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### ► **To cite this version:**

Maud Pelletier, Nathalie Bonvallot, Olivier Ramalho, Olivier Blanchard, Corinne Mandin, et al.. Dermal absorption of semivolatile organic compounds from the gas phase: Sensitivity of exposure assessment by steady state modeling to key parameters. *Environment International*, 2017, 102, pp.106-113. 10.1016/j.envint.2017.02.005 . hal-01635769

**HAL Id: hal-01635769**

**<https://hal.science/hal-01635769v1>**

Submitted on 15 Nov 2017

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1 **Dermal absorption of semivolatile organic compounds from the gas phase: sensitivity of**  
2 **exposure assessment by steady state modeling to key parameters**

3

4 Maud Pelletier<sup>1,2</sup>, Nathalie Bonvalot<sup>1,2</sup>, Olivier Ramalho<sup>3</sup>, Olivier Blanchard<sup>1,2</sup>, Fabien  
5 Mercier<sup>1,2,4</sup>, Corinne Mandin<sup>2,3,4</sup>, Barbara Le Bot<sup>1,2,4</sup>, Philippe Glorennec<sup>1,2\*</sup>

6

7 <sup>1</sup>EHESP-School of Public Health, Sorbonne Paris Cité, Rennes, France

8 <sup>2</sup>INSERM-U1085, Irset-Research Institute for Environmental and Occupational Health,  
9 Rennes, France

10 <sup>3</sup>University of Paris-Est, Scientific and Technical Center for Building (CSTB), Health and  
11 Comfort Department, French Indoor Air Quality Observatory (OQAI), 84 Avenue Jean  
12 Jaurès, Champs sur Marne, 77447 Marne la Vallée Cedex 2, France

13 <sup>4</sup>LERES-Environment and Health Research Laboratory (Irset and EHESP Technologic  
14 Platform), Rennes, France

15 \*Corresponding author:

16 INSERM-U1085, Irset-Research Institute for Environmental and Occupational Health,  
17 Rennes, France. EHESP-School of Public Health, Sorbonne Paris Cité, Rennes, France.

18 Tel.: +33-2 99 02 26 80.

19 E-mail address: philippe.glorennec@ehesp.fr

## 20 **ABSTRACT**

21 Recent research has demonstrated the importance of dermal exposure for some semivolatile  
22 organic compounds (SVOCs) present in the gas phase of indoor air. Though models for  
23 estimating dermal intake from gaseous SVOCs exist, their predictions can be subject to  
24 variations in input parameters, which can lead to large variation in exposure estimations. In  
25 this sensitivity analysis for a steady state model, we aimed to assess these variations and their  
26 determinants using probabilistic Monte Carlo sampling for 8 SVOCs from different chemical  
27 families: phthalates, bisphenols, polycyclic aromatic hydrocarbons (PAHs),  
28 organophosphorus (OPs), organochlorines (OCs), synthetic musks, polychlorinated biphenyls  
29 (PCBs) and polybromodiphenylethers (PBDEs). Indoor SVOC concentrations were found to  
30 be the most influential parameters. Both Henry's law constant ( $H$ ) and octanol/water partition  
31 coefficient ( $K_{ow}$ ) uncertainty also had significant influence. While exposure media properties  
32 such as volume fraction of organic matter in the particle phase ( $f_{om-part}$ ), particle density ( $\rho_{part}$ ),  
33 concentration ( $[TSP]$ ) and transport coefficient ( $\gamma_d$ ) had a slight influence for some  
34 compounds, human parameters such as body weight ( $W$ ), body surface area ( $A$ ) and daily  
35 exposure ( $t$ ) make a marginal or null contribution to the variance of dermal intake for a given  
36 age group. Inclusion of a parameter sensitivity analysis appears essential to reporting  
37 uncertainties in dermal exposure assessment.

## 38 **KEY WORDS**

39 Percutaneous, indoor air, chemical, contact, sensitivity analysis, Monte Carlo.

## 40 **HIGHLIGHTS**

- 41 • Sensitivity analysis was conducted for 8 SVOCs with MC simulations.
- 42 • SVOC air concentration is the most influential variable parameter.

43 • Octanol-water partition coefficient and Henry's law constant are influential uncertain  
44 parameters.

45 • Influence of airborne particles characteristics and human parameters is minimal.

#### 46 **ABBREVIATIONS**

47 *A* Body surface area ( $m^2$ )

48 *C<sub>a</sub>* SVOC total concentration in indoor air ( $C_g+F$ ) ( $ng/m^3$ )

49 *C<sub>g</sub>* SVOC concentration in the gas phase ( $ng/m^3$ )

50 *DI<sub>dermal-gas</sub>* Daily intake by dermal exposure through the gas phase (micrograms per kilogram  
51 of body weight per day,  $\mu g/kg\text{-}bw/d$ )

52 *F* SVOC concentration in the particulate phase ( $ng/m^3$  of air)

53 *f<sub>om-part</sub>* Volume fraction of organic matter associated with airborne particles

54 *H* Henry's law constant ( $Pa\cdot m^3/mol$ )

55 *K<sub>oa</sub>* Octanol/air partition coefficient

56 *K<sub>ow</sub>* Octanol/water partition coefficient

57 *K<sub>p</sub>* Gas/particle distribution coefficient ( $m^3/\mu g$ )

58 *k<sub>pg</sub>* Indoor air transdermal permeability coefficient, describing the transport of a gas phase  
59 SVOC from bulk indoor air to dermal capillaries, through the boundary layer adjacent  
60 to skin, the stratum corneum and viable epidermis composite ( $m/h$ )

61 *MW* SVOC molecular weight ( $g/mol$ )

62 *P<sub>s</sub>* SVOC vapor pressure ( $Pa$ )

- 63  $R$  Ideal gas constant ( $=8.314 \text{ Pa}\cdot\text{m}^3/\text{mol}\cdot\text{K}$ )
- 64  $\rho_{part}$  Density of airborne particles ( $\text{g}/\text{m}^3$ )
- 65  $SVOC$  Semivolatile organic compound
- 66  $t$  Daily exposure duration (h/d)
- 67  $T$  Temperature (K)
- 68  $[TSP]$  Total suspended particle concentration ( $\mu\text{g}/\text{m}^3$ )
- 69  $W$  Body weight (kg)
- 70  $y_d$  Coefficient describing the external transport of a gas phase SVOC from the bulk  
71 indoor air to the boundary layer adjacent to the skin (m/h)

## 72 INTRODUCTION

73 People spend more than 80% of their time in enclosed spaces, largely in dwellings in which  
74 they are exposed to an increasing number of chemicals from various sources and via different  
75 exposure routes. In addition to other pollutants found in indoor environments (radon, carbon  
76 monoxide, formaldehyde and other volatile organic compounds), semivolatile organic  
77 compounds (SVOCs) have received a great deal of attention, due to a rise in their use in  
78 consumer products as well as improved analytical techniques that have shown their ubiquity  
79 in dwellings (Rudel et al., 2003; Weschler and Nazaroff, 2008).

80 SVOCs include organic molecules from many different chemical families (phthalates,  
81 bisphenols, polycyclic aromatic hydrocarbons (PAHs), organophosphorus (OPs),  
82 organochlorines (OCs), synthetic musks, polychlorinated biphenyls (PCBs),  
83 polybromodiphenylethers (PBDEs), etc.). They are emitted from multiple household sources:  
84 flooring and wall materials, furniture, cosmetics, cleaning products, combustion products,

85 packaging, etc. Due to their physical-chemical properties, they are able to migrate to, and  
86 partition between, different indoor compartments, including gas phase and airborne particles  
87 and settled dust (Weschler and Nazaroff, 2008), as well as other available surfaces such as  
88 walls, ceiling and flooring materials – or human skin and clothing. The scientific  
89 community’s growing interest in studying exposure to these compounds is motivated by  
90 suspicion of reprotoxic, (Rubin, 2011; Moreau-Guigon and Chevreuil, 2014), neurotoxic  
91 (Baldi et al., 2001; Elbaz et al., 2009; Blanc-Lapierre et al., 2012; Zaganas et al., 2013) and  
92 carcinogenic (Armstrong et al., 2004; IARC, 2010a, 2010b) health effects, as well as the  
93 acknowledged presence of these compounds and their metabolites in human biological fluids  
94 (blood and urine) (NHANES, 2015). This interest is even more pronounced with regard to  
95 pregnant women and young children, considered more sensitive to these toxic effects  
96 (Grandjean et al., 2008).

97 At home, in addition to food ingestion (the main exposure pathway for many SVOCs), people  
98 are exposed through a variety of pathways: direct contact with the SVOC source, inhalation  
99 and contact with indoor air (gaseous and particulate phases), ingestion and contact with  
100 settled dust (on floor and furniture). Young children are more exposed than the rest of the  
101 population due to their more frequent contact with the ground and deposited dust, carrying  
102 objects in their mouths, etc. Though dust ingestion and inhalation of gaseous and particulate  
103 phases are the two best-documented exposure pathways in the literature, dermal absorption is  
104 rarely assessed in the course of environmental exposure assessments, because it is presumed  
105 to be negligible. Nevertheless, recent research has hypothesized the significance of dermal  
106 exposure (Weschler and Nazaroff, 2012; Gong et al. 2014) and more recently Weschler et al.  
107 (2015) and Morrison et al. (2016) have corroborated these findings via experimental human  
108 dermal exposure to two gaseous phthalates: diethyl phthalate (DEP) and di-n-butyl phthalate  
109 (DnBP). The results confirm that transdermal uptake directly from air can be a meaningful

110 exposure pathway for DEP and DnBP, and that direct dermal absorption from air is also  
111 expected to be significant for other SVOCs, where the molecular weight and  $K_{ow}$  are in a  
112 similar range. Bekö et al. (2013) estimated daily intakes resulting from four different indoor  
113 exposure pathways: dust ingestion, inhalation and dermal exposure through gas phase and  
114 through dust adhering to skin, based on phthalates' metabolites levels in urine samples of  
115 DEP, DnBP, di(isobutyl) phthalate (DiBP), butyl benzyl phthalate (BBzP) and di(2-  
116 ethylhexyl) phthalate (DEHP) and their concentration in dust samples collected at the same  
117 time. They found that gas phase dermal absorption was the major exposure pathway for the  
118 more volatile compounds, in comparison with the other pathways involved. They also found  
119 that intake through dermal contact with dust contributed only very slightly to total intake for  
120 all studied phthalates. In order to assess SVOC gas phase dermal exposure, some authors have  
121 adapted and used a model based on mass-transfer resistance to calculate an indoor air  
122 transdermal permeability coefficient  $k_{p-g}$  (Weschler and Nazaroff, 2012; Bekö et al., 2013).  
123 This mass-transfer model describes the transport of a gas phase SVOC from bulk indoor air to  
124 dermal capillaries, through the boundary layer adjacent to skin, the stratum corneum and the  
125 viable epidermis composite.

126 At equilibrium and due to their physical-chemical properties, especially  $K_{oa}$ , SVOCs partition  
127 between the gas and particle phases of indoor air (Finizio et al., 1997; Pankow, 1998;  
128 Weschler and Nazaroff, 2008). When assessing gaseous SVOC dermal exposure,  
129 concentration in the gas phase ( $C_g$ ) is required and could be either measured or modeled from  
130 total concentration in indoor air ( $C_a$ ) - which is the sum of gas and particulate (F) phases  
131 ( $C_g+F=C_a$ ). Salthammer and Schripp (2015) have highlighted the importance of taking  
132 parameter uncertainty and variability into account when assessing SVOC partitioning and  
133 exposure. Weschler and Nazaroff (2014) have already assessed the sensitivity of  $k_{p-g}$  and  
134 other partitioning coefficient calculations, such as  $K_p$ , to the octanol/water partition

135 coefficient ( $K_{ow}$ ), the octanol/air partition coefficient ( $K_{oa}$ ) and Henry's law constant (H)  
136 uncertainties. We would like to continue this work here by evaluating the sensitivity of dermal  
137 intake, i.e. the mass of pollutant entering the body per kg of body weight and unit of time, to  
138 these parameters using a steady-state model.

139 The objective of this sensitivity analysis is to evaluate dermal intake variation caused by the  
140 uncertainty and variability of input parameters when using the model described by Weschler  
141 and Nazaroff (2012) for dermal absorption of gas phase SVOCs. We chose the study by  
142 Blanchard et al. (2014), in which 57 indoor SVOCs of health interest (Bonvallet et al., 2010)  
143 were measured with separation of their gas phase and airborne particle concentrations, as a  
144 starting study. We selected eight compounds from different chemical families having varied  
145  $K_{oa}$  and volatility to represent contrasting situations: dimethyl phthalate (DMP),  
146 phenanthrene, galaxolide (HHCB), PCB 105, diazinon, permethrin, bisphenol A (BPA) and  
147 BDE 154.

## 148 **METHODS**

### 149 1. Equation tested for dermal intake modeling

150 For a given human, chronic daily intake of gas phase SVOC via the dermal pathway,  $DI_{dermal-}$   
151  $gas$ , can be estimated in steady-state conditions using the following equation adapted by Bekö  
152 et al. (2013).

$$DI_{dermal-gas} = \frac{C_g \times k_{p-g} \times A \times t}{1000 \times W} \quad (1)$$

153  
154 Where  $C_g$  is the SVOC gas phase concentration ( $ng/m^3$ ),  $A$  is the body surface area ( $m^2$ ),  $t$  is  
155 the daily duration of exposure (h/d),  $W$  is the body weight (kg),  $k_{p-g}$  is the SVOC transdermal  
156 permeability coefficient (m/h) and  $DI_{dermal-gas}$  is expressed in  $\mu g/kg-bw/d$ .



157 The indoor air transdermal permeability coefficient ( $k_{p-g}$ ) can be estimated using the steady-  
 158 state model adapted by Weschler and Nazaroff (2012, 2014), (see supplementary material for  
 159 the intermediate equations used to derive Equation 2):

$$k_{p-g} = 1 / \left( \frac{1}{\gamma_d} + \left( \frac{H}{RT} / \frac{10^{(0.7 \times \log(K_{ow}) - 0.0722 \times MW^{2/3} - 5.252)} \times 3600 \times 10^{-2}}{1 + (10^{(0.7 \times \log(K_{ow}) - 0.0722 \times MW^{2/3} - 5.252)} \times MW^{0.5} \times 3600 / 2.6)} \right) \right) \quad (2)$$

160  
 161 Where  $K_{ow}$  is the SVOC octanol-water partition coefficient, MW is the SVOC molecular  
 162 weight (g/mol), H is the Henry's law constant (Pa.m<sup>3</sup>/mol), R is the ideal gas constant (8.314  
 163 Pa.m<sup>3</sup>/mol.K), T is the air temperature (K) and  $\gamma_d$  is the coefficient that describes the external  
 164 transport of an SVOC from the gas phase in the core of a room through the boundary layer  
 165 adjacent to the skin (m/h).

166 When estimating dermal intake from the gas phase (see Equations 1 and 2), exposure  
 167 assessors may face two situations which can be distinguished in terms of availability of the  $C_g$   
 168 value: A)  $C_g$  is measured experimentally or B)  $C_g$  is calculated from total indoor air  
 169 concentration ( $C_a$ ). Assuming that SVOCs are in equilibrium between gas and particulate  
 170 phases,  $C_g$  can be estimated from  $C_a$  using the partitioning model proposed by Weschler and  
 171 Nazaroff (2010) and can be expressed as follows (see supplementary material for the detailed  
 172 calculation):

$$C_g = \frac{C_a}{1 + ([TSP] \times \frac{f_{om-part} \times K_{ow} \times R \times T}{\rho_{part} \times 10^6 \times H})} \quad (3)$$

173  
 174 Where [TSP] is the total suspended particle concentration (μg/m<sup>3</sup>),  $f_{om-part}$  is the volume  
 175 fraction of organic matter associated with airborne particles and  $\rho_{part}$  is the density of airborne  
 176 particles.

177 2. Parameter estimation

178 The impact of uncertainty or variability of equation parameters on  $DI_{\text{dermal-gas}}$  variability was  
179 assessed. Parameter distributions were constructed or retrieved from the literature as detailed  
180 below. For a given group of occupants in a given indoor setting, some of these parameters  
181 will be the same for all SVOCs ( $\gamma_d$ , [TSP],  $f_{\text{om-part}}$ ,  $\rho_{\text{part}}$ , W, A and t) while others will vary  
182 from one compound to another ( $K_{\text{ow}}$ , H,  $C_g$  and  $C_a$ ).

### 183 *Physical-chemical parameters*

184 For each SVOC, measured or estimated values of  $\log(K_{\text{ow}})$  and Henry's law constant (H) at  
185 25°C were retrieved from: online databases - Hazardous Substances Data Bank (HSBD) and  
186 ChemIDplus (<http://toxnet.nlm.nih.gov/>), Chemspider (<http://www.chemspider.com/>), and  
187 Chemicalize (<http://www.chemicalize.org/>); toxicological and environmental data sheets from  
188 the French National Competence Centre for Industrial Safety and Environmental Protection  
189 (INERIS) (<http://www.ineris.fr/substances/fr/page/21>); online calculators - Chemexper  
190 (<https://www.chemexper.com/>) and ACD/Labs (<http://www.acdlabs.com/>); EPI Suite software  
191 (US EPA, v4.1) and the Handbook of Physical-Chemical Properties and Environment Fate for  
192 Organic Chemicals (Mackay et al., 2010a, 2010b, 2010c, 2010d). Only values at 25°C  
193 (reference temperature) were selected, in order to be consistent and to estimate  $DI_{\text{dermal-gas}}$  at a  
194 constant temperature. For each SVOC, where at least 15 values for their  $\log(K_{\text{ow}})$  and H were  
195 retrieved from the sources mentioned above, distributions were fitted – and otherwise we used  
196 triangular distributions (between minimum, average and maximum values). Corresponding  
197 distributions are displayed in Table 1.

### 198 *Contamination data*

199 In situation A ( $C_g$  measured): SVOC gas phase concentration values ( $C_g$ ) were retrieved from  
200 Blanchard et al. (2014). When 100% (i.e. in 30 out of 30 dwellings) of the data were > the  
201 limit of quantification (LOQ), log-normal distributions were fitted. When 100% of the values

202 were < LOQ, uniform distributions between 0 and LOQ were used. Lastly when single values  
203 were > LOQ, custom distributions were constructed, related to their probabilities of  
204 occurrence, with continuous range between 0 and LOQ and discrete ranges for the values >  
205 LOQ. In situation B ( $C_g$  measured from  $C_a$ ): SVOC indoor air concentration values ( $C_a$ ) were  
206 retrieved from the literature (Fromme et al., 2004; Fromme et al., 2009 and Rudel et al.,  
207 2010). Log-normal distributions were fitted where possible; otherwise custom distributions  
208 were constructed, related to their probabilities of occurrence, with continuous ranges from 0  
209 to LOQ and from LOQ to maximum value, or triangular distributions between minimum,  
210 average and maximum values. Corresponding distributions are displayed in Table 1.

#### 211 *Human parameters*

212 We considered a 4-year-old male child to be representative of the sensitive population in  
213 terms of the identified toxicological effects and exposure behavior. As an example we  
214 searched literature for body weight ( $W$ ), surface area ( $A$ ), and time spent in dwellings ( $t$ ) for a  
215 child living in France. Log-normal distributions were used for weight and body surface area.  
216 Normal distribution was used for the space-time-budget. Corresponding distributions are  
217 displayed in Table 1.

#### 218 *Exposure media properties*

219 The assessment of gas phase SVOC dermal transfer requires the use of exposure media  
220 properties such as  $\gamma_d$ , [TSP],  $f_{om-part}$  and  $\rho_{part}$ . Triangular distribution was used for  $\gamma_d$ , using the  
221 minimum and maximum values found in the literature and the generally-assumed 6 m/h as the  
222 most likely value to occur. Log-normal distribution was used for [TSP] and  $f_{om-part}$ . Normal  
223 distribution was used for  $\rho_{part}$ . Corresponding distributions are displayed in Table 1.

### 224 3. Simulation

225 Dermal intake sensitivity analysis (Eq. 1) was performed using Crystal Ball® software  
226 (Oracle®, version 11.1.1.3.00). For each SVOC and for each of the two situations regarding  
227  $C_g$  (A:  $C_g$  measured, B:  $C_g$  modeled), Latin Hypercube one-dimensional simulations were  
228 carried out with  $10^5$  runs.

229 **Table 1:** Parameter distributions used in the dermal intake sensitivity analysis from gas phase for 8 SVOCs.

Parameter	DMP	Phenanthrene	HHCB	Permethrin	Diazinon	PCB105	BPA	BDE154	Sources
log (K <sub>ow</sub> )*	Log-normal min=1.35 μ=1.65 σ=0.14	Logistic μ=4.50 scale=0.09	Triangular min=3.42 μ=5.23 max=6.26	Minimum extreme likeliest=6.51 scale=0.73	Minimum extreme likeliest=3.74 scale=0.37	Minimum extreme likeliest=6.72 scale=0.35	Triangular min=3.32 μ=3.48 max=4.04	Triangular min=6.86 μ=7.89 max=8.83	Internet databases***, Chemexper, ACD/Labs calculators, EPI Suite software (US EPA, v4.1), Mackay et al., 2010a, 2010b, 2010c, 2010d.
H* (Pa.m <sup>3</sup> /mol)	Triangular min=6.20E-3 μ=3.60E-2 max=1.11E-1	Triangular min=2.38 μ=3.74 max=5.55	Uniform min=7.66E-2 max=1.34E+1	Triangular min=2.33E-6 μ=5.34E-2 max=1.89E-1	Triangular min=7.00E-3 μ=4.25E-2 max=1.44E-1	Triangular min=2.43 μ=2.24E+1 max=8.36E+1	Triangular min=9.28E-7 μ=2.49E-6 max=4.05E-6	Triangular min=4.77E-2 μ=1.46E-1 max=2.40E-1	
C <sub>g</sub> (ng/m <sup>3</sup> )	Log-normal min=0 μ <sub>g</sub> =8.57 σ <sub>g</sub> =2.23	Log-normal min=4.16 μ <sub>g</sub> =3.88 σ <sub>g</sub> =2.98	Log-normal min=0 μ <sub>g</sub> =6.67E+1 σ <sub>g</sub> =2.57	Uniform [0 - 0.6(LOQ)]	Discrete (p=0.03) 2.45 Uniform (p=0.97) [0 - 0.6(LOQ)]	Discrete (p=0.03) 0.4 Uniform (p=0.97) [0 - 0.25(LOQ)]	Uniform [0 - 0.6(LOQ)]	Uniform [0 - 0.6(LOQ)]	Blanchard et al., 2014.
C <sub>a</sub> (ng/m <sup>3</sup> )	Log-normal μ=1.18E+3 p50=4.36E+2 p95=4.65E+3	Log-normal μ=6.1 p50=1.10E+1 p95=2.90E+1	Log-normal μ=1.19E+2 p50=1.01E+2 p95=2.45E+2	Uniform (p=0.92) [0 - 0.3(LOQ)] Uniform (p=0.08) [0.3(LOQ) - 2]	Uniform (p=0.98) [0 - 0.3 (LOQ)] Uniform (p=0.02) [0.3(LOQ) - 3.1E+1]	Uniform (p=0.88) [0 - 0.3 (LOQ)] Uniform (p=0.12) [0.3(LOQ) - 1.2]	Uniform (p=0.84) [0 - 0.8(LOQ)] Uniform (p=0.16) [0.8(LOQ) - 2.2E+1]	Triangular min=0 μ=6.20E-4 max=1.09E-2	Rudel et al., 2010 (phen, per, diaz, PCB105, BPA), Fromme et al., 2004 (DMP, HHCB), Fromme et al., 2009 (BDE154).
Y <sub>d</sub> (m/h)	Triangular min=5, likeliest=6, max=10								Tamas et al., 2006; Pandrangi and Morrison, 2008; Weschler and Nazaroff, 2008.
[TSP] (μg/m <sup>3</sup> )	Log-normal, μ <sub>g</sub> =37.34, σ <sub>g</sub> =2.17, p95=182								Ramalho et al., 2012.
f <sub>om-part</sub>	Normal, μ=0.35, σ=0.2								Salthammer and Schripp, 2015.
ρ <sub>part</sub> (g/m <sup>3</sup> )	Normal, μ=1.6E+6, σ=0.5E+6								Pitz et al., 2003.
W <sup>**</sup> (kg)	Log-normal, μ <sub>lnx</sub> =2.68, σ <sub>lnx</sub> =0.17, p95=22.0								Tanguy et al., 2007.
A <sup>**</sup> (m <sup>2</sup> )	Log-normal, μ=-0.28, σ=0.12, p95=0.94								Sabaterie et al., 2013.
T (h/d)	Normal, μ=17.17, σ=0.63								Zeghnoun and Dor, 2010.

230 A: body surface area; BPA: bisphenol A;  $C_a$ : SVOC total concentration in indoor air;  $C_g$ : SVOC concentration in the gas phase; diaz: diazinon; DMP:  
231 dimethyl phthalate;  $f_{om-part}$ : volume fraction of organic matter associated with airborne particles;  $\gamma_d$ : coefficient describing the external transport of a gas phase  
232 SVOC from the bulk indoor air to the boundary layer adjacent to the skin; H: Henry's law constant; HHCB: galaxolide;  $K_{ow}$ : octanol/water partition  
233 coefficient;  $\mu$ : arithmetic mean;  $\mu_g$ : geometric mean; per: permethrin; phe: phenanthrene;  $\rho_{part}$ : density of airborne particles;  $\sigma$ : standard deviation;  $\sigma_g$ :  
234 geometric standard deviation; t: daily exposure duration; [TSP]: total suspended particle concentration; W: body weight. Distribution parameters may be  
235 different for a same distribution law because of diversity of reporting data in the literature.

236 \*: Measured or estimated at 25°C.

237 \*\*: Spearman's rank correlation between mass and body surface area is 0.99 for a 4-year-old male child (Sabaterie et al., 2013).

238 \*\*\*: Hazardous Substances Data Bank (HSBD), ChemIDplus, Chemspider, Chemicalize and the French toxicological and environmental data sheets from  
239 INERIS.

240 **RESULTS AND DISCUSSION**

241 1. Daily dermal intake variation

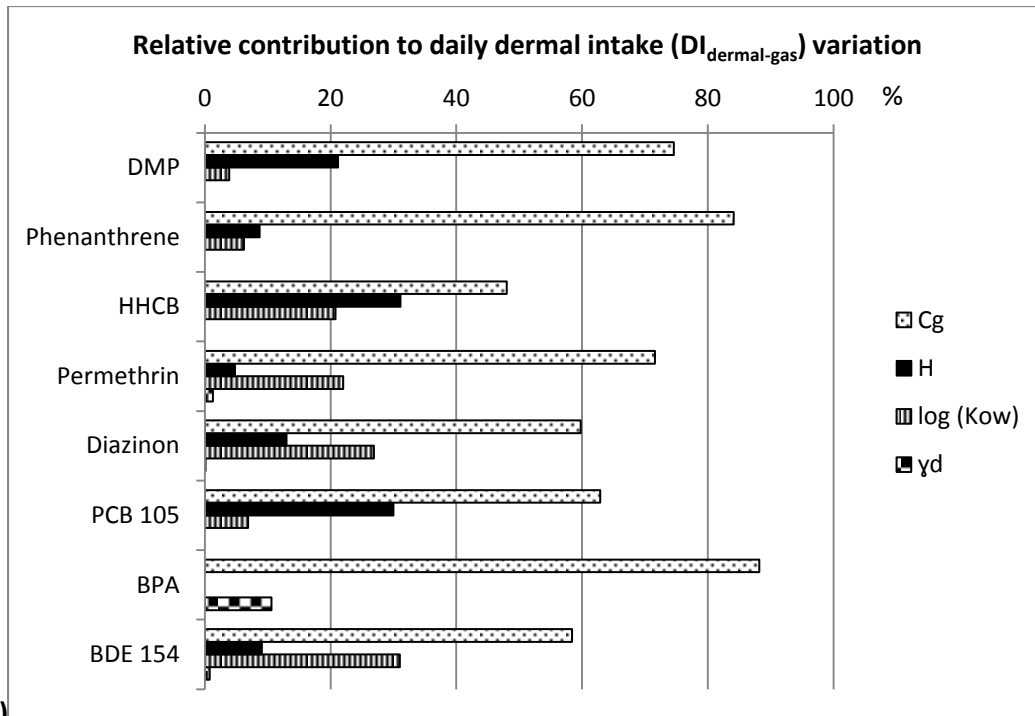
242 Daily dermal intake variations are presented in Table 2.

243 **Table 2:** Variations in daily dermal intake ( $DI_{\text{dermal-gas}}$ ), expressed as the relative interdecile  
 244 range, for each SVOC and for each of the two situations regarding gas phase concentration  
 245 (A:  $C_g$  measured, B:  $C_g$  modeled).

Situation	SVOC	Relative interdecile range $= \frac{d90 - d10}{d50}$
A	Dimethyl phthalate (DMP)	3.1
	Phenanthrene	1.9
	Galaxolide (HHCB)	6.3
	Permethrin	2.2
	Diazinon	3.1
	PCB 105	2.8
	Bisphenol A (BPA)	1.2
	BDE 154	1.6
B	Dimethyl phthalate (DMP)	6.2
	Phenanthrene	1.4
	Galaxolide (HHCB)	2.8
	Permethrin	3.6
	Diazinon	3.1
	PCB 105	3.7
	Bisphenol A (BPA)	17.8
	BDE 154	7.3

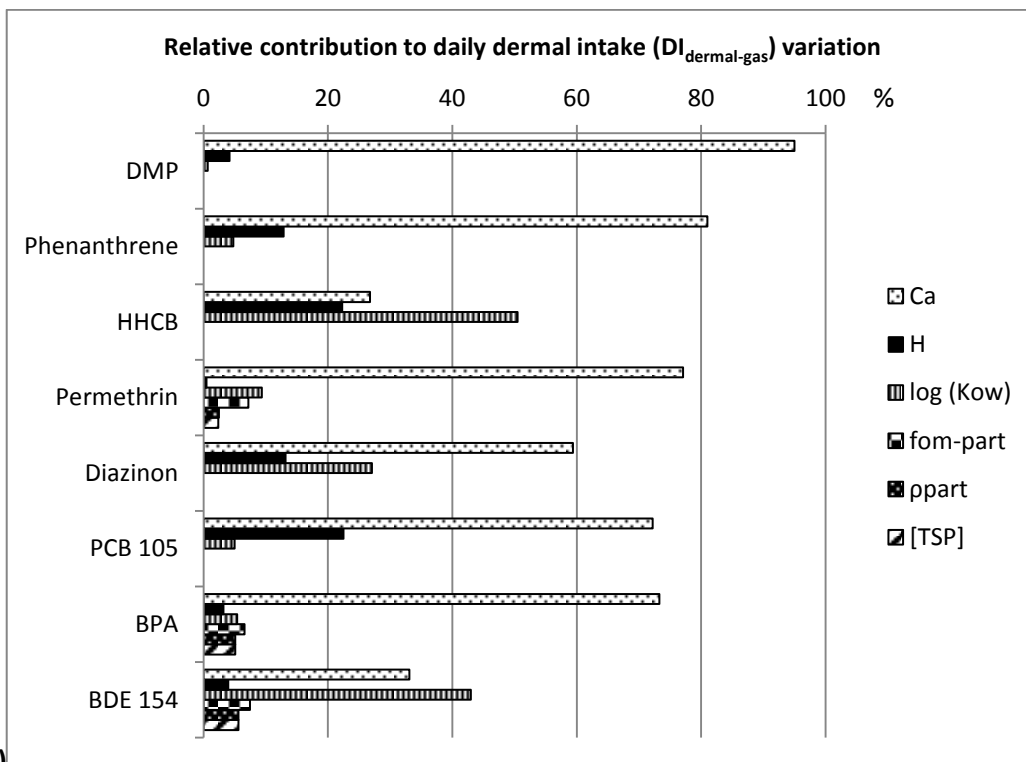
246  
 247 The relative interdecile range of  $DI_{\text{dermal-gas}}$ , when  $C_g$  value is measured (situation A), ranged  
 248 from 1.2 for BPA to 6.3 for galaxolide (HHCB). When  $C_g$  is estimated from  $C_a$  (situation B),  
 249  $DI_{\text{dermal-gas}}$  variation was highest for BPA having a relative interdecile range of 17.8 and  
 250 lowest for phenanthrene, at 1.4. The following sensitivity analysis results allow us to interpret  
 251  $DI_{\text{dermal-gas}}$  variation, particularly for compounds having high relative interdecile ranges.

252 2. Sensitivity to model parameters



253 A)

254



255 B)

256

257 **Figure 1:** Relative contribution (%) to total variation of gaseous SVOC daily dermal intakes  
 258 ( $\mu\text{g}/\text{kg-bw}/\text{d}$ ) according to key parameters:  $C_g$ ,  $C_a$ ,  $\log(K_{ow})$ , H,  $\gamma_d$ ,  $f_{\text{om-part}}$ ,  $\rho_{\text{part}}$ , and [TSP] for  
 259 both situations: A)  $C_g$  is measured and B)  $C_g$  is estimated from  $C_a$ .



260

261 Results of the sensitivity analysis are presented in Figure 1 as tornado charts, showing relative  
262 contributions to total variation of gas phase SVOC daily dermal intakes ( $\mu\text{g}/\text{kg}\text{-bw}/\text{d}$ )  
263 according to key parameters. Parameters having a relative contribution that is always lower  
264 than 5% are not shown in Figure 1, that is, each of the human parameters: W, A and t  
265 (situation A and B) and  $\gamma_d$  (only for situation B).

266 The sensitivity analysis of  $\text{DI}_{\text{dermal-gas}}$  calculation, when  $C_g$  is measured (situation A), reveals  
267 that for the studied compounds the most influential parameters are:  $C_g$ , H and  $\log(K_{ow})$ .  
268  $\text{DI}_{\text{dermal-gas}}$  estimation is mainly driven by variability in  $C_g$ .

269 When  $C_g$  is estimated from  $C_a$  (situation B),  $\text{DI}_{\text{dermal-gas}}$  variation is dominated by variability in  
270  $C_a$ , with the exception of HHCB and BDE 154 for which it is mainly driven by uncertainty in  
271  $\log(K_{ow})$ . For permethrin and BDE 154, uncertainties in  $f_{\text{om-part}}$ ,  $\rho_{\text{part}}$  and [TSP] are also  
272 significant, though less influential in  $\text{DI}_{\text{dermal-gas}}$  calculation.

273 For a given age group in both situations, the following parameters make a marginal or null  
274 contribution to  $\text{DI}_{\text{dermal-gas}}$  variation for all of the studied SVOCs: t, W, A, and  $\gamma_d$  - with the  
275 exception of BPA in situation A, for which  $\gamma_d$  makes a significant contribution.

### 276 3. Variability and uncertainty in $C_g$ and $C_a$ measurements

277 Indoor air SVOCs concentrations can be variable and/or uncertain. Variabilities in indoor air  
278 concentrations ( $C_g$  and  $C_a$ ) are high due to several conditions, such as differences in occupant  
279 habits, variety of sources, and dwelling characteristics. For example, Clausen et al. (2012)  
280 found that indoor temperature has a significant influence on DEHP air concentrations: this  
281 increases by a factor of about 10 with an increase of  $12^\circ\text{C}$  in indoor temperature.  
282 Furthermore, these concentrations may vary considerably from one country to another

283 depending, for example, on national regulations regarding the use of specific SVOCs  
284 (Weschler and Nazaroff, 2008). In this sensitivity analysis,  $C_g$  and  $C_a$  were retrieved from  
285 studies (Fromme et al., 2004; Fromme et al., 2009; Rudel et al., 2010 and Blanchard et al.,  
286 2014) having measured these concentrations at various indoor temperatures and in different  
287 countries. But  $C_g$  and  $C_a$  are also uncertain, especially when all or most values are below the  
288 LOQ. When  $C_g$  and  $C_a$  values were below the LOQ (permethrin, diazinon, PCB 105, BPA and  
289 BDE 154), the applied distribution shape (custom, triangular and uniform) also brings  
290 uncertainty to these unknown values. In order to assess the impact of the distribution on  $C_g$   
291 and  $C_a$  uncertainty, triangular distributions (between 0, LOQ/2 and LOQ) were also tested in  
292 place of uniform distributions (data not shown). The same results were found, providing  
293 evidence that distribution shape does not influence the relative contribution of  $C_g$  and  $C_a$   
294 uncertainty in  $DI_{\text{dermal-gas}}$  result variation.

295 Moreover, some LOQs used as maxima in uniform distribution regarding  $C_g$  (permethrin,  
296 BPA and BDE 154) are open to discussion because they are high in comparison with other  
297 studies in which fewer compounds were measured at the same time. This uncertainty can lead  
298 to discrepancy between the two situations regarding the distribution we used, especially for  
299 BDE 154 concentrations in indoor air (see Table 1) where  $C_g$  maximum value ( $0.6=LOQ$ ) is  
300 larger than  $C_a$  maximum (0.01). This unrealistic situation is the only one in our study - but it is  
301 important to bear in mind that the objective here is a sensitivity analysis linked to information  
302 availability, rather than an exposure assessment.

#### 303 4. Uncertainty in physical-chemical parameter values

304 Physical-chemical parameters can either be measured experimentally or calculated using other  
305 chemical properties. Depending on which of these methods is used, it follows that values vary  
306 by one order of magnitude or more (Finizio et al., 1997) - and these uncertainties will be

307 propagated in the calculation of  $k_{p-g}$ ,  $C_g$  and  $DI_{\text{dermal-gas}}$ . On this topic, Weschler and Nazaroff  
308 (2008) warned that  $H$ ,  $K_{ow}$  and  $K_{oa}$  values, calculated using the SPARC Online Calculator  
309 v4.0 (Hilal et al., 2003, 2004), sometimes vary substantially from experimentally derived  
310 values. For the same parameters, Schossler et al. (2011) demonstrated that values vary by one  
311 order of magnitude or more between the results obtained using software tools such as EPI  
312 Suite (US EPA, 2013) or the SPARC Online Calculator v4.6. In this study,  $\log(K_{ow})$  and  $H$   
313 values were collected from several databases, online calculators and literature sources at  
314 25°C, and for each SVOC (see Table 1). The wide intervals obtained for certain compounds -  
315 such as  $H$  for HHCB - corroborate the relatively high level of uncertainty for these  
316 parameters.

317 When assessing gas/particle SVOC distribution (see Equation 8 in the supplementary material  
318 for the detailed calculation), Salthammer and Schripp (2015) assumed normal distributions for  
319  $H$  and  $\log(K_{ow})$ . Due to the lack of data (as discussed above) the authors calculated mean and  
320 standard deviation, in order to build normal distribution using just two values for certain  
321 compounds. In our study, where at least 15 values for these parameters were retrieved from  
322 literature, log-normal distributions were fitted - otherwise we used triangular, minimum  
323 extreme or logistic distributions (see Table 1). However, in order to assess the impact of  
324 distribution, other distributions, such as uniform, were also tested and the same parameters  
325 were found to be most sensitive regarding  $DI_{\text{dermal-gas}}$  variability, providing evidence that  
326 choice of distribution shape does not influence the relative contribution made by  $H$  and  $\log$   
327 ( $K_{ow}$ ) in  $DI_{\text{dermal-gas}}$  result variation.

328 In each situation (A and B),  $H$  and  $\log(K_{ow})$  have a significant influence on  $DI_{\text{dermal-gas}}$   
329 variability, depending on the range of  $H$  and  $\log(K_{ow})$  values. For example,  $H$  values for  
330 HHCB range from [7.66E-2 to 13.4], which logically leads to this parameter having a greater  
331 influence on  $DI_{\text{dermal-gas}}$  variability (see Figure 1). These results are consistent with previous

332 studies: Weschler and Nazaroff (2014) assessed  $k_{p-g}$  sensitivity (see Equation 2) to the same  
333 key parameters and also found that the permeability coefficient was more sensitive to H. In  
334 the same way, Salthammer and Schripp (2015) assessed the sensitivity of  $K_p$  (see Equations 8  
335 and 9 in the supplementary material for the detailed calculation) and found that the error  
336 margin in  $K_p$  calculation was dominated by H uncertainty. Because  $\log(K_{ow})$  and H are two  
337 of the most influential parameters on  $DI_{\text{dermal-gas}}$  variation for every SVOC and in both  
338 situations, reducing their uncertainties could significantly reduce variation on  $DI_{\text{dermal-gas}}$  and  
339 uncertainty in exposure analysis.

#### 340 5. Uncertainty in exposure media properties

341 In earlier studies, default values were used for  $f_{\text{om-part}}$ ,  $\rho_{\text{part}}$ , [TSP] and  $\gamma_d$ . Regarding the  
342 volume fraction of organic matter associated with airborne particles,  $f_{\text{om-part}}$ , we assumed the  
343 same normal distribution parameters as Salthammer and Schripp (2015). Regarding particle  
344 density,  $\rho_{\text{part}}$ , several values are found in the literature and  $1.10^6 \text{ g/m}^3$  is often assumed, as a  
345 default value (Turpin and Lim, 2001). In order to assess  $DI_{\text{dermal-gas}}$  sensitivity to this  
346 parameter, normal distribution was used rather than a single value (Pitz et al., 2003).  
347 Regarding total suspended particle concentration, [TSP], a default value of  $20\mu\text{g/m}^3$  was  
348 assumed by Weschler and Nazaroff (2008). More recently, Salthammer and Schripp (2015)  
349 found that [TSP] strongly influenced gas/particle partitioning, and we decided to build a log-  
350 normal distribution for this parameter using data on indoor  $PM_{10}$  concentrations (Ramalho et  
351 al., 2012). These data are weekly-averaged, and cover different climate zones and seasons.  
352 Regarding  $\gamma_d$ , a value of 6m/h is assumed for the coefficient describing the external transport  
353 of a gas phase SVOC from bulk indoor air to the boundary layer adjacent to the skin  
354 (Weschler and Nazaroff, 2012). The authors have previously estimated this parameter to  
355 range between 5 and 10 m/h (Weschler and Nazaroff, 2008). In this study, triangular  
356 distribution was built between these three values. This parameter variation's influence on

357  $DI_{\text{dermal-gas}}$  variation was insignificant, with the exception of BPA in situation A (see Figure 1,  
358 A). Nevertheless, Weschler and Nazaroff (2008) also proposed an estimate of 3 m/h for the  
359 mass-transfer coefficient. In order to provide a comprehensive sensitivity analysis we  
360 assessed different distribution shapes: triangular with a minimum of 3, a likeliest value of 6  
361 and a maximum of 10 and uniform between 3 and 10. The results (not shown) were identical  
362 and  $v_d$  did not become an influential parameter.

363 While exposure media properties such as  $f_{\text{om-part}}$ ,  $\rho_{\text{part}}$ , [TSP] and  $v_d$  have a slight influence  
364 (less than 10%, see Figure 1) for some compounds (permethrin, BPA and BDE 154), they  
365 make a marginal or null contribution (less than 5%) to the variance of dermal intake for a  
366 given age group, for the other SVOCs and in both situations.

#### 367 6. Variability in human parameters

368 Human parameters such as body weight (W), body surface area (A) and daily exposure (t)  
369 make a marginal or null contribution (less than 5%) to the variance of dermal intake for a  
370 given age group, for each SVOC and in each situation. However, one has to bear in mind that  
371 we ran the model for a given age group and that these parameters would have a larger impact  
372 when applied on a more diverse population. Regarding the role of clothing in dermal  
373 exposure, Piotrovski (1971) assessed the exposed body surface area (A) and found little  
374 difference in dermal absorption between clothed and naked people exposed to phenol vapor.  
375 More recently Morrison et al. (2016) assessed the influence of clothing on the dermal uptake  
376 of two phthalates (DEP and DnBP). The authors found that clean clothes were protective  
377 against air pollutants; whereas worn clothes, because they have adsorbed air pollutants,  
378 increased dermal intake. Because only clean clothes could be considered protective, we  
379 decided to not take into account the role of clothing in this sensitivity analysis, and to assume  
380 total body surface area exposed to indoor air when calculating  $DI_{\text{dermal-gas}}$ . However, the fact

381 that we found A to make a marginal or null contribution (less than 5%) to the variance of  
382 dermal intake, does not mean that clothing should not, when possible, be taken into account in  
383 assessing dermal exposure. In this case the proportion of exposed body surface area has to be  
384 taken into account.

385 In addition to the important role of clothing, other parameters not included in this model are  
386 suspected of influencing or playing a role in dermal exposure were not taken into account in  
387 this sensitivity analysis: skin temperature, metabolic processes on (e.g. ionization) or in the  
388 skin, the effects of bathing on SVOC levels in skin-surface lipids, etc.

389 Furthermore, it is important to bear in mind that we did not assess model uncertainty - only  
390 parametric uncertainty when using this model. A first source of uncertainty is model  
391 boundaries. Indeed our model relies partly on equation of Mitragotri (2002) that may lead to  
392 greater uncertainty when MW is higher than 400, which is the case for BDE 154. In addition,  
393 more sophisticated, and recent, models exist taking into account the dynamics of aerosols  
394 and/or the clothing effect. Regarding the dynamics of aerosols, Shi and Zao (2015) showed  
395 that, in their model, air exchange rate and surfaces cleaning frequency were influential  
396 parameters, while density of settled dust and its organic fraction were important media  
397 properties. Also, the transient model proposed by Gong et al. (2014), addresses the rapidly  
398 changing conditions and concentrations and considers a convective mass transfer resistance in  
399 the boundary air layer adjacent to the skin, and leads to lower estimates of dermal uptake.  
400 Morrison et al. (2016) improved this model taking clothing effect modeling, and showed that  
401 bathing frequency and change of clothes frequency were influential. Also, the introduction of  
402 a skin surface lipid film in the models and its interactions with clothing may affect the results,  
403 so do the corresponding additional parameters, such as for instance the thickness of this  
404 lipidic film.

## 405 CONCLUSION

406 When assessing dermal absorption of gas phase SVOCs, variation of dermal intake estimation  
407 is driven firstly by variability and uncertainty in indoor air concentration ( $C_g$  or  $C_a$ ), and  
408 secondly by uncertainty in SVOC physical-chemical parameters:  $\log(K_{ow})$  and  $H$ . While  
409 exposure media properties such as volume fraction of organic matter in the particle phase ( $f_{om-$   
410  $_{part}$ ), particle density ( $\rho_{part}$ ), concentration ([TSP]) and transport coefficient ( $\gamma_d$ ) do have a  
411 slight influence (less than 10%) for some compounds, human parameters such as body weight  
412 ( $W$ ), body surface area ( $A$ ) and daily exposure ( $t$ ) make a marginal or null contribution (less  
413 than 5%) to the variance of dermal intake for a given age group.

414 Considering that  $DI_{dermal-gas}$  variation can be high for some compounds, exposure assessors  
415 aiming to assess SVOC  $DI_{dermal-gas}$  using the  $k_{p-g}$ , or to estimate  $C_g$  from  $C_a$ , must pay  
416 particular attention to the determination, estimation, and selection of the following SVOC-  
417 specific parameters: concentration in gas phase ( $C_g$ ) or indoor air ( $C_a$ ),  $K_{ow}$  and  $H$ .

418 It is however important to remain aware, when analyzing these results, that exposure to an  
419 SVOC is strongly dependent on its partition between gas phase and particulate phase. When  
420 an SVOC is more abundant in the gas phase, dermal absorption will be greater than dust  
421 ingestion, and conversely, when a SVOC is more present in the particulate phase, dust  
422 ingestion is likely to be greater than dermal absorption (Weschler and Nazaroff, 2012).  
423 Therefore, less volatile SVOCs ( $P_s < 10^{-6}$  Pa), which are more present in the particulate phase  
424 and have a low predicted dermal absorption, do not require the same caution in estimation of  
425 dermal intake in order to assess their total exposure to indoor SVOC.

426 In general, inclusion of an uncertainty analysis in exposure assessment appears to be essential.  
427 In view of these sensitivity analysis results, reducing  $\log(K_{ow})$  and  $H$  uncertainties could  
428 significantly reduce uncertainties in  $DI_{dermal-gas}$  assessment.

429 **ACKNOWLEDGMENTS**

430 This research did not receive any specific grant from funding agencies in the public,  
431 commercial, or not-for-profit sectors.

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