundering and storage has been reported for permethrin-treated garments.<sup>105</sup>

 Chemicals on clothing can be chemically transformed to other species. Of longstanding concern are the abiotic and microbial reduction of azo dyes to carcinogenic aromatic amines such as 244 aniline, benzidine, and 2-naphthylamine.<sup>106−111</sup> For example, analysis of 86 textile products purchased in Japan detected aromatic amines at low concentrations in socks, undershorts, pants 246 and other garments.<sup>64</sup> Oxidants can also degrade azo dyes, as shown by reactions initiated by the hydroxy radical, generating benzene and substituted benzenes.<sup>112</sup> Photolytic debromination has 248 been shown to produce low levels of polybrominated dibenzofurans<sup>113</sup> when clothing containing the flame retardant hexabromocyclodecane (HBCD) is dried in the sun.

250 During wear, clothing acquires skin oils, whose constituents can be altered via microbial activity. 251 Different fiber types promote the growth of different microbes, influencing malodor generation 252 from microbial metabolism of apocrine and sebaceous secretions.114,115 Squalene, a major 253 constituent of skin oil,<sup>116</sup> has been shown to react with ozone on T-shirts generating products 254 with a range of volatilities.<sup>117−119</sup> The less volatile products remain on the apparel item, exposing 255 the wearer to species such as  $C_{27}$ -pentaenal,  $C_{22}$ -tetraenal,  $C_{17}$ -trienal and their carboxylic acid

 counterparts.<sup>120</sup> Squalene also reacts with HOCl, the active ingredient in chlorine bleach, to generate chlorinated squalene products. Three to four chlorine atoms become covalently 258 incorporated into the squalene molecule during a one-hour exposure to 1 ppb HOCl.<sup>121</sup> Such species may not be fully removed from clothing during washing. More generally, bleach oxidizes chemicals on clothing, increasing the water solubility of the contaminants but perhaps leaving behind oxidized and chlorinated residues. Numerous low volatility oxidation products, starting with primary carbonyls and carboxylic acids and evolving to products with high O to C ratios, result when ozone reacts with terpenes or sesquiterpenes transferred to clothing from personal care products. Other examples of chemicals generated via reactions that occur on clothing include nonylphenol, a known endocrine disruptor, from the degradation of nonylphenol 266 ethoxylate detergent residues<sup>65</sup> and formaldehyde from urea–formaldehyde and melamine/formaldehyde resins used as antiwrinkling agents.40,41,86 The potential for chemical transformations to occur on clothing is commonly overlooked during assessments of exposures to environmental chemicals.

#### **2.3 Mechanisms, quantification and prediction of exposure and transfer of chemicals**

 Clothing influences chemical exposure by a variety of mechanisms, including some that are complex and poorly characterized. Organizations such as the US Environmental Protection Agency, the World Health Organization and the European Chemicals Agency provide guidance 274 on estimating exposure from consumer articles;<sup>122−124</sup> however, such recommendations are based on a far-from-complete understanding and are therefore of limited utility in accurately characterizing complex chemical exposures mediated by clothing. Notwithstanding their limitations, these recommendations and models can be combined with stochastic representations 278 of exposure factors and behaviors to estimate population distributions of exposure.<sup>125</sup>

### 2.3.1 Dermal transfer and absorption

 Most exposure models of skin contact transfer of chemicals from surfaces are conservative by design, i.e. they account, realistically, for the maximum potential exposure for risk assessment and risk management purposes. Exposure is derived from factors including the skin area in contact, the concentration of the chemical in the material, the number, frequency or duration of contact events, the type of contact and a transfer efficiency.122,124 The transfer efficiency is the fraction of the chemical in the material that transfers during contact events. Experimental 286 measurements of the transfer of pesticides<sup>126</sup> and fluorescent tracers<sup>127</sup> from carpet and of 287 permethrin from military uniforms<sup>128</sup> have been used to quantify transfer efficiency of residues from textiles. Some experimental results used to derive residue transfer efficiency are based on low-volatility chemicals directly applied to the side of the textile in contact with the skin. Therefore, the residue transfer model may inaccurately characterize exposure from clothing that has volatile or semivolatile chemicals distributed throughout the fabric. Recognizing that diffusive migration can occur within consumer materials, it has been proposed that a transfer efficiency can be derived from the amount that can diffuse from a thin "contact layer" of the material. The thickness of this layer can be specified for consumer products or can be estimated 295 if diffusion coefficients are known for specific chemical-material combinations.<sup>129</sup> These models 296 generally do not account for the uptake resistance of skin itself.<sup>130</sup>

 Models of sweat-mediated transfer of chemicals from clothing also use a transfer efficiency approach. The leachable fraction is derived from experiments using artificial sweat to extract 299 substances such as dyes<sup>17,43</sup> and trace elements.<sup>57</sup> (See also §3.1.3.) Often, the extracted fraction is assumed to be entirely transferred to the skin. Such an approach is likely to overestimate exposures, since only a fraction of the sweat will return to the skin from clothing. For example,

 Meinke et al.<sup>17</sup> extracted fluorescent dyes from a polyester/cotton blend shirt using a sweat simulant and compared the predicted exposure (based on 100% transfer) to that observed in volunteers wearing the shirt during 30 minutes of exercise or for 12 hours of normal activity. In these experiments, less than 1% of the estimated amount of a dye was transferred to volunteers during normal wear or sweating.

 Indirect (non-contact) exposure to environmental contaminants can also be influenced by 308 clothing. Clothing has been observed to reduce the transfer of airborne insecticides,<sup>131</sup> phthalate esters<sup>26</sup> and organophosphate flame retardants<sup>33</sup> to skin. Some models of indirect dermal exposure to airborne contaminants have assumed that clothing is fully permeable.<sup>132</sup> Other models assume that clothing is fully impermeable. For example, in estimating dermal uptake of 312 polycyclic aromatic hydrocarbons from barbeque fumes to bare skin, Lao et al.<sup>88</sup> assumed that areas covered by clothing were fully protected. Between these extremes, a mechanistic modeling approach has been introduced that accounts for the history of clothing, contaminant-transfer between clothing and the environment, sorptive partitioning of chemicals to clothing, diffusive and advective transfer through clothing and to skin lipids, as well as resistance to uptake through skin.133,134 The clothing component of these models is similar to that used to assess clothing for 318 chemical protection<sup>135</sup> and can account for uptake through clothing from air as well as exposure to contaminants present in clothing when donned. Predictions using such mechanistic models agree reasonably well with urinary excretion rate measurements for the limited number of human-subject studies in which adequate information is available to populate the model 322 parameters.<sup>30,134,136</sup> These models indicate that clothing can either reduce or increase dermal uptake relative to bare-skin uptake (Figure 2). The extent of exposure is predicted to be sensitive 324 to a chemical's partition coefficient between clothing and air  $(K_{ca})$ ,<sup>89−93,137,138</sup> the efficiency of

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325 chemical removal during laundering, 31,68,92,139 the air-gap between fabric and skin, laundering frequency, and the history of the clothing items prior to wear.<sup>133</sup> A key advantage of dynamic mechanistic models is that they can predict how clothing accumulates chemicals under non- equilibrium conditions. Such models can be used to derive simpler exposure heuristics for 329 classes of chemicals, types of clothing and exposure scenarios for risk assessment purposes.<sup>140</sup>

#### 2.3.2 Inhalation

 Inhalation exposures for clothing-associated chemicals can be modeled using methods similar to those used to estimate inhalation of chemicals emitted by consumer products. For example, the emission rate of dry-cleaning solvents from clothing hung in a closet can be combined with 334 building air-exchange rates to predict indoor air concentrations,<sup>103</sup> which are then used to assess inhalation exposures. Inhalation exposure from the emissions that are generated while wearing an article of clothing may be enhanced owing to the "personal cloud" effect, as described for particles in §3.1.5. For gaseous pollutants, personal-cloud type alterations have been illustrated in climate chamber experiments investigating transport and pollutant distribution in the breathing 339 zone,<sup>141</sup> as well as using computational fluid dynamics to predict breathing zone concentrations of volatile products that result from ozone reactions with the surface of the body and 341 clothing.<sup>142,143</sup> For a seated person under typical indoor conditions, inhalation exposure to volatile ozone reaction products with skin oils was predicted to be up to 2.5 times higher than the corresponding value for room-average concentrations. Predicted exposure to ozone itself was 344 estimated to be 0.6 to 0.9 times the corresponding condition for room-average concentrations.<sup>142</sup> Simulations are currently limited to simple scenarios, such as stationary seated or standing individuals. Experimental validation of personal cloud effects for clothing-associated chemical exposures are lacking.

2.3.3 Ingestion

 Ingestion by mouthing of fabrics can be a significant exposure pathway, especially for young children. Exposure estimation requires information on the extractability of compounds in saliva, the frequency of mouthing clothing and the area of the fabric mouthed. Extractability can be quantified using a broader set of *in vitro* bioavailability methods,<sup>144</sup> which have been applied to 353 determine extractability in saliva simulants of azo dyes<sup>145</sup> and for silver from nanoparticles.<sup>146</sup> For highly water-soluble species, upper bounds on exposure can be established by assuming that the chemical is completely extractable. In an evaluation of indirect exposure to environmental airborne methamphetamine in former residential methamphetamine labs, mouthing of cotton fabric by toddlers was predicted to generate intakes approximately 10 times greater than all other exposure pathways combined.<sup>90</sup>

 A diagramatic summary of these exposure pathways as influenced by physical-chemical properties is shown in Figure 3. Excepting particles and particle-associated chemicals, inhalation requires a chemical to be volatile enough to become airborne. Ingestion is important for more water-soluble chemicals. Most species can be transferred to skin by contact or transfer through the clothing-skin air gap. For both ingestion and transfer to skin surface, the chemical must be of lower volatility to be present in clothing at meaningful concentrations. Transdermal uptake from the skin surface tends to be highest for chemicals with intermediate volatilities and relatively low water-solubilities.



 **Figure 3.** The relative importance of clothing-associated exposure pathways based on a chemical's volatility and water solubility.

## **3. Exposures to particles**

## **3.1 Clothing-associated exposures to biotic and abiotic particles**

Ample evidence from environmental and occupational exposure studies indicate that clothing

can act as an important source of particle-borne agents that contribute to human exposures.

Clothing-associated exposures have been observed for biotic and abiotic particles, with varied

acquisition, retention and release mechanisms, exposure routes and potential health outcomes.

This section consolidates evidence from relevant empirical and field studies in the context of a

- providing an overview of exposure to biotic and abiotic particles associated with clothing.
- 3.1.1 Allergens
- Exposure to allergenic biological particles from clothing has been well-studied, including cat
- 381 allergen (Fel d 1), dog allergen (Can d 1), dust-mite allergen, and pollens. Tovey et al.<sup>147</sup> were
- among the first to identify clothing as a significant source of inhalable allergens. They showed

 that allergenic dust particles can become resuspended directly from clothing by body movement and can travel to the wearer's breathing zone by means of the thermal plume, thus causing increased allergenic exposures. Other studies found that exposures to mite and cat allergens were closely related to the quantity of particle-bound allergen found on wearer's clothing, suggesting that personal clothing could be an important factor influencing both mite and cat allergen exposure.148,149 Evidence of allergen exposure also has been reported for people that are not in 389 direct contact or proximity to any allergenic source.<sup>148,150</sup> These studies identified clothing as an

important indirect exposure vector, transporting particle-borne allergens from one space to

another.

 Much prior evidence concerning clothing-mediated exposures to pet allergens has focused on school environments.151,152 Studies have found that allergens can be transported on children's clothing from homes to schools, including both the cat allergen, Fel d 1,148,153 and the dog 395 allergen, Can f 1.<sup>154</sup> Children without pets can also acquire allergens while in school and 396 subsequently bring them back to their homes.<sup>155</sup> A study focusing on exposure interventions found that the level of airborne cat allergens in schools could be effectively mitigated either by pet ownership prohibition or by using school uniforms.<sup>156</sup> Additional evidence has shown that 399 clothing can be a transport vector for the mite allergens, Der f 1 and Der p  $1,^{154,157}$  and for allergenic pollen.<sup>158</sup>−<sup>160</sup> Taken together, this body of research persuasively documents that clothing can be an important secondary source of allergenic exposures in buildings, including environments that are free of direct allergenic sources.

3.1.2 Pathogenic microbes

 A second category of clothing-related biological particles is pathogenic microorganisms that pose threats for the transmission of infectious diseases. Most research about clothing-associated



 During the past few decades, embedded nanomaterials have emerged as a class of technological innovations for improving certain features of clothing fabrics, such as reducing microbial growth and survival, protecting against ultraviolet radiation, and improving water repellency. To achieve specific targeted functions, prevalent nanostructured clothing additives have included titanium 428 dioxide (TiO<sub>2</sub>), silver, zinc oxide (ZnO), gold, copper, carbon nanotubes, and nanoclays.<sup>185</sup> An



 Overall, there is a need for more research to characterize the influence of antimicrobial agents, including nanoparticles, on microbial diversity in clothing and on the development of microbial resistance over time. Whether the presence of nanomaterials on fabrics in contact with the skin could alter the local skin microbiota remains a key open question.

3.1.4 Para-occupational exposures



3.1.5 Personal cloud

 An enhancement of inhalation exposure to particles beyond the room-average levels may occur for clothing-associated particle releases. This feature, termed the "personal cloud," was introduced for clothing-mediated chemical exposures in §2.3.2.

 There are multiple dimensions to the clothing-associated personal cloud effect. Key determinants involve size-dependent emission rates of particles from clothing, proximity of clothing to the breathing zone, and local air movement in relation to personal activities. Several studies suggest that direct shedding from clothing surfaces may be a noteworthy source of coarse-mode particles 469 and bioaerosols indoors,<sup>198–201</sup> but none of them quantified contributions to the personal-cloud 470 effect. A recent study by Licina et al.<sup>202</sup> reported that clothing movement can release coarse particles into the perihuman space of a seated person, which can then be transported upwards by means of the metabolically induced thermal plume. In that study, the contribution of such

473 releases to the personal cloud was substantial — from 2 to 13  $\mu$ g/m<sup>3</sup> in the particle diameter range 1-10 μm. The contribution of clothing-associated particle release to a personal cloud effect was observed only for seated occupants and specifically not observed during walking. The study suggests that the personal cloud is contingent on physical activities and that manipulating the metabolic thermal plume could alter exposure to clothing-released particles. Additionally, during more intensive clothing manipulations, such as putting on and taking off a shirt, or folding and 479 unfolding a shirt, sharp peaks in the breathing zone  $PM_{10}$  mass concentration were detected, at 480 times exceeding 40  $\mu$ g/m<sup>3</sup>. Overall, the emerging evidence regarding the personal cloud combined with evidence that clothing can harbor allergens, potentially pathogenic microorganisms, and other harmful substances suggest that clothing surfaces may be an underappreciated factor influencing particle exposure, possibly with public-health relevance.

### **3.2 Toward quantifying clothing-mediated particle exposures**

 The previous section summarized evidence that clothing-mediated exposures to particles are potentially meaningful in diverse circumstances. It is important to characterize exposures quantitatively and – in as far as it is possible – mechanistically, so that one is able to extract generalizable findings from limited experimental evidence. In this section, we outline a framework that could guide and support systematic knowledge acquisition for better understanding how clothing influences inhalation exposures to biotic and abiotic particles.

 The central element in this framework is the determination of size-resolved and composition specific emission rates of particles associated with clothing. Such emission rates can be expressed in terms of particle mass per time or particle number per time. Composition is key in relation to health outcomes of concern: allergenic particles, infectious microbes, and abiotic particles each contribute to increased yet distinct adverse health risks.

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 As with other indoor particle sources, emission rates can be inferred from measuring time- and size-resolved particle concentrations in chamber experiments with controlled environmental conditions (e.g. known ventilation rate and low particle backgrounds) and simulated activities.199,202 It is also plausible to infer emission rates from field observations; however, doing so for clothing-associated particle emissions poses the challenge of separately accounting for 509 resuspension from flooring, commonly an important source of coarse particles indoors.<sup>205</sup>

It is reasonable to expect that particle exposures associated with clothing occur mainly indoors.

 In assessing clothing-associated emissions, it is worthwhile to differentiate broadly among three particle source categories (see Figure 4). One category is skin flakes, known as squames, generated through frictional interaction between clothing and skin. These squames consist of skin fragments with associated microbes, especially bacteria. A second category would be particles endogenous to the clothing fabric, such as fabric fragments and nanoparticle additives. A third category, the broadest, is exogenous particles that become associated with clothing articles by means of environmental transfer. The first category has been studied most carefully in connection with concerns about hospital-acquired infections. Concern about the second category is increasing, in part due to the emerging use of nanoparticle fabric treatments. The third

- 519 category would be relevant for concerns as diverse as allergen exposure, para-occupational
- 520 exposure, and general enhancements of airborne particles via the personal cloud.



522 **Figure 4.** Particle source categories associated with clothing (left); and mechanisms of size 523 dependent particle release and resuspension characteristics (right). Corresponding references: 524 dominant particle size mode reported by Bhangar et al.<sup>199</sup>; size-resolved emission rates from 525 sitting and walking person reported by Licina et al.<sup>202</sup>; release of previously deposited particles 526 from clothing (transport vector effect) reported by Licina and Nazaroff.<sup>206</sup>

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 one expects that important factors affecting emissions would include initial particle loading of the fabric, the nature of bonding with fibers, the nature and intensity of movement generating frictional forces, and the overall wear of the fabric.

 For emissions of exogenous particles, one might envision clothing articles as environmental reservoirs and aim to account for the net movement of particles between these reservoirs and the surroundings. Consider an article of clothing, such as a T-shirt, passing through a cycle starting with laundering. The washing cycle might effectively remove previously deposited particles, but could conceivably add particles from dissolved salts in the wash water and from detergent residue. A tumble-dry cycle could effectively add some airborne exogenous particles filtered by the clothing items from the drying air that passes through the drum. The clothing article might then lose some of these particles, and contribute an increment of exposure, during the post- laundry handling of folding and placing in storage. When worn, the T-shirt can acquire exogenous particles by deposition from the air and by direct contact with particle-laden surfaces. Exogenous particles may also be acquired during storage intervals, especially if exposed in a manner that would be influenced by settling dust. The accumulation of particles during these processes could be quantified through deposition assessments, for example through the multiplicative combination of exposure concentrations of particles, a suitable deposition velocity, and duration of exposure. Knowing the size-resolved and composition-specific quantities of exogenous particles on a clothing article, one could assess the emission rate through the use of loss-rate coefficients. An analogous approach has been used to systematically investigate particle 555 resuspension from walking.<sup>205</sup>

## **3.3 Factors affecting clothing-mediated particle exposures**

 The previous section outlined an approach that could be used to systematically assess clothing- associated exposures to particles. Specific information about relevant factors and processes is sparse. This section describes what is known from prior experimental investigations about the major factors that influence the size-dependent emissions of particles associated with clothing, emphasizing the relationship to inhalation exposures.

 Early studies revealed important findings about clothing-skin surface interaction as a means of liberating bacteria-laden skin flakes.178,179,182−184,208−<sup>211</sup> Recent advances in DNA-based measurements have enabled rapid progress in characterizing the human microbiome, including 565 detailed descriptions of diverse communities of bacteria<sup>212,213</sup> and fungi<sup>214</sup> present on human skin. Analyses of clothing surface samples or air exposed to clothing have revealed populations 567 of pathogenic bacteria, <sup>164, 176</sup> respiratory syncytial virus, <sup>177</sup> fungi, <sup>215, 216</sup> dust-mite and cat allergens, endotoxins157,217 and allergenic pollen.<sup>158</sup>−<sup>160</sup> Some quantitative evidence exists 569 documenting microbial transfer to clothing from skin<sup>210</sup> and by hands,<sup>218,219</sup> although more studies are needed to quantify this phenomenon and to better characterize the process mechanistically.

 Available evidence suggests that the rate of particle release from clothing fibers is influenced by a combination of three main factors: properties of clothing, environmental conditions, and human factors. A dominant factor influencing release is the intensity of movement. Up to an order of magnitude higher emission rates have been observed during vigorous bodily movement compared to slight activity, presumably owing to increased frictional interactions between 577 clothing fibers and skin.<sup>182,199,202,220</sup> Men have been found to release significantly more particles compared to women.184,211,221−<sup>224</sup> Application of skin lotion has been linked to reduced dispersal 579 rate of biotic particles.<sup>201,223</sup> Some studies,<sup>221,225,226</sup> but not all,<sup>223,227</sup> have found that the emission

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rate of biotic particles from clothing-skin interactions increases within an hour after showering.

Transport of particles through clothing surfaces and subsequent dispersal can be reduced by

582 wearing tightly-woven and non-woven fabrics.<sup>179,224,228</sup>

 A few recent studies have applied a material-balance approach to infer size-resolved biotic 584 particle emission rates associated with human occupancy. Qian et al.<sup>229</sup> used quantitative PCR to infer that a single university classroom occupant contributes effective emissions of 37 million bacterial genomes per hour, with a modal aerodynamic diameter of 3–5 µm. However, that study could not differentiate between emissions associated with clothing and those from other sources 588 such as resuspension from a carpeted floor. Bhangar et al.<sup>230</sup> applied a laser-induced fluorescence technique to quantify the per person emission rate of fluorescent biological aerosol particles (FBAP) in the size range 1–15 µm diameter in an uncarpeted university classroom. Their work, which again did not isolate the contribution of clothing, yielded an average emission rate of 2 million FBAP per hour with modal diameters of 3–4 µm. In a subsequent chamber study, 593 Bhangar et al.<sup>199</sup> found that at least 60-70% of occupancy-associated FBAP emissions originated from the floor. However, they also found that "clothing, or its frictional interaction with human skin, was … a source of coarse particles, and especially of the highly fluorescent fraction." That study also revealed a dominant size mode for FBAP of 3-5 µm diameter.

 When considering the specific issue of infectious disease transmission in relation to clothing, the persistence and survival of infectious agents on fabrics needs to be considered. Variation in building environmental conditions and properties of clothing fabrics produce various effects on microbial persistence and survival.231,232 Longitudinal assessment of bacteria survival across different studies showed a remarkably high persistence — from several days up to more than 90 days for isolates of VRE and MRSA.176,233,234 Survival and persistence of viruses and fungi on

 clothing fabrics has similar days- to months-long time scales.215,235 Among different factors influencing survival and persistence, relative humidity and fabric material have been explored. Increased relative humidity (from 35 to 78%) has been linked to reduced stability of both bacterial and viral strains in clothing.235,236 Survival and persistence of bacteria, virus and fungi is higher on commonly used polyester and wool fabrics compared to cotton materials.215,235−<sup>237</sup>

 Another common theme in the literature concerning clothing-mediated exposure to pathogenic microbes considers the effectiveness of laundering practices such as washing, drying and ironing. Mechanical removal includes fabric agitation assisted by surfactant properties of detergents, while inactivation processes can occur as a consequence of elevated water temperature combined with laundry additives such as sodium hypochlorite. Among relevant studies, Callewaert et al.<sup>238</sup> documented microbial exchanges among clothing articles during washing. Nordstrom et al.<sup>239</sup> found that home-washed hospital scrubs had increased prevalence of bacterial species compared to those laundered in hospitals, presumably due to low temperature washing. A 7-log reduction 616 in bacterial load can be achieved by 10-minutes of washing with 60  $^{\circ}$ C water.<sup>240</sup> Adding sodium hypochlorite to a detergent is an effective way to eliminate bacteria and inactivate enteric and respiratory viruses;241,242 however, it might also lead to increases in the abundance of chlorinated organic compounds on clothing.<sup>121</sup> Detergents free of bleach can reduce the prevalence of *Staphyloccus aureus*, <sup>243</sup> while adding bleach-enriched detergents completely eliminates the same. Recent adjustments in laundering procedures include addition of enzymes, reduced water use, lower water temperature and bleach-free detergents.<sup>244</sup>−<sup>246</sup>

Both biotic and abiotic material can be deposited onto clothing surfaces from various

- environmental sources including outdoor air,158,159,247 grassland,<sup>160</sup> residential air,<sup>248</sup> public
- 625 transport microenvironments, and from physical contact with items such as furniture, storage

 surfaces and car seats.<sup>250</sup>−<sup>252</sup> The rate of deposition from air to clothing can be described using the deposition velocity concept.<sup>253</sup> Studies have found that particle size and local air movement are dominant influencing factors.249,254

 Research has clearly documented that previously deposited material can be released into air from clothing.197,206,255,256 For example, using a controlled chamber study approach, Licina and 631 Nazaroff<sup>206</sup> found that 0.3-3% of deposited particles (size range  $1-10 \mu m$ ) deposited through settling could be released via fabric motion. In that work, the release fraction monotonically increased with particle size.

 The degree of particle binding to clothing fibers and the rate of resuspension may arise from a combined influence of different forces acting upon the fibers. The forces governing the release of clothing-embedded particles are abrasive actions between clothing surfaces – a consequence of physical activity of a wearer.182,184 Forces influencing release are strongly linked to particle size. Because detachment forces increase more strongly with particle diameter than do adhesion forces, clothing-associated emissions are more discernible among coarse-mode than fine-mode particles.199–202

 Common clothing fibers are wool, cotton and polyester. Wool has been reported to have particle 642 release rates up to  $10\times$  times higher than the other two materials;<sup>149,257</sup> cotton exhibits higher emissions than polyester.<sup>220</sup> The higher particle emissions from wool garments could be linked to 644 different surface roughness and weave pattern,<sup>258</sup> but also to less frequent laundering as compared to cotton and polyester fabrics.<sup>257</sup> Other clothing conditions found to increase particle 646 release rate include increased clothing age<sup>259</sup> and reduced cleanliness.<sup>1496,257</sup> While it is generally understood that adhesion forces acting on particles increase with relative humidity, we know of

 only two studies that have examined its effect on clothing-associated emissions. Yoon and 649 Brimblecombe<sup>257</sup> found an association between low relative humidity and increased particle 650 emission rate, whereas Zhou et al.<sup>201</sup> reported an insignificant influence.

## **4. Future outlook**

 There is ample evidence that clothing influences human exposure to chemicals and particles. Yet, only a few studies have quantified clothing-mediated exposure by means of direct measurements.7,11,15,24−26,28−30,149,202

 We know surprisingly little about the occurrence of contaminants acquired by everyday clothing after purchase. For a relatively low cost, we could learn a large amount from simply assessing the occurrence, concentrations and extractability (e.g., by sweat and saliva) of chemicals and particles in everyday clothing. Cross-sectional exposure studies would greatly benefit from the addition of clothing analyses, potentially identifying direct connections between clothing-associated exposure and health.

 The diversity of clothing, environmental and human factors make predicting exposures challenging. Therefore, it will be important to reduce the many variables to those that are most influential. Progress can be achieved through models, laboratory and field investigations of human exposure and uptake. In addition to chemical properties, important factors affecting exposures may include textile materials, weave, thickness and permeability; wear, care and storage practices; environmental conditions; intensity and types of activities; skin-oil transfer to clothing and its aging; human physiology (skin integrity, lipid generation, sweating) and personal hygiene habits. Simulated exposures with human subjects also should consider pollutant transfer

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- increasing.<sup>260</sup> Synthetics have chemical partitioning behaviors and moisture holding capacities
- that differ from those of natural fibers, altering the capacity to be reservoirs of contaminants.
- Advances in materials and adjustments in laundering procedures may also influence how
- clothing is cared for and how chemicals and particles are acquired and retained in clothing.
- Increased recycling and re-use of clothing can influence tertiary exposures.
- People spend nearly their entire lives in intimate contact with clothing and other textiles. The
- evidence reviewed in this article supports a view that this environmental compartment plays
- important roles in exposure and health risk. Consequently, clothing as a mediator of chemical
- and particle exposure deserves substantial attention from the environmental science research and
- regulatory communities.

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Figure 1. Non-dietary routes of human exposure for contaminants of concern: Inhalation, ingestion and dermal absorption.



Figure 2. Dynamic urinary excretion rates due to dermal uptake for nicotine and two urinary metabolites (summed) after exposing participants, who were wearing breathing hoods, to airborne nicotine.<sup>29</sup> (a) Freshly laundered clothing, unexposed to nicotine, is protective; this is analogous to a tree at the beginning of a rainstorm that protects a person from getting wet. (b) Clothing that has been previously exposed to airborne nicotine dramatically increases urinary excretion rate for days after wearing the clothing, just as a standing under tree after a long rainstorm is most certain to get the person wet. This exposure occurs while wearing the clothes in the environment containing the contaminant (third tree from left) and can continue after leaving this environment

224x129mm (300 x 300 DPI)



Figure 3. The relative importance of clothing-associated exposure pathways based on a chemical's volatility and water solubility.

133x106mm (300 x 300 DPI)



Figure 4. Particle source categories associated with clothing (left); and mechanisms of size dependent particle release and resuspension characteristics (right). Corresponding references: dominant particle size mode reported by Bhangar et al.<sup>199</sup>; size-resolved emission rates from sitting and walking person reported by Licina et al.<sup>202</sup>; release of previously deposited particles from clothing (transport vector effect) reported by Licina and Nazaroff.<sup>206</sup>