



## Characterizing Aggregated Exposure to Primary Particulate Matter: Recommended Intake Fractions for Indoor and Outdoor Sources

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## Article

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1 **Characterizing Aggregated Exposure to Primary Particulate Matter:**

2 **Recommended Intake Fractions for Indoor and Outdoor Sources**

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25

## 26 **Abstract**

27 Exposure to fine particulate matter (PM<sub>2.5</sub>) from indoor and outdoor sources is a  
28 leading environmental contributor to global disease burden. In response, we established under  
29 the auspices of the UNEP/SETAC Life Cycle Initiative a coupled indoor-outdoor emission-to-  
30 exposure framework to provide a set of consistent primary PM<sub>2.5</sub> aggregated exposure factors.  
31 We followed a matrix-based mass balance approach for quantifying exposure from indoor and  
32 ground-level urban and rural outdoor sources using an effective indoor-outdoor population  
33 intake fraction and a system of archetypes to represent different levels of spatial detail.  
34 Emission-to-exposure archetypes range from global indoor and outdoor averages, via  
35 archetypal urban and indoor settings, to 3646 real-world cities in 16 parameterized sub-  
36 continental regions. Population intake fractions from urban and rural outdoor sources are  
37 lowest in Northern regions and Oceania and highest in Southeast Asia with population-  
38 weighted means across 3646 cities and 16 sub-continental regions of, respectively, 39 ppm  
39 (95% confidence interval: 4.3–160 ppm) and 2 ppm (95% confidence interval: 0.2–6.3 ppm).  
40 Intake fractions from residential and occupational indoor sources range from 470 ppm to  
41 62,000 ppm, mainly as function of air exchange rate and occupancy. Indoor exposure  
42 typically contributes 80–90% to overall exposure from outdoor sources. Our framework  
43 facilitates improvements in air pollution reduction strategies and life cycle impact  
44 assessments.

45

46 **Keywords:** PM<sub>2.5</sub>, intake fraction, emission-to-exposure framework, exposure archetypes,  
47 global guidance, air pollution, LCIA

48

## 49 Introduction

50 Over the last three decades, multiple epidemiological and toxicological studies have  
51 attributed a range of adverse health impacts including chronic and acute respiratory and  
52 cardiovascular diseases and premature mortality to exposures to fine particulate matter (PM<sub>2.5</sub>,  
53 representing particles with aerodynamic diameter of 2.5 μm or smaller) both outdoors and  
54 indoors. In the Global Burden of Disease (GBD) study series, exposure to PM<sub>2.5</sub> is identified  
55 as a leading environmental risk factor contributing to global human disease burden. PM<sub>2.5</sub> in  
56 outdoor air and household air is reported to contribute to estimated 4.2 and 2.9 million  
57 premature deaths, respectively, corresponding to 103 and 86 million disability-adjusted life  
58 years (DALY), respectively, in 2015.<sup>1,2</sup> Indoor and outdoor emissions of primary PM<sub>2.5</sub> from  
59 anthropogenic sources contribute substantially to human exposures, which take place both  
60 indoors and outdoors. Outdoor emissions in urban and rural areas are mainly associated with  
61 road traffic including fuel combustion-related vehicle exhaust and road dust, coal- and gas-  
62 fired power plants, and other industrial sources.<sup>3,4</sup> Indoor emissions in residential,  
63 commercial, and occupational settings are mainly from combustion processes (e.g., cooking,  
64 smoking, candles). Approximately 2.8 billion people, primarily in Africa and Southeast Asia,  
65 are exposed to indoor emissions from the use of solid fuels including coal, charcoal, wood,  
66 dung, and crop residues, with substantial impacts on both indoor and outdoor air quality.<sup>5-8</sup>

67 To inform decisions for comparing and reducing PM<sub>2.5</sub> exposure from anthropogenic  
68 sources, a quantitative framework is required to link indoor and outdoor environments.  
69 Multiple studies have monitored PM<sub>2.5</sub> concentrations outdoors<sup>9,10</sup> and indoors,<sup>11,12</sup> and  
70 estimated related inhalation exposure outdoors<sup>13,14</sup> and indoors.<sup>15,16</sup> Intake fractions  
71 (population inhalation intake per emission unit) have been determined as related exposure  
72 metric either for indoor or for outdoor urban or rural environments.<sup>14,17,18</sup> However, a  
73 consistently coupled indoor-outdoor exposure assessment framework is currently missing that  
74 allows for comparing PM<sub>2.5</sub>-related intake fractions from a range of human activities that lead

75 to outdoor and indoor sources resulting in human exposures to PM<sub>2.5</sub> both indoors and  
76 outdoors. According to earlier recommendations,<sup>19</sup> such a framework needs to (a) integrate  
77 indoor and outdoor air on a consistent mass-balance basis, thereby accounting for multiple  
78 emission sources along product system life cycles, (b) distinguish among relevant emission  
79 and exposure scenarios in different indoor, urban and rural outdoor environments, (c)  
80 conceptually integrate indoor and outdoor exposure as starting point for linking exposure  
81 levels to exposure-response considering that humans spend most of their time indoors,<sup>18</sup> (d)  
82 build on an archetypal structure to capture variability in PM<sub>2.5</sub> air concentrations and  
83 population density among different indoor and urban- and rural-outdoor environments, and (e)  
84 incorporate uncertainty into results at different levels of detail. Hodas et al.<sup>18</sup> and Milner et  
85 al.<sup>20</sup> further underline the need to include indoor PM<sub>2.5</sub> in exposure estimates and to consider  
86 distinct archetypes to capture important differences among indoor environments and building  
87 types. For outdoor scenarios, spatial approaches are unable to capture higher exposure in  
88 urban areas, unless they build on grid-resolutions that allow distinguishing between urban and  
89 rural environments in all regions, i.e. using resolutions on the order of at least 0.1°.<sup>21,22</sup> For  
90 example, although intake fractions based on global, spatially gridded 1° × 1° PM<sub>2.5</sub> outdoor  
91 air concentrations are estimated to only vary between 1.6 and 9.6 ppm,<sup>23</sup> intraurban intake  
92 fractions estimated globally for all cities with more than 100,000 inhabitants reach 260 ppm  
93 with a population-weighted average of 39 ppm.<sup>14</sup> Archetypes are best capable of capturing  
94 relevant differences between urban and rural areas, where city-specific intake fractions (e.g.  
95 Apte et al.<sup>14</sup>) need to be integrated into a background continental environment and account for  
96 the fact that the population spends most of its time indoors.<sup>18</sup> Understanding the interaction  
97 between indoor and outdoor environments is also important (for example, when exposure-  
98 response functions obtained in a region with low indoor air exchange rates are applied to  
99 regions with substantially higher air exchange rates). Therefore, a modeling framework is

100 needed that accounts for various indoor and outdoor settings, interactions between urban and  
101 rural areas, and operates at multiple scales of integration, while capturing high variability.<sup>24</sup>

102 In response to these needs, the United Nations Environment Program/Society of  
103 Environmental Toxicology and Chemistry (UNEP/SETAC) Life Cycle Initiative established a  
104 task force to provide guidance for quantifying health effects from PM<sub>2.5</sub> exposure associated  
105 with indoor and outdoor sources for use in life-cycle-based impact assessments. The aim is to  
106 compare human activities and product systems with respect to their contribution to PM<sub>2.5</sub>  
107 emission related disease burden.<sup>19,25,26</sup> As a first step toward such a PM<sub>2.5</sub> impact assessment  
108 framework, we aim in the present paper at characterizing for primary PM<sub>2.5</sub> the intake  
109 fraction, which is the long-term population intake mass per unit mass emitted into different  
110 indoor and outdoor environments. Building on the rich literature on PM<sub>2.5</sub> exposure research,  
111 we organize the present work as follows: First, we structure the PM<sub>2.5</sub> emission-to-intake  
112 pathway into a system of archetypes representing a tiered approach following different levels  
113 of detail for indoor and urban- and rural-outdoor environments. Levels of detail range from  
114 generic (global average) level to city-specific level, representing 3646 real-world cities and a  
115 set of residential and occupational indoor environments. Second, we describe our system as a  
116 fully mass balance based framework for relating indoor and outdoor emissions to aggregate  
117 PM<sub>2.5</sub> exposure. Third, we analyze the variations of intake fraction among different emission  
118 locations in our framework as a function of advection rates and population densities, based on  
119 differentiating for each source scenario the contribution of each environment to overall  
120 population exposure. Finally, we discuss how the proposed framework is aligned with state-  
121 of-the-art indoor- and outdoor-exposure models, and how it can be consistently coupled with  
122 exposure-response information.

123

## 124 **Methods**

### 125 **Coupled indoor and outdoor source-to-exposure framework**

126 Environmental fate and transport processes of PM<sub>2.5</sub>, linking emissions in different  
127 indoor or outdoor environments to human inhalation exposure indoors and outdoors, are  
128 represented by a mass balance system of homogeneous air compartments (**Figure 1**) described  
129 by a set of first-order differential equations. In order to address PM<sub>2.5</sub> emissions, and complex  
130 issues, such as spatially heterogeneous concentrations in urban environments and different  
131 applications of exposure-response functions in indoor, urban, and rural environments based  
132 on earlier recommendations,<sup>19</sup> we made several modifications to existing fate modeling  
133 approaches. Most importantly, we couple indoor and outdoor environments, incorporate  
134 inhalation as a removal process in the fate model (in addition to using inhalation in the  
135 exposure model), and capture exposure-related variability among different indoor, and urban-  
136 and rural-outdoor environments. We address variability using a set of interconnected  
137 archetypal environments ranging outdoors from global averages of urban conditions to 3646  
138 specific cities and different indoor settings. We finally provide the basis for consistently  
139 linking both indoor and outdoor exposure to exposure-response.

140

141 <**Figure 1**>

142

143 The overall source-to-exposure modeling framework builds on four main  
144 compartments, namely outdoor and indoor environments in urban and rural areas, where both  
145 indoor and outdoor urban environments are nested within rural areas. **Figure 1** provides a  
146 general illustration of the PM<sub>2.5</sub> transport and loss processes considered in and between all  
147 compartments. For consistency and completeness we build for each compartment a mass  
148 balance equation that addresses emissions; deposition to soil, water, and vegetation surfaces  
149 outdoors; advection losses outdoors (including losses beyond the continental rural boundaries

150 to the global atmosphere); transfers between outdoor air and indoor air; deposition to surfaces  
 151 indoors; removal from indoor environments by cleaning and filtration; and removal by  
 152 inhalation indoors and outdoors.

153

#### 154 **Overall emission-to-impact matrix system**

155 Exposure-pathway-specific PM<sub>2.5</sub> intake fractions relate the population inhaled mass  
 156 of PM<sub>2.5</sub> to the mass emitted and provide the exposure information for the impact assessment  
 157 framework. Intake fractions are calculated from combining PM<sub>2.5</sub> removal via inhalation  
 158 (exposure factors) with PM<sub>2.5</sub> transfer and removal from air (fate factors):

$$159 \quad \mathbf{iF} = \mathbf{XF} \mathbf{FF} = \mathbf{XF} (-\mathbf{K}^{-1}) \quad (1)$$

160 where matrix  $\mathbf{XF} \in \mathbb{R}^{p \times n}$  contains exposure factors expressed as PM<sub>2.5</sub> removal rate  
 161 coefficients (further detailed in [Table 1](#)) via inhalation with exposure pathways in rows and  
 162 receptor compartments in columns, and matrix  $\mathbf{FF} \in \mathbb{R}^{n \times n}$  contains fate factors representing  
 163 PM<sub>2.5</sub> mass received in receptor compartments (rows) per unit emissions into source  
 164 compartments (columns).  $\mathbf{FF}$  main diagonal elements represent PM<sub>2.5</sub> residence times,  
 165 accounting for all multiple inter-compartment transfers between the different indoor and  
 166 outdoor environments.<sup>18</sup> This allows for assessing not only exposure in the indoor or urban-  
 167 outdoor emission compartments, but also subsequent exposure after transfer to the continental  
 168 rural environment, which may be especially relevant for small cities. Fate factors for PM<sub>2.5</sub> in  
 169 eq. 1 under steady-state conditions are obtained from inverting the matrix of rate coefficients  
 170  $\mathbf{K} \in \mathbb{R}^{n \times n}$  describing transfers between adjacent compartments and removal (i.e. deposition  
 171 and inhalation) within compartments. In our framework, matrix  $\mathbf{K}$  consists of elements (rate  
 172 coefficients,  $k$ ) representing for outdoor emissions environmental processes within and  
 173 between four compartments, namely default indoor air in urban (denoted ‘i,u’) and rural (‘i,r’)  
 174 areas and scenario-specific outdoor air in urban (‘o,u’) and rural (‘o,r’) areas, and for indoor  
 175 emissions environmental processes in a scenario-specific residential or occupational indoor

176 environment. Compartments are further detailed in the next section. Each main diagonal  
 177 element of **K** represents the bulk removal or loss via all considered processes (denoted ‘loss’  
 178 and per convention negative to indicate losses) in the respective compartment and all other  
 179 non-zero non-diagonal elements represent individual intercompartmental transfers.  
 180  
 181 **Table 1.** Equations to calculate fate-related rate coefficients, human exposure factors, and  
 182 underlying parameters used in the PM<sub>2.5</sub> matrix framework. Default and constant inputs are  
 183 provided in the Supporting Information.

Variable or parameter	Equation <sup>a</sup>
<i>Coefficients for outdoor air compartment in urban areas</i>	
Bulk removal (loss) rate coefficient from outdoor air in urban areas	$k_{o,u,loss} = k_{i,u \leftarrow o,u} + k_{o,r \leftarrow o,u} + k_{o,u,dep} + k_{o,u,att} + XF_{o,u}$
Transfer rate coefficient from outdoor urban air to indoor air in urban areas	$k_{i,l \leftarrow o,u} = \frac{k_{o,u \leftarrow i,u} \times P_u \times V_{i,u}}{V_{o,u}}$
Transfer rate coefficient from outdoor urban air to outdoor rural air	$k_{o,r \leftarrow o,u} = \frac{DR_{o,u}}{h_{o,u} \times \sqrt{A_{o,u}}} \times f_{u,corr}$
Removal rate coefficient from outdoor urban air via bulk deposition	$k_{o,u,dep} = \frac{v_{o,u,dep}}{h_{o,u}}$
Removal rate coefficient from outdoor urban air via indoor attenuation	$k_{o,u,att} = \frac{ACH_u \times (1 - P_u) \times V_{i,u}}{V_{o,u}}$
Removal rate coefficient (exposure factor) from outdoor urban air via inhalation	$XF_{o,u} = \frac{BR_o \times (1 - f_{t,i}) \times POP_u}{V_{o,u}}$
<i>Coefficients for outdoor air compartment in rural areas</i>	
Bulk removal (loss) rate coefficient from outdoor rural air	$k_{o,r,loss} = k_{i,r \leftarrow o,r} + k_{o,u \leftarrow o,r} + k_{o,r,glob} + k_{o,r,dep} + k_{o,r,att} + XF_{o,r}$
Transfer rate coefficient from outdoor rural air to indoor air in rural areas	$k_{i,r \leftarrow o,r} = \frac{k_{o,r \leftarrow i,r} \times P_r \times V_{i,r}}{V_{o,r}}$
Transfer rate coefficient from outdoor rural air to outdoor urban air	$k_{o,u \leftarrow o,r} = \frac{k_{o,r \leftarrow o,u} \times V_{o,u}}{V_{o,r}}$
Advective loss rate coefficient from outdoor rural air to global air	$k_{o,r,glob} = \frac{u_{o,r}}{\sqrt{A_{o,r}}}$
Removal rate coefficient from outdoor rural air via bulk deposition	$k_{o,r,dep} = \frac{v_{o,r,dep}}{h_{o,r}}$

Variable or parameter	Equation <sup>a</sup>
Removal rate coefficient from outdoor rural air via indoor attenuation	$k_{o,r,att} = \frac{ACH_r \times (1 - P_r) \times V_{i,r}}{V_{o,r}}$
Removal rate coefficient (exposure factor) from outdoor rural air via inhalation	$XF_{o,r} = \frac{BR_o \times (1 - f_{t,i}) \times POP_r}{V_{o,r}}$
<i>Coefficients for indoor air compartments in urban and rural areas*</i>	
Bulk removal (loss) rate coefficient from indoor air	$k_{i,loss} = k_{o \leftarrow i} + k_{i,dep} + k_{i,circ} + XF_i$
Transfer rate coefficient from indoor air to outdoor air	$k_{o \leftarrow i} = ACH$
Removal rate coefficient from indoor air via bulk deposition	$k_{i,dep} = v_{i,dep} \times (S/V_i)$
Removal rate coefficient from indoor air via filtration of recirculated air	$k_{i,circ} = k_{recirc} \times \varepsilon_{filter}$
Removal rate coefficient (exposure factor) from indoor air via inhalation	$XF_i = \frac{BR_i \times f_{t,i} \times POP}{V_i}$
<i>Parameter used to calculate fate-related rate coefficients and human exposure factors</i>	
Area of urban areas within region $x$	$A_{o,u} = (POP_u/LPD)^2$
Volume of indoor air in urban areas	$V_{i,u} = V_{i,u,pers} \times POP_u$
Volume of indoor air in rural areas	$V_{i,r} = V_{i,r,pers} \times POP_r$
Volume of outdoor air in urban areas	$V_{o,u} = A_{o,u} \times h_{o,u}$
Volume of outdoor air in rural areas	$V_{o,r} = A_{o,r} \times h_{o,r}$
Correction factor accounting for city-specific dynamics in area and dilution rate	$f_{u,corr} = 4.95 \times A^{0.0508} \times DR^{-0.124}$
Penetration factor from outdoor urban air to indoor air in urban areas	$P_u = F_{u,inf} \times \frac{k_{i,u,dep} + XF_{i,u} + ACH_u}{ACH_u}$
Penetration factor from outdoor rural air to indoor air in rural areas	$P_r = F_{r,inf} \times \frac{k_{i,r,dep} + XF_{i,r} + ACH_r}{ACH_r}$

184 \*Indoor environment equations apply to both urban and rural environments, where relevant  
 185 parameters, such as population, are environment-specific. <sup>a</sup> $A$ : air cross section area [m<sup>2</sup>] (see  
 186 eq. 2 for archetypal city areas);  $ACH$ : air exchange rate [d<sup>-1</sup>];  $BR$ : breathing rate [m<sup>3</sup>/d];  $DR$ :  
 187 normalized atmospheric dilution rate [m<sup>2</sup>/d];  $f_{t,i}$ : fraction of time per day spent indoors [-];  
 188  $f_{u,corr}$ : correction factor accounting for city-specific dynamics in area and dilution rate [-];  
 189  $F_{inf}$ : infiltration factor representing an indoor/outdoor air concentration ratio in the absence of  
 190 indoor sources that is obtained from dividing elements of the fate factors matrix and volumes  
 191 of the respective indoor and outdoor compartments [-];  $h$ : atmospheric mixing height [m];  $k$ :  
 192 first order rate coefficient for individual transfer or bulk removal processes [d<sup>-1</sup>];  $LPD$ : linear  
 193 population density [capita/m] based on Figure 2;  $P$ : penetration factor from outdoor to indoor  
 194 air [-];  $POP$ : human population count [capita];  $S/V_i$ : total material area to air volume ratio  
 195 indoors [m<sup>2</sup>/m<sup>3</sup>];  $V$ : air volume [m<sup>3</sup>];  $v_{dep}$ : bulk deposition velocity combining deposition to  
 196 vertical and upward-facing and downward-facing surfaces [m/d];  $u$ : mean wind speed at

197 ground-level [m/d];  $XF$ : human inhalation exposure factor [ $d^{-1}$ ]. Indices: i, o, r, u denote  
198 indoor, outdoor, rural, and urban, respectively; att, circ, dep, filter, inf, glob, loss, pers, recirc  
199 refer to indoor attenuation, air circulation, bulk deposition, air filter, infiltration, global air,  
200 bulk removal or loss, per individual person, and air recirculation, respectively; and arrows  
201 between indices indicate inter-compartment transfer processes.  
202

### 203 **Determination of rate constants**

204 We focused on adapting and consistently integrating our model elements from existing  
205  $PM_{2.5}$  transport and exposure studies. For addressing transport and fate in outdoor air, we  
206 build on an earlier consensus effort<sup>17</sup> and work by Apte et al.,<sup>14</sup> using respectively a set of  
207 parameterized generic (urban and rural) and city-specific archetypes at the global scale. Both  
208 studies developed the foundations for our exposure assessment for  $PM_{2.5}$  in outdoor air  
209 compartments. While Apte et al. provide a set of theoretical outdoor intake fractions for  
210 ground-level emissions in the 3646 cities globally with more than 100,000 inhabitants, we  
211 linked urban areas to the rural background using 16 sub-continental regions parameterized by  
212 Kounina et al.<sup>27</sup> This ensures full integration of the outdoor environment, while capturing  
213 important differences in  $PM_{2.5}$  air concentrations and related intake fractions across urban  
214 areas and between urban and rural areas. City-specific dynamics related to area and dilution  
215 rate were considered by adjusting the transfer from outdoor urban to rural air in a correction  
216 factor as a function of area and dilution rate across cities (see [Table 1](#)). This correction factor  
217 improves the correlation between intake fraction and the rate coefficient linking the urban  
218 area to its background rural environment compared to a direct transfer based only on dilution  
219 rate and size of the urban area.<sup>14</sup> Since our initial correlation makes the fate factor dependent  
220 on population density, we recalculated a new correlation of comparable accuracy ( $R^2 = 0.96$ ,  
221 instead of  $R^2 = 0.99$ ) that uses only the city-specific area and dilution rate as independent  
222 variables without involving the linear population density. How we obtained the correction  
223 factor is further detailed in the [Supporting Information \(SI\), Section S-2](#). Differences in  
224 atmospheric mixing height are linked to lower dilution rates in urban areas at night and the

225 short residence time of the air in urban areas of only a few hours compared to a longer  
226 residence time of air in rural areas of several days, which allows for mixing between day and  
227 night over the full mixing height.

228 For fate factors for indoor emission scenarios, we calculate indoor-to-outdoor transfer  
229 fractions for the default outdoor environment, obtaining the removal rates of PM<sub>2.5</sub> in  
230 different residential and occupational indoor archetypes as a function of ventilation,  
231 occupancy, and recirculation/filter efficiency. As a starting point for our indoor transport and  
232 fate model, we build on key studies by Thatcher and Layton,<sup>28</sup> Riley et al.,<sup>29</sup> and Bennett and  
233 Furtaw.<sup>30</sup> For the subsequent exposure assessment indoors and, in particular for exposure  
234 from indoor emissions, we build on work by Klepeis et al.<sup>31</sup> and Weschler and Nazaroff.<sup>32</sup> In  
235 our multimedia framework, we also account for transfer and related exposure to sources  
236 emitted elsewhere, primarily building on work by Diapouli et al.,<sup>33</sup> Riley et al.,<sup>29</sup> Thatcher and  
237 Layton,<sup>28</sup> Hänninen et al.,<sup>34</sup> and Meng et al.,<sup>35</sup> assessing PM<sub>2.5</sub> exposure indoors attributable  
238 to outdoor sources and PM<sub>2.5</sub> exposure outdoors attributable to indoor sources. All rate  
239 coefficients are further detailed in [Table 1](#), while default model settings are detailed in the [SI](#)  
240 ([Tables S1-S2](#)).

241 We determine exposure factors from indoor and outdoor breathing rates, the fraction  
242 of time spent indoors and outdoors, and air volume and population in each compartment,  
243 characterizing the fraction of air volume inhaled per day by the compartment-specific  
244 population. To arrive at aggregated exposure, intake fractions are calculated separately for  
245 each of the interlinked compartments accounting for exposure in all four environments (see  
246 [Figure 1](#)). For indoor exposure from emissions outdoors in a given region, we parameterize  
247 the indoor environment according to the average or most prominent air exchange rate and  
248 occupancy in the considered region. For studying emissions in a specific indoor environment  
249 when air exchange rates and building occupancies differ from the typical values in the  
250 considered region, we created a decoupled indoor model for first calculating the intake

251 fraction attributable to indoor emissions. We then add to this indoor intake fraction the  
252 fraction of the indoor emission transferred to the outdoor environment multiplied by the  
253 average outdoor intake fraction for the considered region to yield the overall effective intake  
254 fractions from indoor sources. The resulting mass-balanced fate and exposure model provides  
255 a mathematical framework that builds on state-of-the-art approaches for indoor and outdoor  
256 exposure assessment. Accounting for variability within our considered compartments, we  
257 introduce criteria described in the following paragraphs for defining consistent sets of  
258 archetypal environments for each compartment at generic, regional/intermediary and city-  
259 specific levels.

260

### 261 **Defining archetypal exposure environments at different levels of detail**

262 We propose a system of archetypes at different levels of detail that provide a higher  
263 level of resolution than can be achieved with currently available spatial models. Different  
264 levels of detail help to provide exposure estimates that are consistent with available data  
265 resolution in different decision contexts. As an example, archetypes at a generic (world  
266 average) level are required when emission source location or other scenario details like  
267 population density are unknown,<sup>17</sup> while archetypes at the city level are useful when details  
268 about city-specific urban emissions and population density are available.<sup>14</sup> Criteria for  
269 identifying a suitable set of archetypes for each compartment and level of detail help to  
270 differentiate and explain variability in emission situations, environmental conditions and  
271 human exposure.

272 In outdoor environments, there is a strong correlation between emission source  
273 strength and population density, where it has been shown that intake fractions for PM<sub>2.5</sub>  
274 emissions from roadways and low stacks can be significantly underestimated by models  
275 without very high resolution (at km scale or finer) emissions-to-population mapping.<sup>14,36</sup>  
276 However, source-specific data on emissions are often unavailable at spatial scales required to

277 account for population heterogeneity across large regions. Archetypes therefore need to  
278 capture the essential variability and heterogeneity for providing reliable outdoor intake  
279 fraction estimates.

280 For a given region, the population and area of a representative city must be defined to  
281 match the urban population-weighted average intake fraction of this region, using a  
282 population-weighted harmonic mean of the urban atmospheric dilution rate across cities with  
283 available data. The relation between population and area must be consistent and reflect the  
284 typical population density in cities of a region. This is ensured by studying the region-specific  
285 linear population density,  $LPD$  [capita/m], which links city area to population in order to  
286 determine intake fraction.<sup>37</sup> To define region-specific city archetypes, we first establish how  
287  $LPD$  varies across cities  $i$  as a function of urban population,  $POP$  [capita], within each region  
288  $x$ , with  $i \in x$ , by fitting a general model  $\log(LPDi) = \alpha_x + \beta \times \log(POP_i)$  based on 3646  
289 cities from Apte et al.<sup>14</sup> (see SI, Table S3). Once, this relation is established, we can derive the  
290 city area,  $A$  [m<sup>2</sup>], that corresponds to a given  $POP$  as follows (see SI, Section S-3):

$$291 \quad A = (POP/LPD)^2 = (10^{-\alpha_x} \times POP_u^{1-\beta})^2 \quad (2)$$

292 We distinguish outdoor archetypes at three levels of detail: First, a generic *level 1* is defined  
293 for situations where emission location or conditions are unknown, reflecting a population-  
294 weighted average intake fraction of 39 ppm across 3646 cities.<sup>14</sup> At finer levels of detail,  
295 additional aspects to discriminate intake fractions from outdoor sources are needed, such as  
296 different air exchange rates and occupancy levels for indoor environments, city size, spatially  
297 differentiated meteorological conditions (dilution rates defined from mixing height and wind  
298 speed), and population distribution in relation to emission source distribution for outdoor  
299 environments. At intermediate detail *level 2*, we define average cities to represent urban areas  
300 at the level of continental and sub-continental regions ensuring consistency between  
301 population, area, and exposure by calculating *level 2* outdoor intake fractions as population-  
302 weighted averages to provide a surrogate for emission-weighted averages in line with

303 Humbert et al.<sup>17</sup> and Lobscheid et al.<sup>36</sup> Finally, if emission scenario information is available  
304 for specific cities, we define respective archetypes at *level 3* to reflect PM<sub>2.5</sub> fate and exposure  
305 conditions as precisely as possible, building on available intraurban outdoor intake fractions  
306 for 3646 global cities parameterized for city-specific population, area, dilution rate, and PM<sub>2.5</sub>  
307 background concentration,<sup>14</sup> and combining these with population, area, and wind speed,  
308 based on high-resolution spatial data<sup>13</sup> for rural environments.

309 For indoor environments, exposure is strongly dependent on air exchange and  
310 available volume per person (occupancy).<sup>18</sup> However, building-specific air exchange and  
311 occupancy are usually not available at the level of detail required to account for variabilities  
312 across residential and occupational building types in different regions.<sup>38</sup> Archetypes therefore  
313 have to be defined to capture heterogeneity in indoor environments for providing reliable  
314 indoor intake fraction estimates. In line with our outdoor archetypes, we distinguish indoor  
315 archetypes at three levels of detail: First, a generic *level 1* is defined when emission location  
316 and building characteristics are unknown, reflecting average exposure conditions under  
317 residential indoor settings (see SI, Tables S1, S2). At the intermediate detail *level 2*, intake  
318 fractions are discriminated according to different air exchange rates, occupancies,  
319 recirculation rates, and filter efficiencies for residential indoor settings based on Hodas et  
320 al.,<sup>18</sup> Rosenbaum et al.,<sup>39</sup> and ASHRAE 62.2,<sup>40</sup> and according to different ventilation rates  
321 and occupant densities for occupational indoor settings obtained from ASHRAE 62.1.<sup>41</sup>  
322 Parameterized continental or sub-continental regions are applied at *level 2* for outdoor urban  
323 and rural environments. Finally, if emission scenarios are available for individual building  
324 types, intake fraction estimates can be derived from specific air exchange, occupancy, and  
325 recirculation/filtration characteristics along with defining the building's specific city or rural  
326 area.

327

## 328 Results

### 329 Archetypes for coupled outdoor and indoor environments

330 Using archetypes at three levels of detail allows us to develop spatially-detailed  
331 assessments, while capturing a representative portfolio of buildings, cities and regions. We  
332 first consider an outdoor archetype for ground-level emissions, differentiated into urban and  
333 rural areas characterized by radial population density. The population of the representative  
334 global average city amounts to 2 million inhabitants with a corresponding average linear  
335 population density of 141 capita per m and a population-weighted harmonic mean of the  
336 urban atmospheric dilution rate of 420 m<sup>2</sup>/s. This corresponds to population-weighted close-  
337 to-average meteorological conditions in urban areas and an average relationship between  
338 linear population density and population count. **Figure 2** shows that city-specific linear  
339 population density is indeed linearly correlated to city population, with a continent-specific  
340 intercept reflecting the variation in urban population density that is highest in Asia and lowest  
341 in Australia and North America. This relationship combined with data for parameterized  
342 continental or sub-continental regions based on Kounina et al.<sup>27</sup> is therefore applied at *level 2*,  
343 where the urban archetypes can, for example, be defined to represent small, medium, large,  
344 and mega cities as shown in **Figure 2** or to identify the population size of a representative  
345 average city for each continental and sub-continental region given in **Table 2**. When we use  
346 the representative average urban area for a given region, the intake fraction is directly  
347 obtained from the city population and average dilution rate by the relationship (see **SI, Section**  
348 **S-3**):

$$349 \quad iF_{o,u} = 10^{1.84} \times DR^{-0.876} \times 10^{1.1016 \times \alpha_x} \times POP^{1.1016 \times \beta - 0.1016} \quad (3)$$

350 When the size of a representative region-specific urban area needs to be defined, it can be  
351 obtained by back-calculating the archetypal population in eq. 3 from the weighted population-  
352 average urban intake fraction (see **SI, eq. S12**). This archetypal population varies from  
353 290,000 inhabitants in Northern Australia and 420,000 inhabitants in the Northern regions of

354 Canada and Europe up to 3.4 million inhabitants in Central America, Indonesia, Japan and  
355 South Korea. At *level 3*, actual population characteristics based on data for 3646 cities  
356 ranging from 100,000 to 40 million inhabitants are used for urban areas combined with  
357 population and parameterized characteristics of 8 continental or 16 sub-continental regions for  
358 rural locations.<sup>27</sup> The area,  $A$  [ $\text{m}^2$ ], for cities currently not included in our dataset can be  
359 obtained as  $A = (POP/LPD)^2$  based on known population  $POP$ , and  $LPD$  estimated from  
360 population (see [Figure 2](#) and [SI, Table S3](#)). The atmospheric dilution rate that can either be  
361 calculated from city-specific wind speed and atmospheric mixing height or, if not available,  
362 the default of  $420 \text{ m}^2/\text{s}$  can be applied as the harmonic average of city-specific dynamics  
363 across 3646 cities. For each region/area, such as Indochina (continental Southeast Asia) or  
364 Scandinavia, intake fractions in rural areas are weighted by the contribution of each region to  
365 total continental emissions.

366

367 <[Figure 2](#)>

368

369 For the default indoor environments that we defined as baseline for urban and rural  
370 areas, we use at *level 1* the global default archetype for residential settings. At regionally  
371 differentiated outdoor *level 2*, indoor archetypes are defined according to region-specific air  
372 exchange rates and occupancy (room volume per person) without recirculation or filters. For  
373 studying emissions into specific indoor environments at *level 2*, we define archetypes based  
374 on low, medium and high air exchange rates and occupancy. We assign these archetypes  
375 either no recirculation and no filters or high recirculation rates assuming daily air  
376 conditioning system runtime of 20% (residential settings) and 100% (occupational settings)  
377 coupled with high filter efficiencies based on an average over the range of ASHRAE 52.2  
378 MERV classes 9-12 for “Intended Dust Spot Efficiency” for residential buildings with  
379 advanced air-filtration systems. At *level 3*, specific data for residential and occupational

380 indoor environments or building types can be applied based on data provided by e.g. Hodas et  
381 al.<sup>18</sup>

382 The application of our archetypes to low-stack (~25 m), high-stack (~100 m), and very  
383 high stack (~250 m) as well as to secondary PM<sub>2.5</sub> formed from precursor emissions will be  
384 addressed in a second stage of this research effort. Outdoor and indoor archetype  
385 characteristics and model coefficients for *level 2* are detailed in the SI (Tables S3-S5).

386

### 387 **Effective intake fractions and contributing source environments**

388 **Figure 3** summarizes the variability across effective population-weighted intake  
389 fractions representing aggregated indoor-outdoor exposure for a specific indoor or outdoor  
390 source environment.

391

392 <**Figure 3**>

393

394 Across 3646 urban areas with more than 100,000 inhabitants, the mean effective  
395 population-weighted intake fraction for urban ground-level emissions is 39 ppm (95%  
396 confidence interval: 4.3–160 ppm, median  $\tilde{x} = 26$  ppm). The full range of effective intake  
397 fractions across urban source environments spans from 0.9 to 280 ppm with a squared  
398 geometric standard deviation (GSD<sup>2</sup>) of 4.7, indicating that 95% of all intake fractions fall  
399 within the range from  $\tilde{x}/\text{GSD}^2$  to  $\tilde{x} \times \text{GSD}^2$ . Population-weighted effective intake fractions  
400 across urban areas per region, summarized in **Table 2**, vary from ~10 ppm in Northern regions  
401 and Oceania to 57 ppm in Southeast Asia, with India as high-end sub-continental region at 70  
402 ppm. This distribution corresponds well with the distribution of effective intake fractions in  
403 rural ground-level source environments showing a global mean population-weighted intake  
404 fraction of 2.2 ppm, ranging from 0.02 in Northern regions with tight buildings (low air  
405 exchange) and low occupancy to 4.2 ppm in Southeast Asia with typically high air exchange

406 and high occupancy (95% confidence interval: 0.2–6.3 ppm, median  $\bar{x}$  = 1.7 ppm, GSD<sup>2</sup> =  
 407 6.9) (Table 2). Even for outdoor emission, between 83% and 90% of the intake takes place  
 408 indoors (see upper label in Figure 3) due to the high fraction of the day spent indoors.

409

410 Table 2. Continental and sub-continental summary population-weighted mean effective intake  
 411 fractions including combined indoor and outdoor exposure from urban and rural **outdoor**  
 412 **sources**, number of cities, population count per average city based on population-weighted  
 413 effective intake fraction, and population count in urban areas and totals for each region.

Region	iF [ppm]		<i>n</i> (cities)	<i>n</i> (million capita)	
	urban areas	rural areas		average city	total
Global average	38.6	2.2	3646	2.00	6731.67
<i>Continental regions</i>					
North America	17.0	0.47	271	2.36	334.81
Latin America	33.7	0.51	402	3.34	578.98
Europe	22.0	1.67	701	1.52	751.22
Africa & Middle East	40.0	1.10	466	1.44	1127.13
Central Asia	20.7	0.60	172	1.18	231.78
Southeast Asia	57.0	4.19	1592	2.04	3666.16
Northern regions	9.6	0.02	22	0.44	16.41
Oceania	10.1	0.04	20	0.78	25.17
<i>Sub-continental regions</i>					
Central Asia	20.7	0.59	172	1.13	231.78
Indochina	50.3	1.08	144	2.02	360.29
Northern Australia	3.3	0.01	2	0.29	3.06
Southern Australia & New Zealand	10.8	0.13	18	0.77	22.11
Southern Africa	29.3	0.72	115	1.41	301.45
North, West, East & Central Africa	40.5	1.22	351	1.36	825.68
Argentina+	22.2	0.23	49	2.47	65.65
Brazil+	26.5	0.41	163	2.94	236.69
Central America+ & Caribbean	44.1	0.63	190	3.37	276.64
USA & Southern Canada	17.3	0.44	271	2.25	334.81
Northern Europe & Northern Canada	9.6	0.01	22	0.42	16.41
Europe	22.0	1.65	701	1.45	751.22
East Indies & Pacific	54.6	1.12	61	3.44	237.44
India+	70.0	6.28	420	3.01	1553.18
Eastern China	40.4	3.73	808	1.26	1326.73
Japan & Korean peninsula	40.3	1.52	159	3.44	188.51

414

415 Across indoor source environment archetypes, the mean effective intake fraction is  
 416 0.013 (13,200 ppm) for residential settings and 0.017 (17,200 ppm) for occupational settings,  
 417 when the distribution of residential and occupational spaces in the different regions has not  
 418 been considered. Effective intake fractions across indoor source environments are detailed in  
 419 **Table 3**. For buildings without recirculation/filtration, effective intake fractions range over  
 420 three orders of magnitude from 470 ppm in regions where buildings have high air exchange  
 421 and low occupancy to 62,200 ppm in regions where buildings have low air exchange and high  
 422 occupancy. Indoor exposure contributes 91–99% to effective intake fractions across indoor  
 423 source environments and is highest for conditions with high occupancy, low air exchange, and  
 424 recirculation/filtration of indoor air. Generally, we observe that for an emission into urban  
 425 areas, rural background exposure becomes important for small cities with low urban intake  
 426 fractions especially in India and eastern China with respective rural intake fractions of 6.3 and  
 427 3.7 ppm. In such situations, neglecting the rural background leads to an underestimation of  
 428 the effective exposure from emissions to urban areas by up to 81%.

429

430 **Table 3**. Summary total intake fractions [ppm] including indoor and outdoor exposure for  
 431 residential and occupational **indoor sources** with different air exchange rates, occupancies  
 432 and recirculation/filtration settings.

<i>Residential settings</i>					
	No recirculation/filtration			Recirculation/filtration	
	Air exchange 0.21 h <sup>-1</sup>	Air exchange 0.62 h <sup>-1</sup>	Air exchange 14 h <sup>-1</sup>	Air exchange 0.21 h <sup>-1</sup>	Air exchange 0.62 h <sup>-1</sup>
Occupancy 100 m <sup>3</sup> /person	19500	8890	470	6900	4900
Occupancy 67 m <sup>3</sup> /person	28900	13200	730	10300	7200
Occupancy 30 m <sup>3</sup> /person	62200	29000	1600	22600	16000
<i>Occupational settings</i>					
	No recirculation/filtration			Recirculation/filtration	
	Air exchange 2.7 L/s/capita	Air exchange 8.5 L/s/capita	Air exchange 13 L/s/capita	Air exchange 2.7 L/s/capita	Air exchange 8.5 L/s/capita

	Occupancy 5 capita/100 m <sup>2</sup>	Occupancy 5 capita/100 m <sup>2</sup>	Occupancy 10 capita/100 m <sup>2</sup>	Occupancy 5 capita/100 m <sup>2</sup>	Occupancy 5 capita/100 m <sup>2</sup>
	37300	17200	13000	3250	2950

433

434 Overall, our exposure estimates are in line with results from previous work.<sup>14,17</sup>

435 However, the population-weighted ratio of our effective total intake fractions for outdoor  
 436 urban sources and outdoor urban intake fractions from Apte et al. (2012)<sup>14</sup> is 0.9, and ranges  
 437 from 0.5 in Yakutsk (Russia) to 1.5 La Paz (Bolivia). This means that intake fractions are  
 438 effectively slightly reduced on average when accounting for indoor exposure attributable to  
 439 outdoor sources, especially where removal from outdoor air is driven by air exchange. This is  
 440 mainly the case in regions with generally low air exchange rates and low rural background  
 441 exposure with an average ratio of effective intake fractions combining indoor and outdoor  
 442 exposure to theoretical outdoor intake fractions of only 0.55 across cities in Northern regions,  
 443 while no reduction in intake fractions is seen when air exchange is high. Furthermore,  
 444 effective indoor-outdoor intake fractions exceed outdoor intake fractions in urban source  
 445 environments where air exchange is high and additionally where background exposure is high  
 446 in related rural environments, which is generally the case in Indochina, India, and Africa.

447

## 448 Discussion

### 449 PM<sub>2.5</sub> framework applicability and limitations

450 In summary, our source-to-exposure framework provides for the first time a modular,  
 451 fully mass balanced and flexible approach to combine PM<sub>2.5</sub> exposure indoors and outdoors  
 452 from emissions to occupational or residential indoor, and urban- and rural-outdoor  
 453 environments. This approach provides a sound basis for integrating PM<sub>2.5</sub> exposure  
 454 assessment with multimedia models used to account for other substances potentially  
 455 contributing to human disease burden. The main output of our framework is a set of effective  
 456 indoor-outdoor population intake fractions reflecting three levels of detail based on a set of

457 archetypes for different source environments. This outcome allows us to highlight and  
 458 evaluate differences between indoor/outdoor and outdoor urban/rural emission situations (see  
 459 [Table 4](#)). We thereby bring together a range of well-established underlying models into a  
 460 coupled indoor-outdoor context and build on well-accepted and robust data sets for  
 461 underlying input parameters. Results from applying our framework highlight that indoor  
 462 exposure is an important contributor to PM<sub>2.5</sub> emissions outdoors and that our set of  
 463 archetypes can much better represent the variability between urban and rural outdoor  
 464 exposure than equally or even more data-intensive spatially detailed models and moreover  
 465 allows us to consider indoor environments.

466

467 [Table 4](#). Application and key features of the coupled indoor-outdoor PM<sub>2.5</sub> source-to-exposure  
 468 framework for calculating effective intake fractions for different emission scenarios. The full  
 469 modeling framework is provided in the [SI \(Section S-4\)](#).

Scenario	Application	Key features
Aggregated indoor and outdoor exposure from PM <sub>2.5</sub> urban or rural <b>outdoor sources</b>	<ul style="list-style-type: none"> <li>• Model worksheet: outdoor</li> <li>• User scenario: defines outdoor archetype (urban area representing global default average city, (sub-) continental average city, or real-world city), and emission stack height</li> <li>• Settings: predefined global average residential indoor settings are used</li> </ul>	<ul style="list-style-type: none"> <li>• A tiered approach of three levels of detail is offered with consistent, region-specific population, linear population density, and area</li> <li>• Existing archetypes can be customized and new archetypes (e.g. cities) can be introduced</li> <li>• Intake fractions from different cities and rural areas can be compared and ranked</li> </ul>
Aggregated indoor and outdoor exposure from PM <sub>2.5</sub> occupational or residential <b>indoor sources</b>	<ul style="list-style-type: none"> <li>• Model worksheet: indoor</li> <li>• User scenario: defines residential or occupational indoor archetype in urban or rural outdoor environment; as optional step urban and rural outdoor archetype for background can be defined (in outdoor worksheet)</li> <li>• Settings: predefined global average urban and rural outdoor settings are used by</li> </ul>	<ul style="list-style-type: none"> <li>• Pre-defined sets of low, medium, and high air exchange rate and occupancy with or without recirculation and filter efficiency are offered</li> <li>• Existing archetypes can be customized and new archetypes (e.g. building types) can be introduced by adjusting air exchange rate, occupancy, and recirculation rate and filter efficiency</li> <li>• Intake fractions from different</li> </ul>

	default	indoor environments can be compared and ranked
Aggregated indoor and outdoor exposure from PM <sub>2.5</sub> <b>combined indoor and outdoor sources</b>	<ul style="list-style-type: none"> <li>• Model worksheets: outdoor and indoor</li> <li>• User scenario: defines outdoor archetype (urban area representing global default average city, (sub-)continental average city, or real-world city), and emission stack height in outdoor model, and residential or occupational indoor archetype in urban or rural outdoor environment in indoor model; sum of intake fraction from both models must be used</li> <li>• Settings: intake fractions from indoor sources are used from indoor model and intake fractions from outdoor sources are used from outdoor model</li> </ul>	<ul style="list-style-type: none"> <li>• Intake fractions from different cities, regions, and indoor environments can be summed up and compared and ranked</li> </ul>

470

471 Our framework is described both in mathematical terms (eq. 1 and Table 1) and

472 parameter values (given in SI) and captures the published state-of-the-science in addressing

473 major contributors to PM<sub>2.5</sub> exposure indoors and outdoors. To be parsimonious, we use

474 generic, reported values for e.g. deposition indoors, which can be further refined (see e.g. Lai

475 and Nazaroff,<sup>42</sup> eq. 24) when data become available. To accommodate new archetypal

476 features, the modular framework structure facilitates a flexible definition of additional

477 archetypes and mass-balance terms. This allows capturing exposure variability among a wide

478 range of urban and rural areas and among a large number of diverse indoor environments.

479 Using archetypes facilitates accuracy in capturing exposure heterogeneity based on the strong

480 correlations of emission strength and population density, which requires high spatiotemporal

481 resolutions for assessing exposure, while daily and long-term population mobility reduce the

482 importance of high-resolution spatiotemporal modeling. The construction of the boundaries of

483 urban areas in the underlying data set from Angel et al.<sup>43</sup> may lead to deviations from actual

484 single-city populations and has to be interpreted with care when comparing intake fractions  
485 across cities.

486 A current limitation is that our exposure estimates for urban sources are exclusively  
487 based on intake fractions for ground-level (10 m reference height) emissions from Apte et  
488 al.,<sup>14</sup> whereas global estimates for stack emissions are missing and could be extrapolated  
489 based on Humbert et al.<sup>17</sup> Our model currently provides the capacity for being consistently  
490 coupled with exposure-response information for exposures indoors and outdoors, where  
491 indoor exposure is driven by outdoor sources as accounted for by introducing a penetration  
492 factor (see Table 1). This factor needs to be applied to intake fractions in both indoor and  
493 outdoor source environments when using exposure-response models, such as presented in  
494 Gronlund et al.<sup>44</sup>

495

#### 496 **Linking to exposure-response and health effects**

497 To facilitate the use of our archetypal exposure assessment framework in human  
498 disease burden estimates, our exposure estimates can be linked to available linear or non-  
499 linear exposure-response relationships for PM<sub>2.5</sub>. Our exposure assessment provides the key  
500 input for the non-linear exposure-response model used to translate human PM<sub>2.5</sub> intake into  
501 health impacts and damages. Based on the available evidence, PM<sub>2.5</sub> mass can be used as an  
502 adequate proxy for toxicity impacts.<sup>45</sup> The starting point for an exposure-response model  
503 would therefore be our broad PM<sub>2.5</sub> exposure concentration range in outdoor air and indoor air  
504 that is assumed to be inhaled by humans. Using a model with a broad PM<sub>2.5</sub> concentration  
505 range allows risk estimations also at high exposure levels currently found in various urban  
506 areas as well as indoors, e.g. from solid fuel combustion. Outdoor and indoor air PM<sub>2.5</sub>  
507 exposure concentrations can finally be translated into human intake dose, accounting for  
508 breathing rates under different exposure situations.

509

## 510 **Future research needs**

511 In developing PM<sub>2.5</sub> intake fractions, four pollutant species need to be considered:  
512 emissions of primary PM<sub>2.5</sub>; formation of secondary PM<sub>2.5</sub> from emissions of precursor  
513 substances SO<sub>2</sub>, NO<sub>x</sub>, and NH<sub>3</sub> (as ammonium sulfate and ammonium nitrate); and secondary  
514 organic aerosols (SOA) resulting from emissions of biogenic and anthropogenic precursors.  
515 Furthermore, in some urban environments, it is necessary to address the interaction of volatile  
516 organic compounds (VOCs) and ozone with NO<sub>x</sub> in forming secondary PM<sub>2.5</sub>. While our  
517 framework currently considers primary PM<sub>2.5</sub>, it needs modifications to account for the  
518 contribution of secondary PM<sub>2.5</sub> formed outdoors<sup>46</sup> and indoors<sup>47</sup> to indoor and outdoor  
519 exposures. For outdoor environments, source- and pollutant-specific global data on stack  
520 height are needed in addition to ground-level emission profiles based on e.g. Pregger and  
521 Friedrich<sup>48</sup> for Europe. Our data for urban areas are currently in the domain of cities with  
522 more than 100,000 inhabitants, whereas atmospheric dilution and the population-linear  
523 population density relation require further research for smaller cities where the influence of  
524 rural background exposure might become more relevant. Compared to the high resolution of  
525 urban areas we apply a resolution in rural areas only at the level of sub-continent, as  
526 variability in intake fraction is generally lower in rural areas compared to variability in intake  
527 fractions between cities or between urban and rural environments. This approach is supported  
528 by studies obtaining a relatively small variability also from higher resolution estimates of  
529 intake fractions that are in addition at the low exposure range.<sup>23,49</sup> This low variability is  
530 expected to increase dramatically when the application moves to high-population density  
531 urban areas. For background exposure to emissions in urban areas, our resolution for rural  
532 environments is hence reasonable, whereas the variability in exposure to emissions in very  
533 remote areas might be somewhat underestimated and requires additional study.

534 For indoor environments, factors for near-person resuspension of PM<sub>2.5</sub> deposited  
535 indoors as well as PM<sub>2.5</sub> formed as a result of near-person chemistry need to be developed.<sup>50</sup>

536 This may be especially important in the instance of high indoor person density and low air  
537 movement, where complete mixing cannot be assumed. Such factors could be derived from  
538 calibrating near-person airflow using computational fluid dynamics (CFD) modeling for  
539 different types of indoor sources. Furthermore, to improve the accuracy of indoor exposure  
540 estimates, the fraction of buildings with different air exchange rates and occupancies and the  
541 population fractions in these building archetypes need to be assessed. This is especially  
542 relevant for outdoor source scenarios, since most of the affected population is indoors, while  
543 this aspect is less relevant when assessing indoor sources occurring in individual buildings.

544 Currently, our framework can help to better characterize exposure across multiple  
545 geographic and scenario scales based on available levels of data. It can be used as a tool in air  
546 pollution reduction strategies to evaluate trade-offs among emission sources in different  
547 indoor and outdoor settings of urban and rural environments. It can also aid in evaluating the  
548 environmental performance of products and services in life cycle impact assessment (LCIA)  
549 with respect to life cycle emissions that contribute to PM<sub>2.5</sub> population exposures.

550

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557

## 558 **Supporting Information**

559 The Supporting Information is available free of charge on the [ACS Publications website](#) at  
560 DOI: 10.1021/acs.est.xxx.

561 Input parameters for emission-to-exposure archetypes, correction of outdoor urban to  
562 rural air transfer, and fitting urban population and area from representative exposure  
563 levels are provided in a supporting document (PDF)  
564 The fully operational emission-to-exposure PM<sub>2.5</sub> model including the matrix  
565 calculation framework, archetypes and spatial background data is provided as macro-  
566 enabled Microsoft® Excel® workbook (XLSM)

567

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743 **Figure captions**

744 **Figure 1.** System of distinct archetypes for a set of coupled indoor and outdoor air  
745 compartments used for assessing PM<sub>2.5</sub> emission-to-exposure pathways in urban and rural  
746 environments.

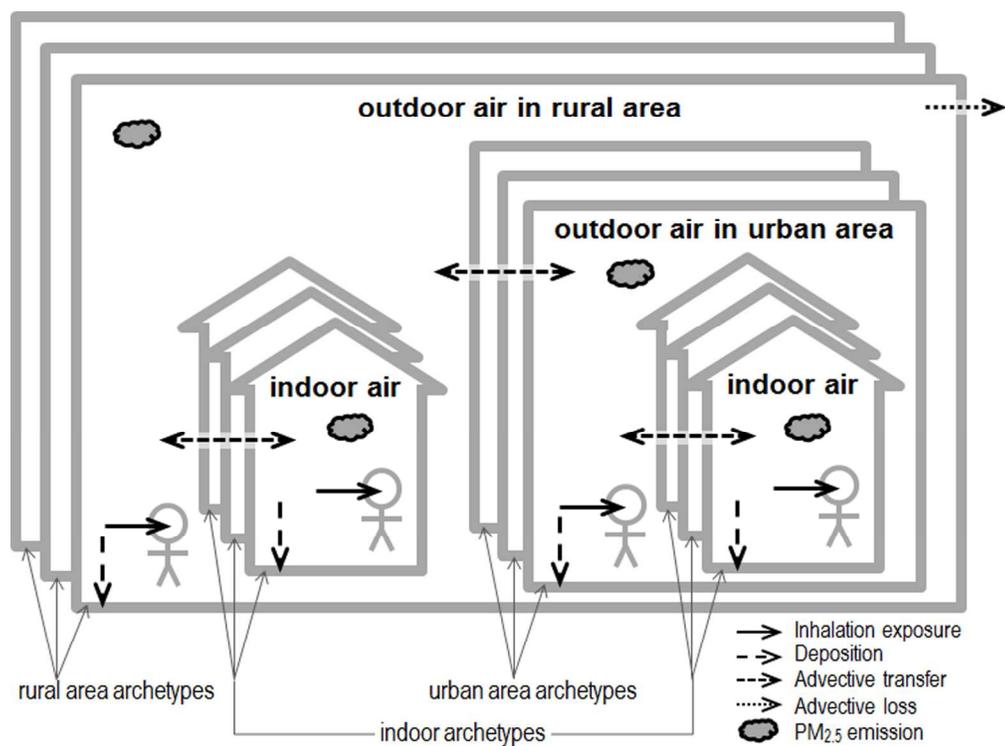
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748 **Figure 2.** Linear population density (*LPD*) and population (*POP*) used for 3646 cities at *level*  
749 *3* grouped according to their corresponding continental region, and location of *level 1*  
750 representative global average urban archetype, and four example *level 2* urban archetypes  
751 obtained from fitting  $\log(LP D) = -1.494 + 0.578 \times \log(P O P)$  with  $R^2 = 0.62$  for the  
752 global average intercept and  $R^2 = 0.77$  for continent-specific intercepts (provided in **SI**,  
753 **Table S3**), and common slope.

754

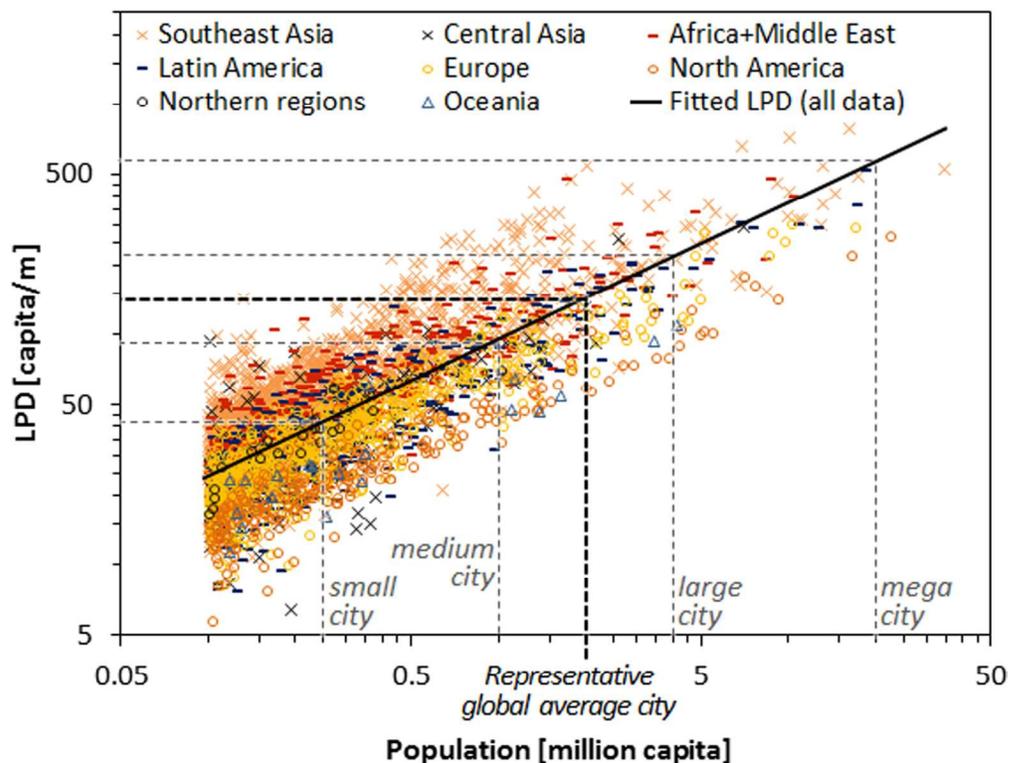
755 **Figure 3.** Population-weighted distribution of effective intake fractions (kg PM<sub>2.5</sub> inhaled per  
756 kg PM<sub>2.5</sub> emitted) and contribution of indoor exposure (percent of total intake fraction) for  
757 residential and occupational indoor emission scenarios and for ground-level urban (range over  
758 all cities per region) and continental rural outdoor emission scenarios.

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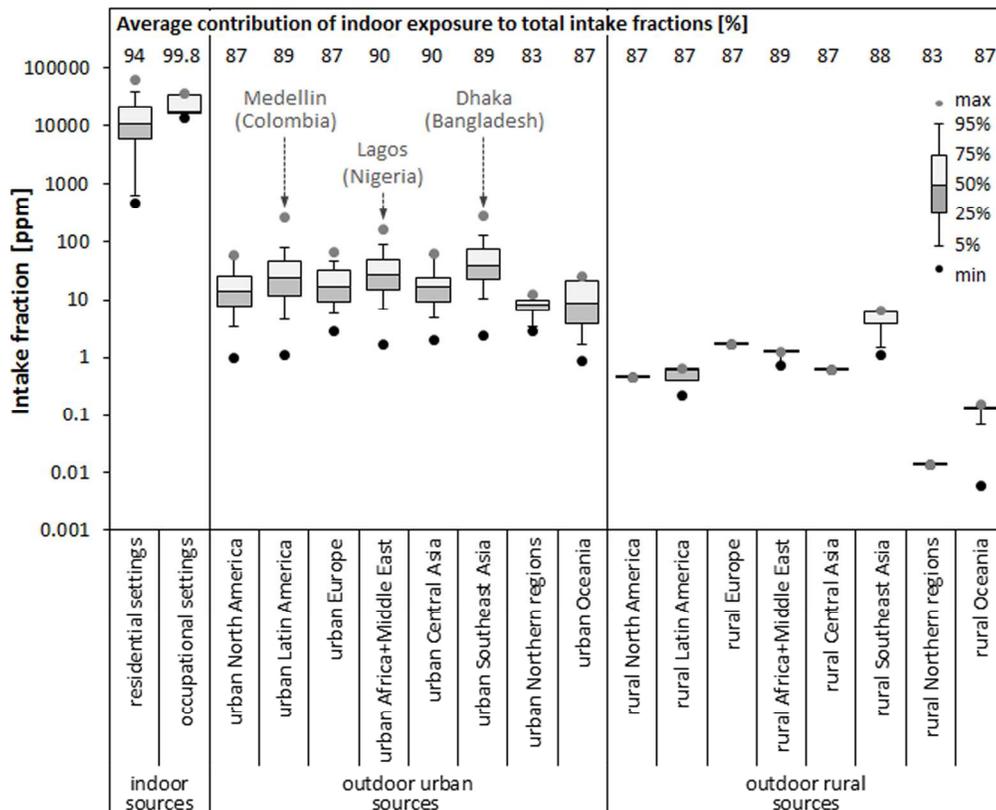
System of distinct archetypes for a set of coupled indoor and outdoor air compartments used for assessing PM<sub>2.5</sub> emission-to-exposure pathways in urban and rural environments.

261x192mm (200 x 200 DPI)



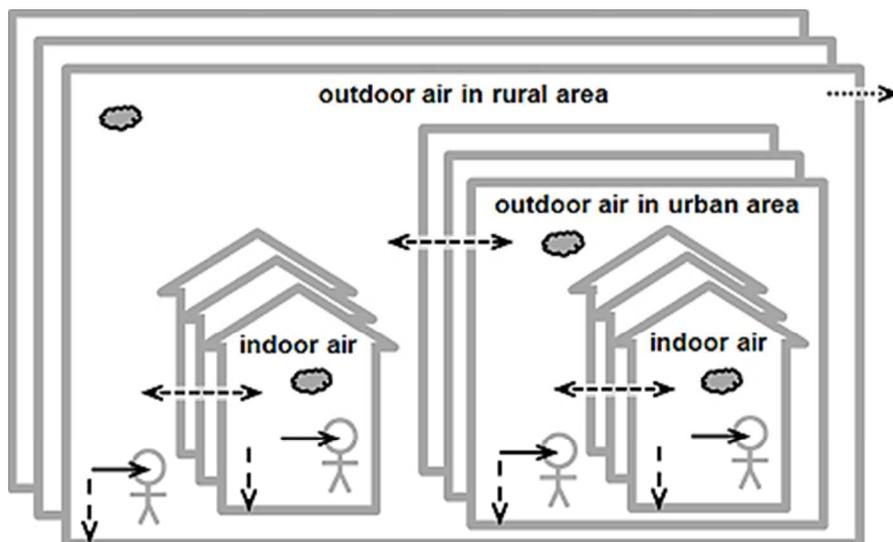
Linear population density ( $LPD$ ) and population ( $POP$ ) used for 3646 cities at *level 3* grouped according to their corresponding continental region, and location of *level 1* representative global average urban archetype, and four example *level 2* urban archetypes obtained from fitting  $\log(LP D) = -1.494 + 0.578 \times \log(POP)$  with  $R^2 = 0.62$  for the global average intercept and  $R^2 = 0.77$  for continent-specific intercepts (provided in SI, Table S3), and common slope.

199x151mm (200 x 200 DPI)



Population-weighted distribution of effective intake fractions (kg PM<sub>2.5</sub> inhaled per kg PM<sub>2.5</sub> emitted) and contribution of indoor exposure (percent of total intake fraction) for residential and occupational indoor emission scenarios and for ground-level urban (range over all cities per region) and continental rural outdoor emission scenarios.

235x190mm (200 x 200 DPI)



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79x47mm (144 x 144 DPI)