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1 Indoor air pollutant exposure for life cycle assessment: 2 regional health impact factors for households

3 Ralph K. Rosenbaum^{*1,2}, Arjen Meijer^{*3}, Evangelia Demou^{4,5}, Stefanie Hellweg⁶, Olivier Jolliet⁷,
4 Nicholas L. Lam⁸, Manuele Margni⁹, Thomas E. McKone⁸

5 1. Irstea, UMR ITAP, ELSA Research group & ELSA-PACT—Industrial Chair for Environmental
6 and Social Sustainability Assessment, 361 rue J.F. Breton, 5095, 34196 Montpellier, France

7 2. Department of Management Engineering, Technical University of Denmark, Lyngby, Denmark

8 3. OTB Research for the Built Environment, Faculty of Architecture and the Built Environment,
9 Delft University of Technology, Delft, The Netherlands

10 4. Healthy Working Lives Group, Institute of Health and Wellbeing, College of Medical,
11 Veterinary and Life Sciences, University of Glasgow, Glasgow, UK

12 5. MRC/CSO Social and Public Health Sciences Unit, University of Glasgow, Glasgow, UK

13 6. Institute of Environmental Engineering, ETH Zurich, Zurich, Switzerland

* Corresponding authors: R.K. Rosenbaum, Irstea, Industrial Chair ELSA-PACT, 361 Rue Jean-François Breton, BP 5095, 34196 Montpellier, France, Ph. +33 499612048, ralph.rosenbaum@irstea.fr;
A. Meijer, TU Delft / Faculty of Architecture and The Built Environment, OTB - Research for the Built Environment, PO Box 5043, 2600 GA Delft, The Netherlands, Ph. +31 152785658, a.meijer@tudelft.nl

14 7. Environmental Health Sciences, School of Public Health, University of Michigan, Ann Arbor,
15 U.S.A.

16 8. School of Public Health, University of California Berkeley, Berkeley, U.S.A.

17 9. Department of Mathematical and Industrial Engineering, CIRAIQ - Polytechnique Montreal,
18 Montreal, Canada

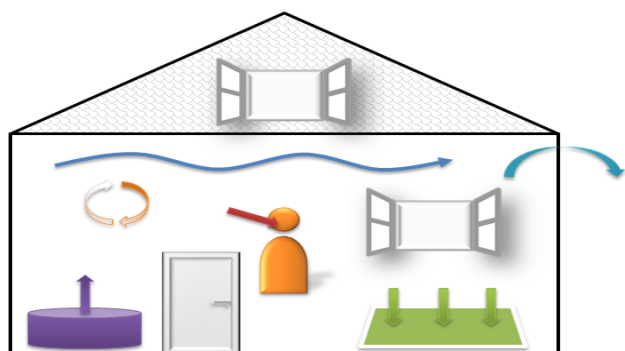
19 **Abstract**

20 Human exposure to indoor pollutant concentrations is receiving increasing interest in Life Cycle
21 Assessment (LCA). We address this issue by incorporating an indoor compartment into the USEtox
22 model, as well as by providing recommended parameter values for households in four different regions
23 of the world differing geographically, economically, and socially. With these parameter values, intake
24 fractions and comparative toxicity potentials for indoor emissions of dwellings for different air tightness
25 levels were calculated. The resulting intake fractions for indoor exposure vary by two orders of
26 magnitude, due to the variability of ventilation rate, building occupation and volume. To compare health
27 impacts as a result of indoor exposure with those from outdoor exposure, the indoor exposure
28 characterization factors determined with the modified USEtox model were applied in a case study on
29 cooking in non-OECD countries. This study demonstrates the appropriateness and significance of
30 integrating indoor environments into LCA, which ensures a more holistic account of all exposure
31 environments and allows for a better accountability of health impacts. The model, intake fractions, and
32 characterization factors are made available for use in standard LCA studies via www.usetox.org and in
33 standard LCA software.

34 **Keywords**

35 Life Cycle Assessment, indoor exposure, USEtox, characterization factor, comparative toxicity
36 potential, cooking, solid fuels, household air pollution

37 **TOC/Abstract Art**



39 **Introduction**

40 Life cycle Assessment (LCA) has broad applications in supply chain management and policy analysis,
41 helps to identify effective improvement strategies for the environmental performance of products or
42 services and to avoid burden shifting between different environmental issues.¹ Current LCA
43 methodology covers more than a dozen impact categories such as climate change, acidification,
44 eutrophication, land-use, or water-use, as well as toxicity, distinguishing ecotoxicity and human
45 toxicity. The latter currently only considers outdoor exposure to ubiquitous chemical concentrations in
46 the environment (or food) from emissions of a product's or service's life cycle, while indoor exposure
47 with proximity to sources emitting in confined (dilution) volumes have not yet been integrated. It is
48 important to note that LCA employs an "emitter perspective" aiming to assess potential impacts of
49 chemical exposure related to a given emission, i.e. marginal exposure or impact attributable to a specific
50 emission source. This is different from Environmental Risk Assessment, which is based on a "receptor
51 perspective" aiming to measure the level of cumulative exposure from single or multiple sources of
52 chemical emission, no matter where these occur.

53 Human exposure to indoor concentrations of chemicals is receiving increasing interest in LCA.²⁻¹¹
54 Due to the often high concentrations of harmful substances in indoor environments and the long periods
55 people spend indoors, the indoor intake per unit of (indoor) emission of these substances can be equal or
56 higher than outdoor intake, by up to several orders of magnitude.^{4,5} Inclusion of indoor exposure in

57 LCA has been acknowledged as an area of need by the UNEP/SETAC Life Cycle Initiative
58 (<http://www.lifecycleinitiative.org>), which is taking up recommendations and conclusions toward the
59 enhancement of the current LCA framework study. Within this initiative, an international expert group
60 on the integration of indoor and outdoor exposure in LCA has formulated a framework for integration of
61 indoor exposure in LCA.⁶ They found that a single-compartment box model is most compatible with
62 LCA and therefore recommended it for use as a default in LCA. Indoor intake fractions were found to
63 be several orders of magnitude higher in many cases than outdoor intake fractions, which highlights the
64 relevance of considering indoor exposure. While an initial set of model parameter values was provided
65 and the integration of the model into the USEtox model was suggested in the previous study, a full set
66 of representative parameter values for various indoor settings is still missing to make this approach
67 operational.⁶ The model parameters given in the framework have been presented as ranges of values.⁶
68 The actual values of the parameters depend on the geographical region of the assessed site, the type and
69 characteristics of the dwelling, and the characteristics and behavior of the occupants. In LCA, when no
70 data are available about the actual dwelling or the occupants, average parameter values are generally
71 used.

72 USEtox is a tool for calculation of comparative toxicity potentials (characterization factors) for
73 human health and freshwater ecosystems, developed under the auspices of the UNEP/SETAC Life
74 Cycle Initiative. It models a cause–effect chain that links emissions to impacts through three steps:
75 environmental fate, exposure, and effects. It was developed as a methodology simple enough to be used
76 on a worldwide basis and for a large number of substances while incorporating broad scientific
77 consensus.^{12,13} It is the recommended LCA (midpoint) toxicity characterization model of the European
78 Union¹⁴, endorsed by the UNEP/SETAC Life Cycle Initiative, and adopted by the US-EPA’s life cycle
79 impact assessment tool TRACI.¹⁵ Therefore, it is regarded as the relevant basis to integrate indoor and
80 outdoor exposure characterization into one consistent method for use in LCA, as also discussed by
81 Hellweg et al.⁶

82 The aims of this paper are 1) extending the USEtox model¹³ to include the indoor environment as a
83 compartment; 2) providing an overview of recommended parameter values to be used as default
84 household model parameters for different geographical settings; 3) comparing intake fractions
85 calculated with these recommended default parameters with intake fractions for outdoor exposure; and,
86 4) applying the new characterization factors for indoor exposure to a comprehensive case study on
87 cooking worldwide. The scope of this paper is restricted to the LCA emitter perspective, i.e. the
88 calculation of potential health effects from indoor emissions modeled as the cumulative impacts from
89 indoor exposure and outdoor exposure due to indoor emissions only. The focus was on indoor emissions
90 of volatile and semi-volatile organic compounds, because pollutants such as particles, ozone or NOx
91 require specific model processes for transport and transformation and are currently not addressed by
92 LCA toxicity models and not included in USEtox. In LCA their impacts on human health are assessed
93 respectively in the separate impact categories “particulate matter formation” and “photochemical ozone
94 formation”.

95 **Materials and Method**

96 The one-box model recommended by Hellweg et al. for estimation of indoor air intake fraction is
97 given as (Equation 1b in ⁶):

$$98 \quad iF = \frac{IR}{V \cdot m \cdot k_{ex}} \cdot N \quad (1)$$

99 where *iF* is the population intake fraction of a chemical (-), *IR* is the daily inhalation rate of air of an
100 individual (m³/day), *N* is the number of people exposed (-), *V* is the volume of the exposure area (m³),
101 *k_{ex}* is the air exchange rate of the volume in the exposure area (-) and *m* is the mixing factor (-). The
102 following sections describe how this has been implemented into the matrix-algebra framework of the
103 USEtox model.¹³

104 **Overall framework:** In the USEtox framework based on Rosenbaum et al.¹⁶, the characterization
105 factor matrix that represents the impact per kg substance emitted is obtained by multiplying an intake

106 fraction matrix (**iF**) by an effect factor matrix (**EF**). The intake fraction is the product of a fate matrix
107 (**FF**) and an exposure matrix (**XF**):¹⁶

$$108 \quad \mathbf{CF} = \mathbf{EF} \cdot \mathbf{iF} = \mathbf{EF} \cdot \mathbf{XF} \cdot \mathbf{FF} \quad (2)$$

109 The unit of the elements in **FF** is [d], in **XF** [1/d], in **iF** [$\text{kg}_{\text{intake}}/\text{kg}_{\text{emitted}}$], in **EF** [disease cases/kg of
110 chemical intake], and in **CF** [disease cases/kg emitted] or CTU_h , which is the name given by the
111 USEtox developers to the results (characterization factors) of their model for human health (as opposed
112 to CTU_e - Comparative Toxic Unit for ecosystems).¹³ For the concept and interpretation of these
113 matrices, their elements and their units we refer to Rosenbaum et al.¹⁶ The matrix-algebra based
114 calculation framework of USEtox allows for the straightforward integration of additional compartments
115 and exposure pathways by simply adding the corresponding columns or rows to the respective fate and
116 exposure matrices.¹⁶ All parameters describing the indoor compartment and the resulting exposure are
117 provided as recommended value sets for household settings in different regions, but can also be
118 modified freely by the user in the model to represent more site-specific conditions.

119 **Fate:** The fate matrix **FF** [d] is calculated as the inverse of the exchange-rate matrix **K** [1/d]:

$$120 \quad \mathbf{FF} = (-\mathbf{K})^{-1} \quad (3)$$

121 The exchange-rate matrix **K** represents the exchange rate between compartments in the non-diagonal
122 terms and the overall removal rate in the diagonal term (with a negative sign). The indoor environment
123 is modeled as a separate air compartment contributing to the overall inhalation exposure of humans.
124 This compartment is added to the existing 11 USEtox compartments.¹³ Three removal mechanisms are
125 considered according to Wenger et al.¹⁷:

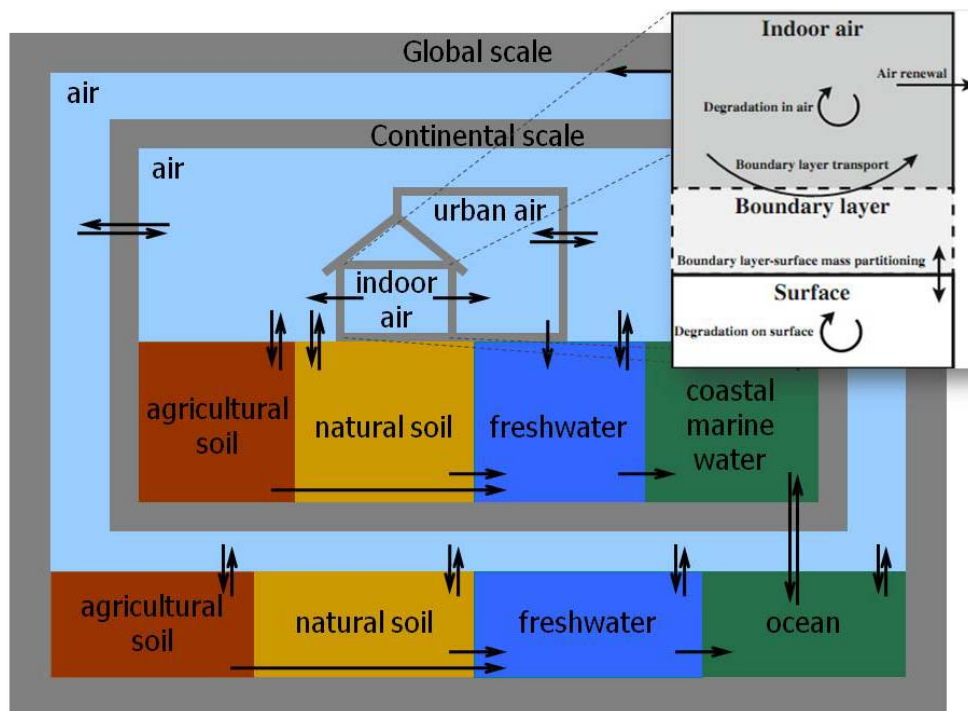
126 1) The advective ventilation flow, parameterized as the air exchange rate k_{ex} [h^{-1}] (as in Equation 1b
127 in ⁶). The air exchange rate does not depend on the substance, but on the building characteristics, such
128 as type and size of windows and doors, type of walls, and the number of cracks in the façades. Average
129 values for several regions are given in Table S1 in the Supporting Information (SI). k_{ex} is not a loss, but
130 an inter-media transport mechanism connecting indoor with outdoor compartments. Based on the
131 average distribution of the global population between urban and rural areas of about 50% respectively¹⁸,

132 half of the ventilation flow is directed to each of the urban and continental rural environments of
133 USEtox (Figure 1). This is taken into account in the model by a non-diagonal term from indoor to
134 compartment *i* given as: $k_{indoor,i} = f_{ex,i} k_{ex}$, with $f_{ex,i} = 0.5$ for transfers to both urban and continental
135 rural air compartments (*i*).

136 2) The gas-phase (g) air-degradation rate $k_{g,deg}$ [h^{-1}] is mainly related to reactions with ozone, hydroxyl
137 radicals, and nitrate radicals (gas-phase degradation). The overall degradation rate in the indoor air is
138 calculated as the average radical concentration ([OH], [O₃], [NO₃]) multiplied by the corresponding
139 second order degradation rate constant: $k_{g,deg} = k_{OH} \cdot [OH] + k_{O_3} \cdot [O_3] + k_{NO_3} \cdot [NO_3]$. Long-term
140 averaged indoor concentrations of ozone ([O₃] = 8 ppb), hydroxyl radical ([OH] = 3×10^{-6} ppb) and
141 nitrate radical ([NO₃] = 10^{-3} ppb) were taken from Wenger et al.¹⁷ and second order degradation rate
142 constants from the EPI Suite v4.1 software¹⁹, which provides OH rate constants for most substances, but
143 only few for O₃ and NO₃.

144 3) An equivalent removal rate by adsorption to indoor surfaces, k_s [h^{-1}] can be calculated as a net
145 removal rate from the air, assuming steady-state conditions between the air and room surface without
146 adding a separate compartment.¹⁷ This approach is similar to the net removal rate calculated in USEtox
147 from the freshwater outdoor environment to the sediments, which are not considered as separate
148 compartments to limit the model complexity.²⁰ Since degradation on surfaces is not well characterized,
149 this removal rate to surfaces is subject to high uncertainty. Surface removal in the current model is
150 applied primarily to Semi-Volatile Organic Compounds (SVOCs), for which additional gaseous dermal
151 exposure may also be relevant and may compensate this removal. If the model is eventually used for
152 particulate matter (PM) and ozone, then surface removal could become more important and requires
153 further assessment of the literature on indoor ozone and PM deposition including the work of
154 Weschler²¹ and Nazaroff²². We therefore do not include the sorption removal pathway in the default
155 model, but only consider it for the sensitivity study together with the dermal gaseous exposure pathway.
156 A more detailed description of the calculation of the equivalent removal rate to the surface k_s is given in
157 SI (section S3).

158 The air degradation rate and the equivalent removal rate to the surface directly add up to the air
 159 exchange rate for the diagonal term of **K**.



160
 161 Figure 1: Schematic representation of the USEtox model with indoor compartment embedded; adapted
 162 from Rosenbaum et al.¹³ and Wenger et al.¹⁷

163
 164 **Exposure:** The exposure pathway considered in this paper is inhalation. The relevant parameters for
 165 inhalation exposure in households are the following: individual daily inhalation (breathing) rate (IR)
 166 [m³/d], average number of people in the building N [dimensionless], building volume V [m³], and daily
 167 time fraction spent indoors f_t [dimensionless]. The latter is the quotient of the time spent indoors and the
 168 total time of a day (24h). Recommendations, assumptions, and choices for these parameter values are
 169 further discussed below. The exposure factor XF [1/d] for the indoor exposure setting is then calculated
 170 based on Equation 1b in Hellweg et al.⁶ (with mixing factor $m = 1$, assuming that complete mixing
 171 within the indoor volume is an inherent hypothesis of the indoor iF model):

$$172 \quad XF = \frac{IR}{V} \cdot f_t \cdot N \quad (4)$$

173 The calculated XF values are placed in the corresponding element of the exposure matrix **XF** in
 174 USEtox. For SVOCs the dermal absorption of gas-phase chemicals may become important and means

175 that the validity of equation (4) is restricted to VOCs.²³⁻²⁵ In this paper the potential influence of the
176 dermal gaseous uptake pathway is considered as a sensitivity study together with the influence of
177 adsorption removal on indoor surfaces which competes with this exposure pathway. Existing
178 approaches^{17,26} were adapted to determine the convective transfer at body surface as a function of heat
179 transfer coefficients²⁷, which might be added to USEtox in a later stage once data will be broadly
180 available and the models further evaluated, in conjunction with the introduction of a dermal pathway
181 within USEtox.

182 **Effect and characterization factor:** The human health effect factor EF is the same as for outdoor
183 exposure in USEtox and thus also independent of the exposure setting or region. Therefore, EF was
184 taken directly from the USEtox database. According to Rosenbaum et al.¹⁶ the characterization factor
185 matrix **CF** (named **HDF** in ¹⁶) is then obtained by multiplying the matrices **FF**, **XF**, and **EF** (Equation
186 2).

187 **Model Parameterization**

188 In order to calculate characterization factors (and intake fractions) for indoor exposure, the parameters
189 discussed above are needed in the USEtox model. In LCA, the exact situation where the indoor
190 exposure takes place is seldom known. In order to calculate characterization factors for generic
191 situations, regions can be defined, for each of which a characterization factor can be calculated using
192 region-specific parameters. Regions can be defined as 1) countries or continents, 2) based on the level
193 of economic development or urbanization, or 3) as a combination of 1) and 2).

194 For several parameters, the data availability is limited for most regions, especially for non-OECD
195 countries. Especially for houses with low air-exchange in non-OECD countries, few data about the
196 parameters needed for the calculations are available, specifically for building volumes (**V**), occupation
197 (**N**), and air exchange rate (**k_{ex}**). You et al. found air exchange rates in 41 elderly homes in China
198 ranging from 0.29 hr⁻¹ to 3.46 hr⁻¹ in fall (median: 1.15 hr⁻¹), and from 0.12 hr⁻¹ to 1.39 hr⁻¹ in winter
199 (median: 0.54 hr⁻¹).²⁸ Massey et al. found air exchange rates in 10 houses in northern India ranging from
200 2.5 hr⁻¹ to 3.1 hr⁻¹ in winter and 4.6 hr⁻¹ to 5.1 hr⁻¹ in summer.²⁹ These data suggest that air exchange

201 rates in houses with low air-exchange in non-OECD countries may be higher than in houses with low
 202 air-exchange in OECD countries. However, it is not clear how representative the dwellings described by
 203 You et al. and Massey et al. are for all houses with low air-exchange in the respective countries.

204 Therefore, four regions have been defined in this study: Europe (EU-27), North America (USA),
 205 OECD countries, and non-OECD countries. We assume that a population-weighted average from EU-27
 206 countries is representative for Europe, that an average from the USA is representative for North
 207 America, that a population-weighted average from EU-27 countries and the USA is representative for
 208 OECD countries, and that a population-weighted average from China, India, Uganda, Brazil, and
 209 Guatemala is representative for non-OECD countries. The region-specific parameters considered are the
 210 building volume (V) and the number of people in the building (N). For the air exchange rate (k_{ex}) data
 211 availability is even less robust than for N and V. Therefore, a distinction has been made between houses
 212 with a low air exchange rate ($k_{ex} < 8 \text{ h}^{-1}$) named “L-AER” and houses with higher air exchange rates
 213 ($k_{ex} > 8 \text{ h}^{-1}$, especially for houses with no windows and/or doors) named “H-AER”. All houses in OECD
 214 countries were assumed as having a relatively low air-exchange, while in non-OECD countries, houses
 215 with both low and high air exchange (e.g. houses with no glass in the windows) exist. In the absence of
 216 data for houses with low air-exchange in non-OECD countries, we assume the same value for k_{ex} as for
 217 OECD countries. In Table 1, the recommended values of the region-specific parameter sets are
 218 summarized. In SI (Table S1), the parameter values are given for the different countries within the
 219 regions.

220 Table 1: Recommended parameter values and standard deviations (SD) for the indoor exposure model
 221 per region, calculated as averages from the individual countries and weighted over the population of
 222 those countries

| Region | V [m ³] | | N [-] | | k _{ex} [h ⁻¹] | | IR [m ³ /d] | f _t [-] |
|-------------------------------------|---------------------|--------------|---------|--------------|------------------------------------|------|------------------------|--------------------|
| | Average | SD | Average | SD | Average | SD | | |
| Non-OECD countries (H-AER building) | 119 | 25.6 | 4.0 | 0.87 | 15.6 | 0.85 | 13 | 0.58 |
| Non-OECD countries (L-AER building) | | | | | 0.64 | 0.08 | | |
| OECD countries | 236 | 37.9 | 2.5 | 0.22 | | | | |
| Europe (EU-27) | 209 | 22.9 | 2.4 | 0.26 | | | | |
| North America (USA) | 277 | ^a | 2.6 | ^a | | | | |

^a single data point (US average) as we are using country averages and hence no variability assessed on sub-country level
 See Table S1 in SI for data per country and literature references

223

224 We assume the daily individual inhalation rate for humans for indoor exposure to be $13 \text{ m}^3/\text{d}$, the same
225 as USEtox assumes for outdoor exposure.¹³ The average time spent indoors needs to be differentiated
226 between time spent at work and time spent at home (which could even be further distinguished between
227 private and public buildings such as shops, restaurants, etc.), where exposure conditions can be very
228 different. As we are focusing here on household exposure, we assume a daily average of 14 hours spent
229 at home. These can be complemented by 7-8 hours at work, leaving 2-3 hours outdoors. The time
230 fraction spent indoors (at home) is then calculated as $f_t = 14\text{h}/24\text{h} = 0.58$.

231 Although, these parameters have a strong regional dependency based on cultural and climatic
232 variability³⁰, it was not possible to consider this due to very limited data availability and a strong bias
233 towards OECD country-data where data are available. The European Expolis study for example,
234 measured between 18 to 23 hours spent indoors (total) and a range of 0.06 to 5 hours spent outdoors
235 (total) for the adult population (25-55 y) in the seven participating urban areas.³¹ The Expolis time-use
236 dataset is the largest multinational European time-use data set, which has been gathered specifically for
237 exposure assessment purposes. Time activity data were gathered from 808 persons in seven European
238 cities: Athens, Basel, Grenoble, Helsinki, Milan, Oxford, and Prague.³⁰⁻³² For North America, the U.S.
239 National Human Activity Pattern Survey (NHAPS) showed that the mean percentage of time spent
240 indoors was 21 hours, with 14 hours of this time spent in a residence and 4 hours of the time spent in
241 other indoor locations.³³ Similar time-patterns were also observed in the Canadian Human Activity
242 Pattern Survey (CHAPS), with some seasonal variations from the U.S. pattern.³⁴ Smith reports that even
243 in developing countries, people spend 70% or more of the day indoors.³⁵

244 **Sensitivity and variability analysis**

245 For those chemicals with an indoor iF dominated by removal via ventilation rather than by
246 degradation or adsorption, a parameter sensitivity and variability analysis was performed, in order to
247 determine their contribution to variance. Since the ranges of these parameters (Table S1, SI) represent
248 variability (between countries, building types, or individual persons) rather than uncertainty, the
249 analysis only quantifies some of the overall variance, essentially being a variability analysis. The

250 following parameters used to calculate indoor iF were included in the variability analysis using Monte
251 Carlo simulation with 50,000 iterations and Latin Hypercube Sampling (Crystal Ball 11.1.2): 1)
252 building volume V; 2) number of people in the building N; 3) air exchange rate k_{ex} ; 4) individual daily
253 inhalation rate (at home) IR, 5) daily time at home t_{home} (used to calculate the daily time fraction spent
254 indoors f_i). For the values of V and N the sampling method has been adapted to reflect the dependency
255 between these parameters: for each Monte Carlo run, a corresponding set of values for N and V for one
256 country was selected out of their discrete distribution over all countries, with a probability-weighting
257 based on its population. The average individual inhalation rate at rest for households was sampled from
258 the reported interval of 0.44-1.04 m³/h³⁶ assuming a beta distribution between these limits. The air
259 exchange rate (k_{ex}) was sampled from a discrete distribution representing L-AER and H-AER buildings
260 respectively from various countries using a probability-weighting based on their respective population.
261 The daily time at home was assumed to be normally distributed with an assumed standard deviation of
262 2, resulting in a 95% confidence interval ranging from 10 to 18 hours per day at home. For further
263 details and values the reader is referred to SI.

264 **Case Study**

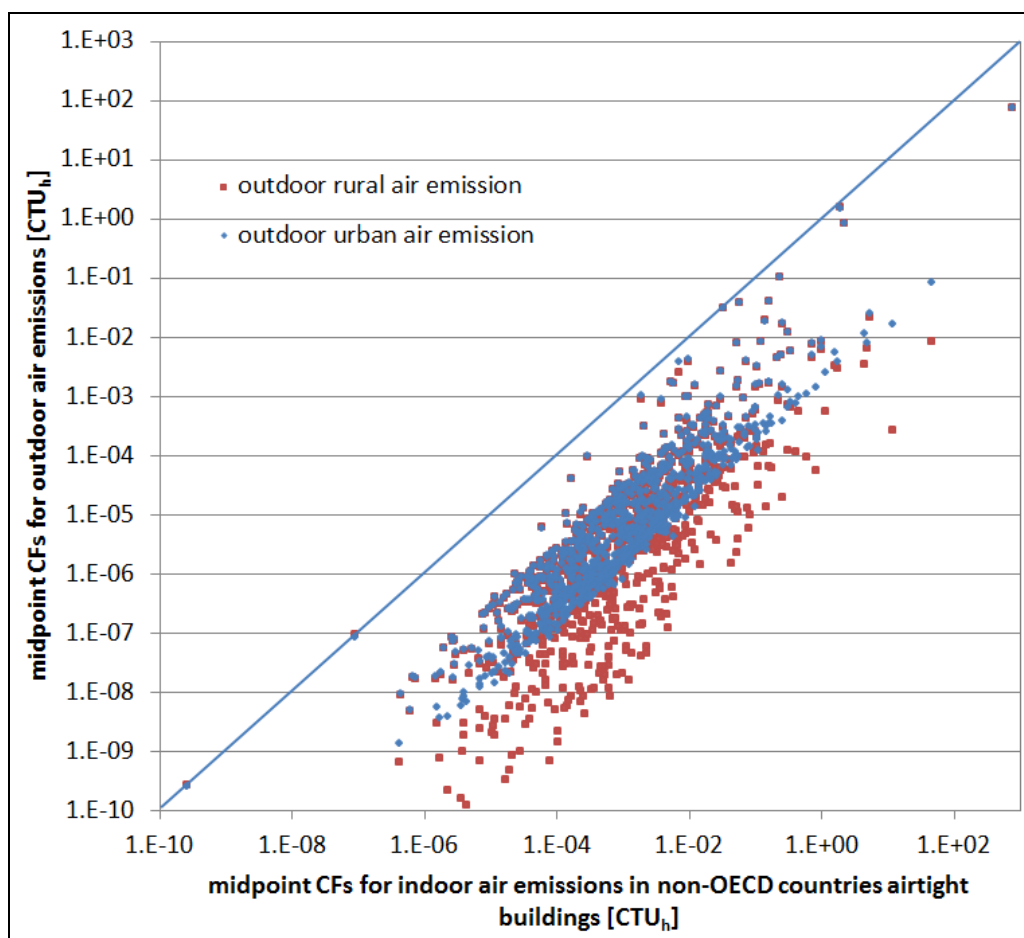
265 To illustrate the application of the method developed, an LCA of cooking in non-OECD countries was
266 performed. This case study was chosen for its relevancy: Air pollution originating from households
267 account for approximately 4% of global health burden and was the leading environmental health risk
268 factor.³⁷ The functional unit was defined as the delivery of 1 MJ of useful heat, delivered with stoves
269 based on different fuels: wood, charcoal, liquefied petroleum gas (LPG), and coal. These fuels are the
270 principal fuels being used in non-OECD countries; for example, in India 78% of the population lives in
271 houses where wood or LPG is used as main cooking fuel.³⁸ Background data for the fuel supply chain of
272 coal, charcoal and LPG were taken from the inventory database *ecoinvent*.³⁹ Wood was assumed to be
273 manually collected (no emissions from transport and harvesting), and only land use and the emissions
274 during combustion were accounted for. For the integrated toxicity assessment of indoor and outdoor
275 emissions, the USEtox outdoor model and effect factors (with integrated indoor model) were used

276 according to equation (2),¹³ extended to endpoint results expressed as Disability Adjusted Life Years
277 (DALY) using the following disability weights: 11.5 DALY/CTU_h for cancerous effects and 2.7
278 DALY/CTU_h for non-cancerous effects (CTU_h – Comparative Toxic Unit for humans¹³ corresponding
279 to cases of cancer or of non-cancer).⁴⁰ Respiratory inorganics impacts of PM_{2.5}, NO_x, SO_x and NH₃ were
280 estimated using the effect and characterization factors from Gronlund et al.⁴¹ The direct emissions are
281 displayed in Table S2 of SI together with further details on the background processes given in section
282 S2 of SI.

283 **Results**

284 **Intake fractions and characterization factors**

285 With the methodology described and the list of parameters given, intake fractions and characterization
286 factors for indoor exposure in residential settings (i.e. households) can be calculated for the defined
287 regions. For volatile substances, ventilation is the only sink in the indoor environment. Since ventilation
288 is chemical independent, no substance-related parameters are used in these calculations. Therefore, the
289 intake fractions for indoor exposure to volatile substances are the same for all substances and are given
290 in Table 2 for the defined regions. Due to the substance-dependency of the toxicity-effect factor, the
291 characterization factors for these substances vary among chemicals (Equation 2). The substance-specific
292 characterization factors for the USEtox chemical database are given in Excel format as part of SI for
293 946 substances. The characterization factors, in literature sometimes also referred to as comparative
294 toxicity potentials, vary over 12 orders of magnitude from least to most toxic and are up to five orders
295 of magnitude higher for household indoor emissions relative to continental rural emissions for the same
296 substance (see Figure 2). However, with future updates to the database, the characterization factors will
297 likely change. Therefore, future updates to the latest (indoor and outdoor) characterization factors will
298 be available on the USEtox website (www.usetox.org) and should always be taken from there.



299

300 Figure 2: Comparison of characterization factors (CFs) for indoor emissions in non-OECD countries
301 and L-AER buildings (x-axis) relative to CFs for continental urban and rural outdoor emissions (y-axis);
302 the difference between indoor and outdoor iFs is smaller for the other regions

303

304 The average house size in non-OECD countries is lower than that in OECD countries, and the average
305 household size is larger (see Table 1). Therefore, intake fractions in L-AER houses in non-OECD
306 countries are about three times higher than those in OECD countries. Intake fractions in H-AER houses
307 in non-OECD countries are a factor of 10 lower because of the higher ventilation rates (Table 1). The
308 results of the variability analysis of household indoor intake fractions are given as standard deviations in
309 Table 2. The variability within the regions is influenced by the amount of data available, which is much
310 lower for non-OECD compared to OECD countries, making those results somewhat less representative
311 for variability between countries.

312 Table 2: Intake fractions (iF) for household indoor exposure with standard deviations (SD) and results
313 of the importance analysis of the parameters used to calculate iF for the defined regions (negative
314 contributions represent an inverse correlation between parameter and result)

| Region | iF [-] | SD | IR/h | t _{home} | N/V | k _{ex} |
|-------------------------------------|----------------------|----------------------|--------------|-------------------|--------------|-----------------|
| Non-OECD countries (H-AER building) | 6.8·10 ⁻⁴ | 8.8·10 ⁻⁴ | 48% | 34% | -16% | -2% |
| Non-OECD countries (L-AER building) | 1.7·10 ⁻² | 1.6·10 ⁻² | 45% | 31% | -15% | -9% |
| OECD countries | 5.2·10 ⁻³ | 1.7·10 ⁻³ | 41% | 29% | -21% | -9% |
| Europe (EU-27) | 5.7·10 ⁻³ | 3.4·10 ⁻³ | 12% | 8% | -7% | -73% |
| North America (USA) | 4.6·10 ⁻³ | ^a | ^a | ^a | ^a | ^a |

315 ^asingle data point (US average) as we are using country averages and hence no variability assessed on sub-country level

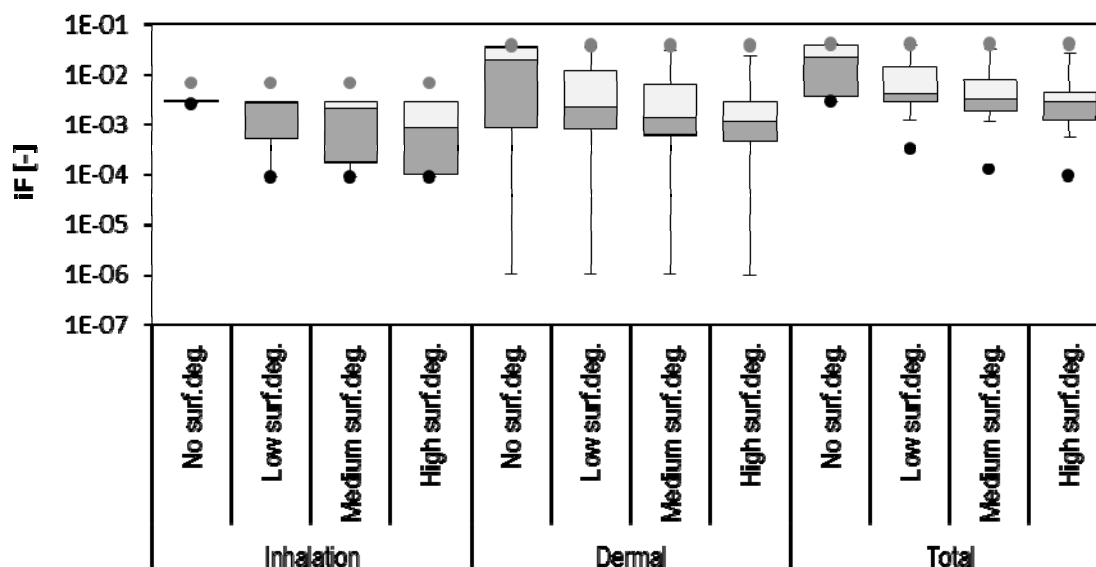
316

317 The results of the importance analysis are given in Table 2. For each region the contribution to total
 318 variance per parameter is given, providing an importance ranking of these parameters. Despite some
 319 variation in the percentage of contribution the ranking is the same for the OECD and Non-OECD
 320 scenarios. Due to the large variability in air-tightness of buildings within Europe, the air exchange rate
 321 varies the most and hence contributes the most to total variance of iF in this region with the remaining
 322 parameters ranking the same way as for the other regions.

323 For substances with significant indoor degradation (e.g. ozone-sensitive substances) or adsorption to
 324 surfaces (e.g. semi-volatile substances), the intake fraction is substance-specific.¹⁷ The intake fractions
 325 and characterization factors for these substances can be calculated using the USEtox model version 2.0.
 326 The sensitivity study carried out to determine the influence of degradation and surface adsorption
 327 delivers the following conclusions: Degradation plays a relatively minor role for the removal of
 328 substances emitted into indoor air, by increasing the removal rate by a maximum 20% (Figure S1, SI).
 329 The effect of adsorption on room surfaces may be more substantial, since it reduces inhalation intake
 330 fraction at high vapor pressure by up to a factor of 60 for substances like benzo[a]pyrene with vapor
 331 pressure below 1 Pa (Figure 3, first 4 columns, Figure S2, SI), even for degradation rates on surfaces as
 332 low as 1 per thousand of the air degradation (low surface degradation). On the contrary, dermal gaseous
 333 exposure uptake increases with the octanol-air partition coefficient K_{oa} and tends to compensate the
 334 reduction due to surface adsorption (Figure 3, 4 central columns) for substances with high K_{oa}, leading
 335 to a total intake with adsorption that is close to the default inhalation intake without adsorption.
 336 However, additional information is needed to better characterize surface adsorption and degradation and
 337 the way it may compensate the increase in dermal gaseous uptake, hence the choice to only consider

338 indoor air advective removal, degradation, and inhalation pathways in the default model at this stage.

339 More details on the sensitivity study can be found in section S3 of SI.



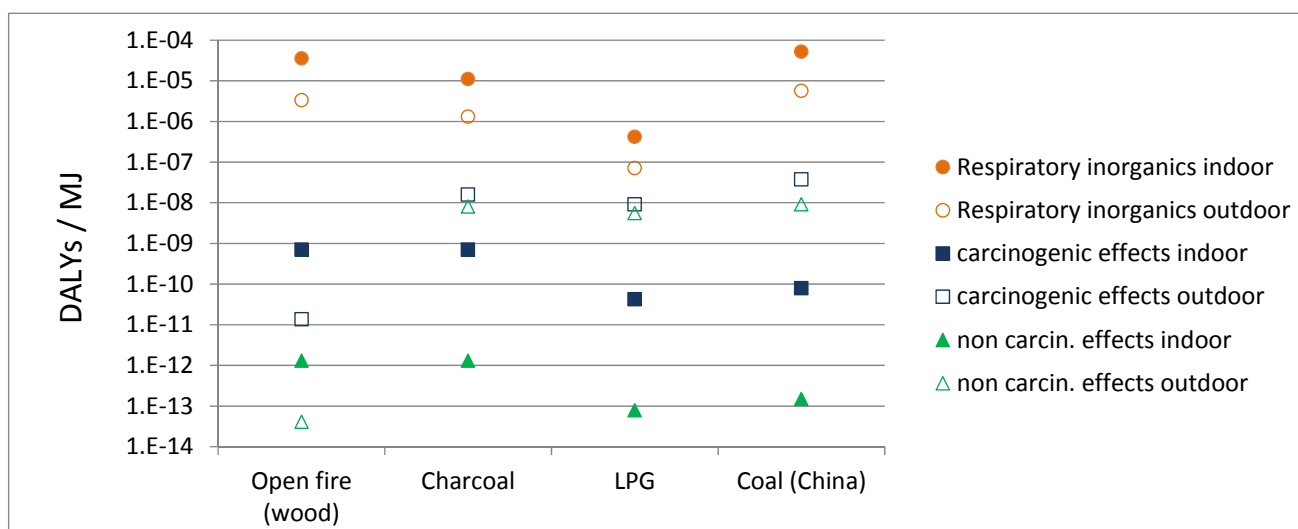
340

341 Figure 3: Variations in indoor intake fractions for the 3073 organic substances in the USEtox 1.01
342 database considering the inhalation, dermal gaseous and sum of these two exposure pathways with four
343 assumptions: No, low, medium, and high surface degradation rates following sorption, respectively
344 corresponding to surface degradation rates of 0, 0.001, 0.01 and 0.1 of the indoor air OH degradation
345 rate

346

347 Case study: world cooking

348 Figure 4 shows that the health impacts from indoor emissions are dominating the overall health
349 effects. Assuming equal weighting between cancer, non-cancer, and respiratory effects, the respiratory
350 effects from PM emissions represent clearly the most relevant effect for all cooking alternatives
351 analyzed. Total health impacts are more than one order of magnitude lower for cooking with gas
352 compared to charcoal and two orders of magnitude smaller compared to wood and coal.



353
354 Figure 4: Human health impacts in DALY from indoor and outdoor exposure

355 Discussion

356 The framework to calculate intake fractions and characterization factors for indoor exposure to
357 substances in households in the USEtox model is described. With this framework and the recommended
358 parameter values given, the iF and characterization factors for household indoor exposure to substances
359 can be calculated for different regions. However, given the uncertainties behind these estimates, the iF
360 for OECD countries, Europe, and North America are essentially equal (Table 2) and we recommend
361 using the OECD value for Europe and North America as well. It should be noted that the distinction
362 between L-AER and H-AER buildings is a strongly simplified, binary classification due to lack of more
363 detailed data. These two classes essentially distinguish between 1) basic constructions ranging from
364 buildings with simple or no sealing and cracked walls to huts or tents without windows and/or doors (H-
365 AER) as opposed to 2) fairly modern buildings eventually with ventilation systems, sealing and
366 insulation (L-AER), which is how they should be used in LCA practice.

367 The observed differences in iF of almost two orders of magnitude between the regions (Table 2) are
368 caused by differences in ventilation rate, building occupation and volume. The dermal absorption of
369 gas-phase chemicals may become important in particular for SVOCs and the calculated intake fractions
370 must be used with care for this class of compounds, as these will require further attention, both for their
371 adsorption and potential degradation rates on surfaces and for dermal uptake.

372 The USEtox intake fractions for inhalation exposure to outdoor emissions range from $3 \cdot 10^{-6}$
373 (continental urban air emission) and $7 \cdot 10^{-9}$ (continental rural air emission) respectively for dioxathion
374 (CAS 78-34-2), and up to $3 \cdot 10^{-4}$ (for continental urban and rural air emission) for 1,1,1,2-
375 tetrafluoroethane (CAS 811-97-2). The intake fractions for indoor air emissions as given in Table 2 are
376 thus at least two and up to seven orders of magnitude higher than the intake fractions for outdoor air
377 emissions.

378 With the indoor exposure model implemented in USEtox and the resulting characterization factors, it
379 is now operational to integrate household indoor exposure to substances into life cycle assessment
380 studies. Both, iF and characterization factors calculated in this study are based on the still sparse data
381 sources available, which highly influenced the number of regions that could be defined. When more
382 data become available the definitions of regions should be revised in order to better represent global
383 variability, and the iF and characterization factors should be updated. Meanwhile, the parameters in
384 Table 1 for the OECD and non-OECD scenarios are recommended for LCA application of Hellweg et
385 al.'s one-box indoor exposure model. Since the present intake fractions are based on average occupancy
386 and continuous emission, further efforts are needed in the future to better assess emissions with non-
387 continuous sources related to the nexus of occupant and source activity patterns (e.g. cooking), in
388 particular emission patterns that involve near-person releases. Another refinement would be to account
389 for substance removal by filters in centrally air-conditioned buildings, a region-specific removal rate
390 that may be substantial in hot climate. Moreover, whereas degradation was not an important removal
391 process we underline that impacts from the products of homogenous reactions in air or other
392 degradation processes may have significant impacts⁴²⁻⁴⁴ but are not taken into account in the CFs
393 calculated by this research work. According to current practice, LCA practitioners can take them into
394 account by adding the amount of reaction products generated from a parent compound to the life cycle
395 emission inventory and characterize them with their corresponding characterization factors.

396 The case study on cooking in non-OECD countries demonstrates the appropriateness and significance
397 of integrating indoor environments into LCA. Approximately 2.4 billion people, concentrated largely

398 within low- and middle-income countries,⁴⁵ continue to rely on solid fuels as main sources of household
399 energy without access to clean energy or appropriate technologies to prevent exposure to harmful levels
400 of indoor air pollutants from inefficient burning of biomass fuels.⁴⁶ The results of the case study confirm
401 that health impacts from indoor exposure are relevant. Neglecting these impacts would have provided
402 an incomplete and misleading picture: While cooking with wood would have performed best if only the
403 outdoor emissions were considered (as usually done in LCA), it was the worst alternative after coal if
404 health impacts from indoor exposure were considered. Given the current limits in data availability to
405 parameterize the indoor exposure model for the most affected regions, more robust datasets will likely
406 increase the discrimination of baseline and proposed alternatives. Thus, incorporating the indoor
407 environment in LCA ensures a more holistic consideration of all exposure environments and allows for
408 a better accountability of health impacts. Furthermore, while developing countries transition towards
409 more processed fuels (e.g. petroleum, or electricity from coal), the holistic approach of LCA remains
410 relevant and necessary for assessing both health and environmental implications.

411 Databases providing emission data for different materials, products, and surfaces are an essential
412 element needed towards operationalization of indoor exposure assessment within LCA. Currently,
413 indoor emission data are not widely available or not in a suitable format for LCA (e.g. given as
414 concentrations whereas emitted mass or emission rates would be required to link with our model
415 results).

416 Adapting current tools, such as the USEtox toxicity characterization model, by investigating their
417 applicability under various situations and providing regional specific parameters, allows for identifying
418 “hot-spots” of disease burdens as well as pointers for solutions using a consistent and transparent
419 method. This study, using an illustrative case of cooking, quantified indoor intake fractions for
420 households in various regions of the world that differ geographically, economically, and socially, and
421 provided information on the impact that human behavior, energy use, and technology can have on
422 human health. The modification to the USEtox model, with the integration of the indoor environment, is
423 part of the official update to USEtox version 2.0 and can contribute in providing a clearer assessment of

424 the source of burden of disease and provide a more informed basis for decision making for all
425 stakeholders.

426 **Supporting information**

427 Parameter values for the individual countries, inventory data for case study on cooking, sensitivity of
428 iF to degradation rates and adsorption on surfaces (PDF), intake fractions, effects factors and
429 characterization factors for household indoor air emissions for three regions calculated with USEtox
430 (Excel). This material is available free of charge via the Internet at <http://pubs.acs.org>.

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