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Intake Fraction for Particulate Matter: Recommendations for Life Cycle Impact Assessment

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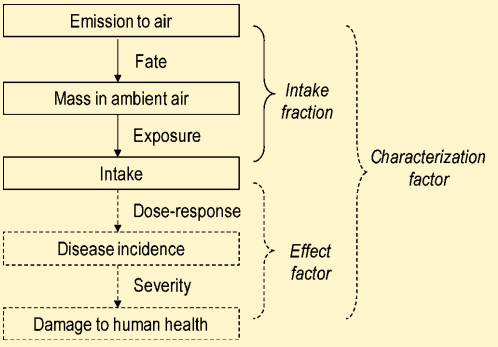
Supporting Information

ABSTRACT: Particulate matter (PM) is a significant contributor to death and disease globally. This paper summarizes the work of an international expert group on the integration of human exposure to PM into life cycle impact assessment (LCIA), within the UNEP/SETAC Life Cycle Initiative. We review literature-derived intake fraction values (the fraction of emissions that are inhaled), based on emission release height and "archetypal" environment (indoor versus outdoor; urban, rural, or remote locations). Recommended intake fraction values are provided for primary $PM_{10-2.5}$ (coarse particles), primary $PM_{2.5}$ (fine particles), and secondary $PM_{2.5}$ from SO_2, NO_x, and NH_3. Intake fraction values vary by orders of magnitude among conditions considered. For outdoor primary $PM_{2.5}$, representative intake fraction values (units: milligrams inhaled per kilogram emitted) for urban, rural, and remote areas, respectively, are 44, 3.8, and 0.1 for ground-level emissions, versus 26, 2.6, and 0.1 for an emission-weighted stack height. For outdoor secondary PM, source location and source characteristics typically have only a minor influence on the magnitude of the intake fraction (exception: intake fraction values can be an order of magnitude lower for remote-location emission than for other locations). Outdoor secondary $PM_{2.5}$ intake fractions averaged over respective locations and stack heights are 0.89 (from SO_2), 0.18 (NO_x), and 1.7 (NH_3). Estimated average intake fractions are greater for primary $PM_{10-2.5}$ than for primary $PM_{2.5}$ (21 versus 15), owing in part to differences in average emission height (lower, and therefore closer to people, for $PM_{10-2.5}$ than $PM_{2.5}$). For indoor emissions, typical intake fraction values are ~1000−7000. This paper aims to provide as complete and consistent an archetype framework as possible, given current understanding of each pollutant. Values presented here facilitate incorporating regional impacts into LCIA for human health damage from PM.



1. INTRODUCTION

This paper aims to review and recommend intake fraction values for primary and secondary particulate matter (PM). Several studies show that PM causes serious adverse health effects, including reduced life expectancy, heart disease, lung cancer, asthma, low birth weight, and premature birth.[1−11] Ambient PM can be primary (directly emitted) or secondary (formed in the atmosphere from precursors). Precursors involved in PM formation include sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3), and volatile and semivolatile organic compounds.

Several life cycle impact assessment (LCIA) methods evaluate the human health damage per mass of PM emitted.[12−15] Hofstetter[12] generated one of the first LCIA approaches evaluating damage factors for PM, based on a consistent integration of data from existing models and epidemiological studies. Since then, researchers have continued to develop fate and exposure

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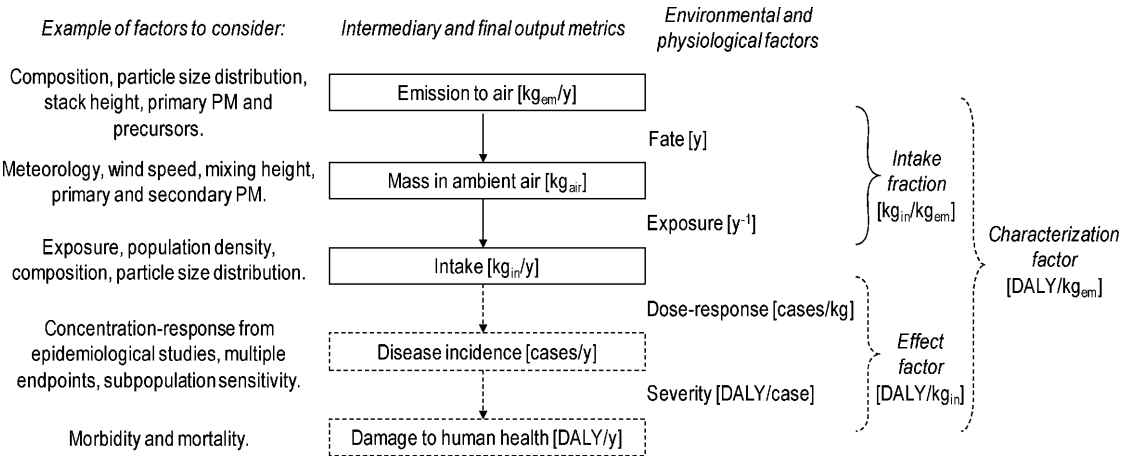

Figure 1. Emission-to-damage framework for particulate matter. Parts represented in dashed lines are outside the scope of this paper. DALY is disability-adjusted life years.

models,[16−20] and conduct epidemiological studies.[4,6,9] Previous reviews suggest that LCIA studies incorporating health effects of PM need to be spatially resolved.[21−24] Potting et al.,[25] reviewing LCIA research on PM, suggest the need for consistency in fate, exposure, and effect evaluation. The present work aims to fill this gap by reviewing intake fraction estimates for PM-related emissions and then recommending a set of intake fraction values that are internally consistent, account for regional differences in fate and exposure ("regionalization"), and facilitate LCIA comparisons with nonparticle pollutants (e.g., organic pollutants).

2. METHODS

2.1. General Framework. Figure 1 illustrates our main approach. Human health impacts associated with a specific source or source category can be estimated via eq 1:[26−28]

$$\text{impact} = \text{emissions} \times \text{intake fraction} \times \text{toxicity} \quad (1)$$

where units are mass or mass per time (emissions), mass inhaled per mass emitted (intake fraction[29]), and health impact (e.g., disease rate, number of adverse outcomes, or risk) per mass inhaled (toxicity). Equation 1 assumes a linear, no-threshold dose−response relationship, an approach that for ambient PM is supported by several studies.[9,30,31] (Where PM concentrations are significantly higher or lower than those observed in epidemiological studies [typically, ∼10−35 $\mu g \cdot m^{-3}$ for chronic exposure to ambient $PM_{2.5}$], linearity might not hold. Recent evidence suggests a log−linear relationship.[32] Grieshop et al.[33] for example, demonstrate the use of intake fraction to investigate exposure concentrations significantly higher than typical outdoor levels.)

2.2. Intake Fraction. Inhalation intake fraction (iF) of a pollutant p is defined as[17,29]

$$iF_p = \frac{\int_{\text{people}} \int_{\text{time}} BR(i,t) C_p(i,t)}{E_{p^*}} \quad (2)$$

where $BR(i, t)$ (m^{-3} person^{-1} d^{-1}) is the breathing rate for person i at time t and C_p (mg m^{-3}) is the incremental exposure concentration attributable to emission E_{p^*} (kg). In the denominator, p^* can refer to emissions of pollutant p (primary pollutant) or of precursory emissions (secondary pollutant), which for PM includes SO_2, NO_x, and NH_3. We employ here a population average breathing rate[34] of 13 $m^3 \cdot person^{-1} \cdot day^{-1}$.

2.3. Factors Influencing the Intake Fraction. *Regionalization.* Recent studies emphasize the importance of "regionalization" (i.e., accounting for local or regional factors) in LCIA[24,30] and fate and exposure of PM.[16,18,36−38] Intake fraction varies by population density[18,19,24,36,39−41] and meteorological conditions,[16,41] especially wind speed and atmospheric mixing height.[17] Temperature and relative concentrations of sulfate−nitrate−ammonium are also important. Averaging method—for example, arithmetic versus harmonic mean—may be important for considering impacts of meteorology.[17]

Height of Emission. Fate and exposure of PM is influenced by the emissions height.[15,16,39,41] Life cycle inventories (e.g., Ecoinvent[42]) often distinguish processes such as power plants, residential wood combustion, and road transportation, which can be attributed to high-stack, low-stack, and ground-level sources, respectively. Levy et al.[16] found that primary $PM_{2.5}$ intake fractions are at least 4 times greater for mobile (ground-level) emissions than for stationary-source (elevated) emissions.

Archetypes Addressing Regionalization and the Height of Emission. Although the exact location of emissions is often unknown in LCIA, some inventories (e.g., Ecoinvent[42]) provide general information (e.g., high versus low population density) and sometimes source types can be derived (e.g., coal power plants generally involve high-stack emissions). Archetypal environments aim to include vertical and horizontal spatial considerations in cases where full details (e.g., exact emission location or population density) are unavailable.[22]

We employ four archetypal environments: indoor sources, and outdoor sources in urban, rural, and remote locations. We further delineate three categories for outdoor emission height: ground-level, low-stack (∼25 m) and high-stack (∼100 m). Here, stack height refers to the physical stack height, not effective height after plume rise. Emissions at high altitude (e.g., from airplanes) are not considered here owing to a lack of relevant studies. We do not distinguish among ground-level sources (e.g., area sources, on-road mobile, off-road mobile); future refinement may be beneficial.

Types of PM. Pollutants considered here are primary PM_{10} (PM smaller than 10 μm), primary $PM_{10-2.5}$ (the share of PM_{10} that is greater than 2.5 μm; "coarse particles"), primary $PM_{2.5}$ (PM smaller than 2.5 μm; "fine particles"), and secondary $PM_{2.5}$

Table 1. Illustrative Parameters and Resulting Intake Fractions for Indoor Emissions[a]

	building volume ($m^3 \cdot person^{-1}$)	k_{ex} (h^{-1})	iF (ppm) for full-time exposure	f_{TE}	iF (ppm) for non-full-time exposure
U.S. household	160	0.5	6770	0.7	4740
office with mechanical ventilation	50	3	3610	0.3	1080

[a] Adapted from Hellweg et al.[47] A mixing factor m (unitless) of 1 is assumed.

Table 2. Model Parameters Used to Generate the Recommended Intake Fractions[a]

	urban emission	rural emission	remote emission
	Exposure Parameters[b]		
population	2 million	1 billion	10 million
area, km^2	240	10 million	10 million
population density, $people \cdot km^{-2}$	8300[c]	100[d]	1
	Meteorological Parameters		
mixing height, m	240	1000	1000
wind speed, $m \cdot s^{-1}$	2.5	2.5	2.5
dilution rate, $m^2 \cdot s^{-1}$	610	2500	2500

[a] Parameters common to all three archetypes include average breathing rate,[34] 13 $m^3 \cdot person^{-1} \cdot d^{-1}$; global average temperature[65] (285 K) and relative humidity, 70%. [b] For all emissions, global exposures are also included. Assumption: year 2015 global population of 7.2 billion people in 75 million km^2.[66] For urban emissions, intake fractions reported in this article also incorporate continental- and global-scale exposures, that is, exposure to PM advected outside the given urban area. [c] Urban parameters are based on weighted averages of United Nations and World Bank data (see Supporting Information). Employing those parameters, linear population density is 130,000 people km^{-1}. [d] Exposure parameters for rural emissions are chosen to match those from the USEtox model.[19]

Table 3. Summary of Recommended Intake Fractions[a]

pollutant and stack height	urban	rural	remote	population-weighted average
		Primary $PM_{10-2.5}$		
high-stack	8.8	0.7	0.04	5.0
low-stack	13	1.1	0.04	7.5
ground-level	40	3.7	0.04	23
emission-weighted average[b]	37	3.4	0.04	21
		Primary $PM_{2.5}$		
high-stack	11	1.6	0.1	6.8
low-stack	15	2.0	0.1	8.9
ground-level	44	3.8	0.1	25
emission-weighted average[b]	26	2.6	0.1	15
		Secondary $PM_{2.5}$		
SO_2	0.99	0.79	0.05	0.89
NO_x	0.20	0.17	0.01	0.18
NH_3	1.7	1.7	0.1	1.7

[a] Here and elsewhere, intake fraction units are parts per million (ppm), indicating mg PM inhaled per kg PM emitted for primary PM, or mg PM inhaled per kg precursor emitted for secondary PM. Precursor species are listed in the table (SO_2, NO_x, NH_3); these species contribute to secondary $PM_{2.5}$ via ammonium nitrate and ammonium sulfate. [b] Although the $PM_{2.5}$ emissions have higher intake fractions than $PM_{10-2.5}$ for each emission height, the emission-weighted average intake fraction can be lower for $PM_{2.5}$ than for $PM_{10-2.5}$ because of differences in typical release height: here, the estimated proportion of ground-level emissions is higher for $PM_{10-2.5}$ (mostly road dust; Table S2, Supporting Information) than for $PM_{2.5}$.

from SO_2, NO_x, and NH_3 (ammonium sulfate and ammonium nitrate). Nearly all secondary ammonium particles are $PM_{2.5}$ (see Supporting Information). Below we do not report separate intake fraction values for primary PM_{10} because they can be estimated as the emission-weighted average of the intake fraction values for $PM_{10-2.5}$ and $PM_{2.5}$. Because of the lack of intake fraction estimates, secondary PM from organic compounds is outside the scope of this paper but is recommended as an area of further research.[43,44] The two main attributes of PM—size and chemical composition—vary among sources and may influence intake fraction and toxicity. Formation rates for secondary PM depend on environmental conditions, including temperature and concentrations of precursor emissions; thus, intake fraction values for secondary PM may differ among precursor species.

Source-Location Framework. The three emission heights (ground-level, low-stack, and high-stack), four emission locations (indoors, outdoor urban, outdoor rural, and outdoor remote), and five independent pollutants (primary $PM_{10-2.5}$ and $PM_{2.5}$ and secondary $PM_{2.5}$ from SO_2, NO_x, and NH_3) yield up to 60 possible combinations. However, many combinations are not pertinent (e.g., stack height is not applicable to indoor emissions and is of limited importance for secondary PM). The actual number of combinations employed here is 27. Calculating primary PM_{10} intake fraction values (based on values for $PM_{10-2.5}$ and $PM_{2.5}$) would add nine additional combinations. Alternatively, providing emission- and population-weighted results (see below, Table 3) increases the number of combinations to 44.

2.4. Archetype Parametrization. This section identifies representative parameters for five archetypal environments: indoor, urban, rural, remote, and unknown environments. This approach is useful when emissions are identified only with a specific archetype (e.g., "urban area"), not a specific location. Continent-specific values are presented in Table S1 (Supporting Information).

Indoor. Intake fraction values generally are orders of magnitude greater for indoor than for outdoor emissions.[26,44−49] A steady-state one-compartment model (eq 3) is commonly used to estimate indoor intake fraction for airborne conserved species; researchers have also investigated episodic emissions,[49] nonconserved species,[49] and emissions to multicompartment indoor environments.[46]

$$iF_{indoor} = f_{TE} N(BR)/V\, m\, k_{ex} \quad (3)$$

Here, f_{TE} (unitless) is the fraction of time people are exposed, that is, for emissions that occur even when people are not present; N is the

number of people exposed; V is the indoor volume (m^3); m is the mixing factor (unitless); and k_{ex} is the air exchange rate (d^{-1}).

Illustrative parameters and resulting intake fractions are provided in Table 1. Those values (~1000−7000 ppm) are consistent with results obtained elsewhere, for example, 6250 ppm for episodic emissions of a conserved species[49] and 1200−3600 ppm for average multicompartment conditions with 1−3 exposed individuals.[46] As with most topics evaluated in this paper, evidence regarding developing country contexts is limited.[33] Hellweg et al.[47] discuss including indoor exposures in LCIA.

Urban. Definitions for "urban area" vary. The U.S. Census Bureau[50] defines a census block as part of an Urban Area if the population density is at least 1000 people·$mile^{-2}$ (390 people·km^{-2}), surrounding blocks have a density of at least 500 people·$mile^{-2}$ (190 people·km^{-2}), and the Urban Area's total population is at least 50 000 people. TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts[13]) employs a threshold population density of 100 people·km^{-2} to distinguish urban versus nonurban. USES-LCA (Uniform System for the Evaluation of Substances−Life Cycle Assessment)[51,52] employs an urban box with average population density of 2000 people·km^{-2}. The population-weighted average urban area in the United States can be represented[17,53] as a 49 × 49 km^2 square with a population density of 753 people·km^{-2}. On average, population density is generally lower in U.S. cities than in cities worldwide.[54]

We propose here to parametrize the default urban box to reflect the population-weighted arithmetic average intake fraction for all urban areas worldwide in a fashion consistent with the one used in USEtox.[19] For intake fraction calculations, "linear population density" (LPD) is often a more useful parameter than population density. Linear population density is the population per width (rather than population per area), that is, the population in a 1-km "strip" extending across the length of a city.[17,54] The default urban box (see Table 2; area 15.5 × 15.5 km^2, population 2 million people) has LPD 130 000 people·km^{-1} and population density 8300 people·km^{-2}. The characteristic mixing height is 240 m and dilution rate (product of mixing height and wind speed) has a harmonic mean of 610 $m^2 \cdot s^{-1}$, based on an analysis[17] of U.S. EPA SCRAM (United States Environmental Protection Agency Support Center for Regulatory Atmospheric Modeling) data[55] for 75 U.S. urban areas. We employ harmonic means rather than arithmetic means because the urban one-compartment intake fraction is inversely proportional to dilution rate.[17] The U.S. value employed here (610 $m^2 \cdot s^{-1}$) is consistent with unpublished estimates[56] for urban areas globally (540 $m^2 \cdot s^{-1}$). See Supporting Information for further discussion.

Rural. We represent rural areas with an average mixing height of 1000 m and a wind speed of 2.5 $m \cdot s^{-1}$.[19] A higher mixing height in rural areas than in urban areas reflects in part the greater average separation between emissions and exposed populations, and the larger residence time in the rural compartment, yielding greater time for vertical mixing to occur in rural areas than in urban areas. Relative to rural and remote exposures, urban exposures are more sensitive to low (e.g., nighttime) mixing heights. When PM emissions occur in rural areas, the population within a few hundred km is exposed,[16,18] which often includes both rural and nearby urban areas, represented here by the global average population density of inhabited regions,[19] 100 people·km^{-2}.

Remote. Ambient emissions in remote areas generally have low intake fractions, because by definition they occur far from population centers. Remote areas are evaluated here as having a population density of 1 person·km^{-2}, which represents the approximate population density over a few million km^2 in remote areas.

Emissions- and Population-Weighted Arithmetic Average. Most life cycle inventories do not specify where emissions occur or else they do so in broad terms (e.g., the country) without specifying whether the emission took place in an urban, rural, or remote area within that country. In such cases, a generic intake fraction for an unknown emission location is useful. If an emission-weighted intake fraction is available for a specific context, then that value should be used. For cases when that value is unavailable, we present here the population-weighted intake fraction, mindful that population-weighted intake fraction may or may not be an appropriate proxy for emission-weighted intake fraction. A justification for this approach is that emissions are typically more correlated with population than with land area. (For example, county-level data from Greco et al.[18] are consistent with our hypothesis; R^2 values for $PM_{2.5}$, NO_x, and SO_2 are, respectively, 0.39, 0.11, and 0.86 for the population−emissions correlation versus 0.14, 0.0001, and 0.02 for the land area−emissions correlation.)

Table 2 summarizes the main parameters used in the models to generate recommended intake fraction values.

2.5. Comparison of Available Models and Data. Several publications provide intake fractions for one or more of the emission archetypes (for example, refs 12, 15−19, 39−41, 57, and 58). When possible, values compared (Figure 2; Table S4, Supporting Information) were adjusted (harmonized) to account for parameter differences (see Table 2; for example, breathing rates[34] were adjusted to 13 $m^3 \cdot person^{-1} \cdot day^{-1}$).

Primary $PM_{2.5}$. Figure 2 indicates, for primary PM intake fraction, an order of magnitude difference between urban and rural areas and an even larger difference between rural and remote areas. Thus, the ability to differentiate between low and high population densities can be at least as important in intake fraction assessment as the choice of model or method. Variations in intake fraction within an archetype are often linked to model limitations that could not be easily harmonized. USEtox[19] and Greco et al.[18] give similar results for the urban archetype when parametrized consistently, with central tendencies of 26 and 20 ppm, respectively (Supporting Information, section 3.2.1). Those values are similar to model- and measurement-based estimates of 14 ± 7 ppm for U.S. urban ground-based emissions.[17] The USEtox rural intake fraction of 2.6 ppm is close to the value reported by Greco et al.[18] of 2.7 ppm. For remote areas, models that can be adapted to low population density conditions give results in the range 0.03−0.1 ppm.

Primary PM_{10}, Primary $PM_{10-2.5}$, and Secondary PM. Figure 3 and Table S5 (Supporting Information) present a summary from multiple models of intake fractions for cases considered here.

Some sources[12,15,40,41,57] suggest no significant intake fraction difference for secondary PM from SO_2 versus NO_x; other sources[16,18,58] suggest that intake fraction is lower for secondary PM from NO_x than from SO_2. For Levy et al.,[16] this difference is derived primarily from their dividing nitrate concentrations by a factor of 4 to reflect their assumption that nitrates form only during winter. Only Hofstetter,[12] Preiss et al.,[40] and Van Zelm et al.[15] provide intake fractions for secondary PM from NH_3; of those three, only the last article[15] is peer-reviewed in a scholarly

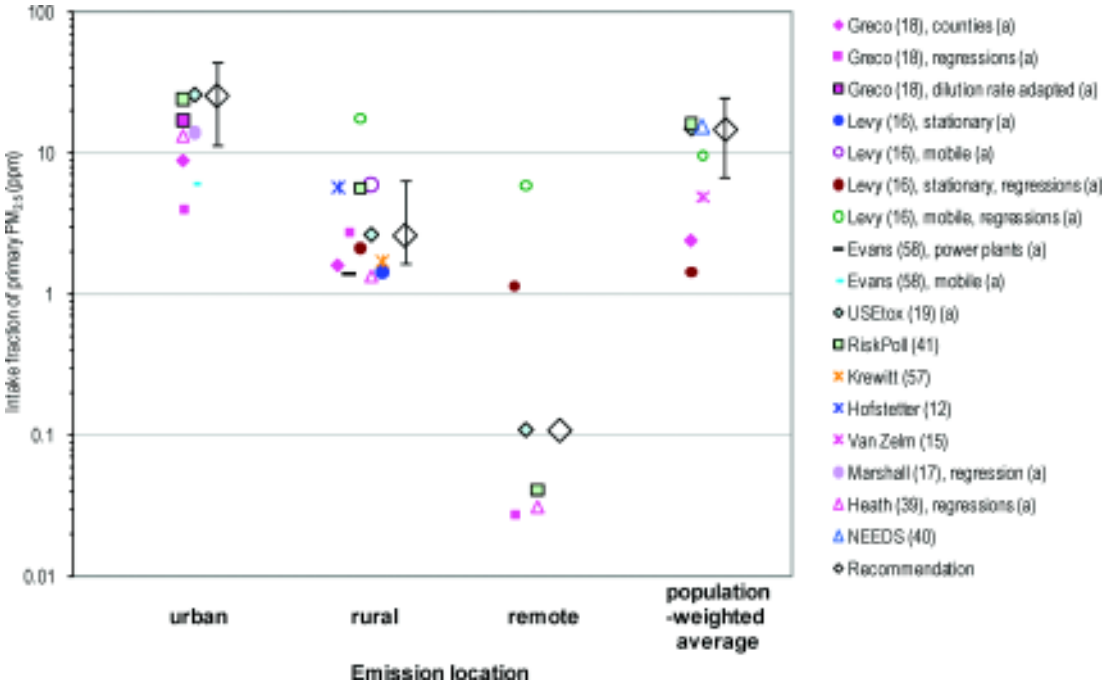

Figure 2. Intake fraction estimates from several models and recommended values for primary $PM_{2.5}$. Values marked "(a)" in legend were modified (harmonized) from the original published values, based on a breathing rate of 13 m^3 $person^{-1}$ d^{-1} and other parameters in Table 2. Recommended values (icon: diamond) are for an emission-weighted average stack height (assumption: 41%, 17%, and 42% of total $PM_{2.5}$ emissions are emitted from high-stack, low-stack, and ground-level sources, respectively; see Supporting Information). Error bars show the range for high-stack versus ground-level. (Low-stack results not shown.) Error bars may extend beyond the displayed literature-derived values, which typically are average stack-height values only.

journal. Several factors might explain differences among results in Figure 3, including different locations studied and different methods employed.

$PM_{10-2.5}$ is generally removed from the environment faster than $PM_{2.5}$. For example, Lai et al.[26] and Liu and Nazaroff[59] report a U-shaped trend, where removal rates are rapid for large and small particles, but intermediate sizes (generally the accumulation mode, ~0.1−1 μm) experience slow removal. Though removal rates are typically faster for $PM_{10-2.5}$ than for $PM_{2.5}$, for each emission archetype, $PM_{10-2.5}$ can experience a higher emission-weighted average intake fraction than $PM_{2.5}$; this counterintuitive result is because a higher proportion of the $PM_{10-2.5}$ emissions are ground-level (mostly road dust; see Table S2, Supporting Information), which have the highest intake fractions of all types of emissions.

When investigating PM_{10}, ideally one would estimate impacts from $PM_{2.5}$ and $PM_{10-2.5}$ separately, since the respective toxicities can differ; eq 4, for primary PM_{10}, is analogous to eq 1:

$$\text{impacts}(PM_{10}) = \text{emissions}(PM_{10})[f_{<2.5,e} iF_{PM2.5} T_{PM2.5} + (1 - f_{<2.5,e}) iF_{PM10-2.5} T_{PM10-2.5}] \quad (4)$$

Here T is toxicity and $f_{<2.5,e}$ is the fraction of emitted PM_{10} that is $PM_{2.5}$. Values for $f_{<2.5,e}$ vary (see Supporting Information section 2); average values in the United States are ~0.12 for transportation emissions (including off-road equipment and road dust), ~0.73 for tailpipe-only emissions from road transportation, ~0.73 for low-stack emissions, and ~0.60 for high-stack emissions.

2.6. Summary of Intake Fraction Values. The intake fraction literature is disparate and at times conflicting; as such, results presented here are suggestive rather than definitive. Our results reflect current understanding based on available scholarship, plus consensus assessment based on the authors' expert judgments. Values aim to reflect "typical" situations; to be as accurate as possible and derived from the most robust methods available; and to be internally consistent (e.g., intake fraction should be smaller for remote than for rural conditions and smaller for rural than for urban conditions).

As discussed next, most results for primary $PM_{2.5}$ were calculated via USEtox, employing the parameter values given above (see also Supporting Information). For primary $PM_{10-2.5}$, we employed the USEtox-derived value for primary $PM_{2.5}$, corrected by use of the RiskPoll-derived[41] intake fraction ratio between $PM_{10-2.5}$ and $PM_{2.5}$ (Tables S4 and S5, Supporting Information). Secondary PM results are based on Greco et al.[18] and Van Zelm et al.[15]

Urban. The urban intake fraction incorporates intraurban intakes, as well as continental-scale plus global intakes (in rural, other urban, and remote areas) attributable to urban emissions. USEtox[19] uses a dilution rate based on a large set of urban data (see Supporting Information) and has the advantage of ensuring a consistent treatment of PM and organic chemicals, which can be assessed by the same model parametrization. The intake fraction for an urban emission of primary PM from an unknown stack height is 26 ppm, calculated by USEtox with the global average urban parameters in Table 2.

For secondary PM from SO_2 and NO_x, the regression model by Greco et al.[18] is employed here. Greco et al.[18] provide an appropriate approach for estimating the intake fractions of these longer-range (several hundred km) pollutants, for which local

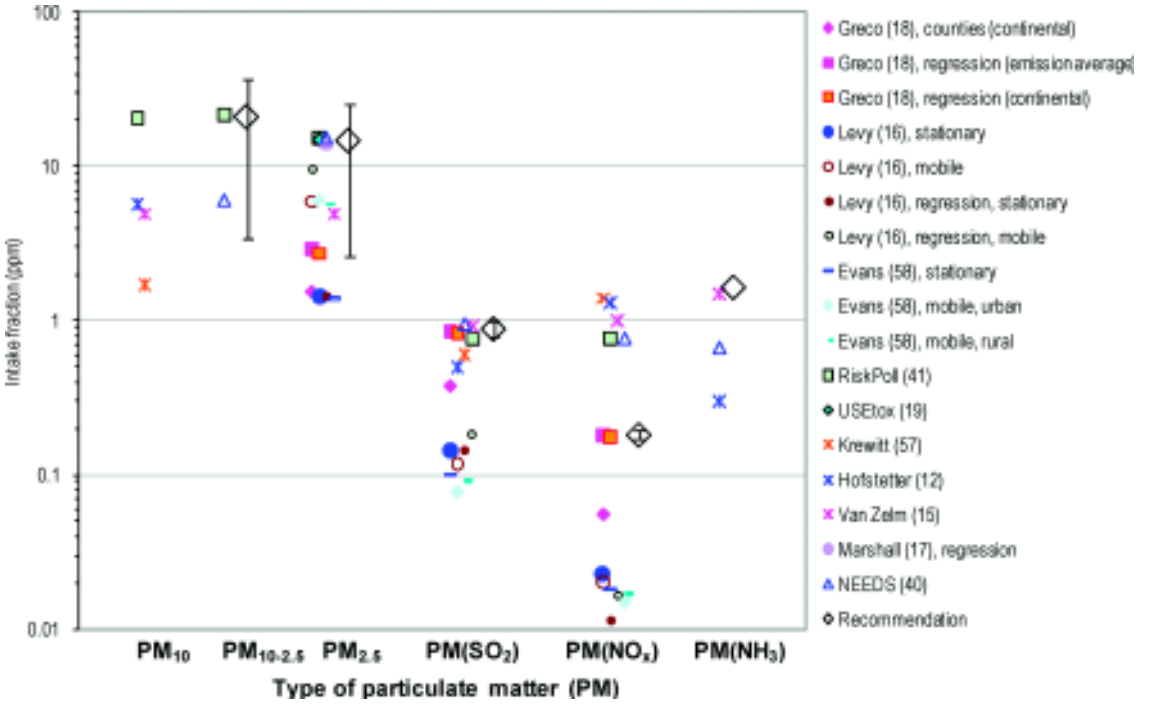

Figure 3. Intake fraction estimates from several model and recommended values for several types of PM. Values are adapted to breathing rates of 13 $m^3 \cdot person^{-1} \cdot day^{-1}$. Recommended values shown here are for a stack-height- and location-weighted average intake fraction. Error bars reflect variations in emission location (rural to urban). PM_{10}, $PM_{10-2.5}$, and $PM_{2.5}$ are primary PM. $PM(SO_2)$, $PM(NO_x)$, and $PM(NH_3)$ are secondary ammonium particles.

population density and dilution rate have modest impacts. We employed results from Van Zelm et al.[15] for secondary PM from NH_3.

Rural. The intake fraction for rural emissions is based on the continental box in USEtox for primary $PM_{2.5}$, on the regressions of Greco et al.[18] for secondary PM from SO_2 and NO_x, and on Van Zelm et al.[15] for secondary PM from NH_3.

Remote. For primary PM, we employed a continental box in USEtox, parametrized as remote and embedded in the world box; the situation modeled is therefore mechanistically consistent with the urban and rural archetypes. For secondary PM, the intake fraction for remote emissions is based on the value obtained for rural areas, corrected by use of the ratio between intake fractions from rural and remote $PM_{2.5}$ emissions for high-stack emissions.

Emission Release Height. For urban and rural primary $PM_{2.5}$, we employ USEtox results, corresponding to an emission from an unknown stack height. Height-specific intake fractions are determined by ratios of modeled intake fractions from different stack heights by use of RiskPoll.[41] Intake fraction differences among release heights (ground-level, low-stack, high-stack) are greater for urban emissions than for rural emissions, because population density is higher in urban than in rural areas. The intake fraction ratio of ground-level to low-stack emissions is 2.9 for urban and 1.9 for rural conditions, and the intake fraction ratio of low-stack to high-stack emissions is 1.3 for urban and 1.2 for rural conditions (see Supporting Information).

Levy et al.[16] found that the secondary PM intake fraction does not differ significantly by source category. Therefore, the intake fractions of secondary PM from high- and low-stack emissions are assumed to be the same as the intake fraction of ground-level emissions. Similarly, for remote emissions we do not differentiate among stack heights, since the air will generally be well-mixed before it reaches most of the exposed population.

Weighted Arithmetic Average. The average continental or rural intake fraction values should not be applied to emissions in unknown locations, because emissions may be correlated to population and therefore situated, on average, closer to urban areas than a continental intake fraction value would suggest. For this reason, the recommended weighted average intake fraction of an emission in an unknown location is calculated as a function of the urban, rural, and remote values:

$$iF_{p,\text{average location}} = \frac{1}{M_p}\sum_l (m_{p,l} iF_{p,l}) \quad (5)$$

where $iF_{p,\text{average location}}$ is the emission-weighted average intake fraction of pollutant p; $m_{p,l}$ and $iF_{p,l}$ are the mass and intake fraction, respectively, of pollutant p emitted in a location l; and M_p is the total mass emitted. A similar equation could be generated employing alternative weighting values. For outdoor emissions, eq 5 can be simplified as

$$iF_{p,\text{average location}} = f_{p,\text{e,urban}} iF_{p,\text{urban}} + f_{p,\text{e,rural}} iF_{p,\text{rural}} + f_{p,\text{e,remote}} iF_{p,\text{remote}} \quad (6)$$

where $f_{p,\text{e,urban}}$, $f_{p,\text{e,rural}}$, and $f_{p,\text{e,remote}}$ represent the respective fractions of PM emissions occurring in urban, rural, and remote regions. As noted above, if mass-weighted values are unavailable, population-weighted values may provide a useful, albeit imperfect, approximation for average intake fraction. For illustration, we set $f_{p,\text{e,urban}}$, $f_{p,\text{e,rural}}$, and $f_{p,\text{e,remote}}$, respectively, to 0.46, 0.53, and 0.01, representing global average population-based values. Secondary pollutants have similar urban and rural intake

fractions (10−20% variation), so the weighting between these two archetypes does not significantly affect the resulting averages.

The emission-weighted average intake fraction developed for an unknown stack height is evaluated by use of eq 7:

$$iF_{p,\text{average stack}} = f_{p,\text{e,high-stack}} iF_{p,\text{high-stack}} + f_{p,\text{e,low-stack}} iF_{p,\text{low-stack}} + f_{p,\text{e,ground-level}} iF_{p,\text{ground-level}} \quad (7)$$

where $f_{p,\text{e,high-stack}}$, $f_{p,\text{e,low-stack}}$, and $f_{p,\text{e,ground-level}}$ are respectively the mass fractions of pollutant p emitted by high-stack, low-stack, and ground-level sources in the same geographical limits. Here, we estimate these fractions using data from U.S. EPA[63] (Table S2, Supporting Information).

3. RESULTS AND DISCUSSION

Table 3 summarizes our recommended intake fraction values for the cases considered here. Assumptions behind those values are given in Tables S6 and S7 (Supporting Information).

3.1. Uncertainty and the Importance of Spatial Differentiation. For intake of PM, there are many sources of variability and uncertainty along the emission-to-intake chain. The intake fraction source-location matrix (Table 3) helps address major sources of variability (urban, rural, and remote location; emission height; and primary versus secondary PM). When the available information allows, accounting for archetypes will reduce the uncertainty of the final LCIA results. Obviously, accounting for local characteristics (e.g., meteorology) would provide further improvement.

We estimate part of the uncertainty of the source-location intake fraction matrix by examining the variability among existing models. Setting the same population density and breathing rate in all emissions-to-intake models, we estimate the squared geometric standard deviation (GSD^2) of the set of modeled intake fractions. (By definition, 95% of the values fall between the median divided by the GSD^2 and the median multiplied by the GSD^2.) The GSD^2 of the primary $PM_{2.5}$ intake fractions for urban, rural, and emission-weighted intake fractions are 3.5, 4.6, and 5.3, respectively. Those values reflect variability among modeled values used here, rather than model uncertainty or variability among all potential situations (e.g., ambient concentrations of precursors influence intake fractions for secondary PM).

These uncertainty estimates can be compared with the variation in intake fraction caused by population density patterns, which can be more than 2 orders of magnitude between emissions in high population density areas (e.g., truck emissions in a city) and low population density areas (e.g., emissions from a diesel train crossing remote areas). The variability between country-average values[40] in Europe is up to 1 order of magnitude. Thus, regionalizing intake fractions and therefore associated characterization factors by considering variability in population density patterns is an important step toward the reduction of overall variability and uncertainty in evaluating human health damage when LCIA is used.

3.2. Future Research Needs. Intake fraction values given above provide a consistent framework for LCIA practitioners to evaluate the fate and exposure of primary and secondary PM, including information on source type and location. Further research should focus on at least two areas: fate and exposure of PM, and health effects. For example, further investigation is needed regarding optimal averaging method of wind speeds and mixing heights, especially for nonurban areas. Low-dilution conditions (calm winds, low mixing height) may be especially important for intake fraction estimation yet are challenging to model. Prior research, for example, on buses and residential wood combustion[60−63] has explored how differences in intake fraction among sources could be exploited to make air quality management more effective; more work along those lines, and in understanding how to reduce life cycle impacts, is warranted. Future work could usefully explore archetypal environments (e.g., oceans, high altitudes) or pollutants (e.g., secondary PM from volatile organic compounds) not evaluated in this paper. Important details regarding chemical composition and (time-evolving) size distributions were ignored here. A major gap in the literature is the small number of investigations on intake fraction in developing country contexts.

While the recommended intake fractions would benefit from further work, the source-location matrix suggested in the present paper provides a framework for researchers to improve their evaluations of adverse health effects caused by primary and secondary PM. In many damage-oriented LCIA studies, PM is estimated to be responsible for a large fraction of the total human health damage. Harmonizing the values used in LCIA studies and making those values consistent with the characterization of organic species[19,64] will increase accuracy, consistency, and comparability among results for human health damage.

■ ASSOCIATED CONTENT

Supporting Information. Seven tables, 20 equations, and text describing calculation methodological details. This material is available free of charge via the Internet at http://pubs.acs.org.

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Supporting Information for

Intake fraction for particulate matter: recommendations for life cycle impact assessment,

S Humbert, JD Marshall, S Shaked, JV Spadaro, Y Nishioka, P Preiss, TE McKone, A Horvath, O Jolliet.

In the Supporting Information, we first describe the archetype parameterization (Section 1) and characterize the emission sources (Section 2). We then detail the intake fraction calculation (Section 3), providing information on the regression models, the values used for the charts comparing the intake fractions, and the formulas used to calculate the recommended values. Last, we provide a discussion on variability and uncertainty (Section 4).

1 Archetype parameterization

1.1 Breathing rate

Modeled average breathing rates vary, for example from 9.5 (*S1*) to 25 m^3 $person^{-1}$ day^{-1} (*S2*); we selected a population average breathing rate of 13 m^3 $person^{-1}$ day^{-1} (*S3*). That value is applied to all intake fractions calculated in this paper, unless specified otherwise.

1.2 Indoor environment parameterization

No strong evidence was found to justify amplification for indoor pollutant exposure above room average conditions. Therefore, in Equation 3 in the main text, the mixing factor m is assumed to be 1. Additional information can be found in Hellweg et al. (*S4*) and Meijer et al. (*S5*).

1.3 'World city' parameterization

The USEtox model calculates the recommended intake fraction of urban emissions, but this must be parameterized to best approximate an 'average' world city. Below, we derive the effective values that result in a globally averaged urban intake fraction.

The intra-urban intake fraction (iF) of a pollutant emitted in an urban area i is approximated in a box model as:

$$iF_{i,\text{intra-urban}} = \frac{a \times BR \times N_i}{u_i \times H_i \times W_i} = \frac{a \times BR}{u_i \times H_i} \times d_i \times L_i, \text{ with } d_i = \frac{N_i}{L_i \times W_i} \quad \text{(S1)}$$

where N (persons) is the number of persons in the urban area; BR (m^3 $person^{-1}$ day^{-1}) is the average breathing rate; a (unitless) is the correction factor to account for the fact that (i) a pollutant can be emitted anywhere in the urban area i and not only along the up-wind periphery, and (ii) the air that left the urban area i can return with some of the pollutant (i.e., a back-and-forth movement of air) (the factor a varies, with a typical value of ~0.75) [*S6*]; u (m day^{-1}) and H (m) are the dominant wind speed and mixing height of the urban area; L (m) is the length of the urban area (measured in the direction of the dominant wind); W (m) is the width of the urban area (often, the urban area is assumed to be a square, thus $L = W$); and d (persons m^{-2}) is the population density of the urban area. Equation S1 assumes that deposition and degradation rates within the urban area are negligible relative to advection out of the area.

Equation S1 shows that the intake fraction is proportional to the term $d \cdot L$ (persons m^{-1}), defined as the linear population density (*S7, S8*). The term $u \cdot H$ (m^2 day^{-1}) is the dilution rate. The harmonic mean dilution rate is 610 m^2 s^{-1}, based on a prior analysis (*S7*) of meteorological conditions in 75 urban areas represented in the USEPA SCRAM database (*S9*).

The emission-weighted average intra-urban intake fraction $iF_{\text{average intra-urban}}$ of a pollutant over all urban areas is computed as:

$$iF_{\text{average intra-urban}} = \frac{\sum_i m_i \times iF_i}{\sum_i m_i} = \frac{a \times BR}{u \times H} \cdot \frac{\sum_i m_i \times d_i \times L_i}{\sum_i m_i} = \frac{a \times BR}{u \times H} \left(d \times L\right)_{\text{effective}} \quad \text{(S2)}$$

where m_i is the mass emitted in urban area i and $\left(d \times L\right)_{\text{effective}}$ is the effective linear population density representative of the average of all the urban areas:

$$\left(d \times L\right)_{\text{effective}} = \frac{1}{\sum_i m_i} \times \sum_i m_i \times d_i \times L_i \quad \text{(S3)}$$

If we assume emissions are proportional to population, then:

$$\left(d \times L\right)_{\text{effective}} = \frac{1}{\sum_i N_i} \times \sum_i N_i \times d_i \times L_i \quad \text{(S4)}$$

The global urban population is $\sum_i N_i$ = 3.82·10^9 persons (year 2015; *S10*). Using data from the United Nations Statistics Division (*S11*) and the World Bank (*S12*), we find that $\sum_i N_i \times d_i \times L_i \approx 5 \cdot 10^{14}$ persons^2 km^{-1}, giving an effective linear population density of $(d \times L)_{\text{effective}} \approx$ 130,000 persons km^{-1} = 130 persons m^{-1}. Accounting for the different sources of uncertainty, the uncertainty in the global population-weighted average linear population density value is estimated to be approximately 25%.

The linear population density of the 'default' world city is therefore set as 130,000 persons km^{-1}, which can be represented by a population density of 8,300 persons km^{-2} over an area of 15.5 km x 15.5 km. These parameters represent a total population of 2,000,000 persons over 240 km^2. With these values, the simple model presented in Equation S2 produces an average intra-urban intake fraction iF_{average} of 24 ppm (Equation S5):

$$iF_{\text{average}} = \frac{0.75 \times (13\ \text{m}^3/\text{person - day})}{(610\ \text{m}^2/\text{s}) \times (3600\ \text{s/hr}) \times (24\ \text{hr/day})} \times (130\ \text{person/m}) = 24\ \text{ppm} \quad \text{(S5)}$$

1.3 Parameterizations by continent

Based on the World Bank (*S12*) data, we find that the population density d (persons km^{-2}) of an urban area i is correlated to its population N, with an approximately linear relationship between the logs of the two variables:

$$d_i = (N_i)^{a_i} \times b_i \quad \text{(S6)}$$

where the parameters a_i and b_i can be specific to each continent. These parameters are sensitive to the population and area of the cities used in the regression, therefore the continent-specific results provided by Equation S6 and presented in Table S1 are a first approximation only and should be used with caution. Table S1 summarizes the parameters by continent and for the World.

TABLE S1. Summary of the parameters by continent and for the world (2015)

Parameter	World	Generic continent	US+	Latin America	Europe	Africa + Middle East	Central Asia	South East Asia	Arctic	Oceania	Antarctica
Area, actual (km^2)	105 M	-	14 M	20 M	8.4 M	33 M	17 M	16 M	17 M	8.0 M	12 M
Population (persons)	7.2 B	1.0 B	360 M	630 M	760 M	1.3 B	230 M	3.9 B	17 M	27 M	1'000
Population density (persons/km^2)	49	-	26	31	90	38	14	240	1.0	3.4	0
'Continental' area (km^2)[a]		10 M	11 M	25 M	9.7 M	35 M	4.6 M	23 M	17 M	27 M	0
'Continental' population (persons) [a]		1.0 B	360 M	630 M	760 M	1.3 B	230 M	3.9 B	17 M	27 M	0
'Continental' population density (persons/km^2) [a,b] (based on GIS analysis)	100	100	33	25	78	37	50	170	13	7.6	0
Urban population (persons)	3.7 B	520 M	200 M	260 M	350 M	370 M	67 M	2.5 B	4.7 M	15 M	0
Urban linear population density (persons/km) (see Equation S4)[c]	130,000	130,000	91,000	190,000	110,000	130,000	120,000	130,000	70,000	75,000	0
Urban population density (persons/km^2)	8,300	8,300	2,200	8,800	6,100	11,000	7,000	16,000	4,100	2,300	-
Urban length (km)[d]	15.5	15.5	38	18	15	12	15	8.5	12	29	-
a_i (see Equation S6)	0.31	0.31	0.16	0.32	0.32	0.37	0.53	0.21	0.29	0.12	-
b_i (see Equation S6)	2.0	2.0	2.3	1.8	1.7	1.6	0.54	2.7	1.9	2.6	-
Rural population (persons)	3.5 B	480 M	160 M	360 M	410 M	910 M	160 M	1.4 B	13 M	12 M	-
$PM_{2.5}$: Urban iF[e]		26	15	29	18	25	20	29	9.3	12	-
$PM_{2.5}$: Rural iF[e]		2.6	0.92	0.75	2.1	1.1	1.3	4.6	0.42	0.30	-
$PM_{2.5}$: Remote iF[e]		0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
$PM_{2.5}$: Population-weighted average iF[f]		15	8.5	12	10	7.9	6.5	20	1.8	4.9	0.1

[a] Continental refers to the archetype defined to model exposure to rural emissions (see main text, Section 2.4 and Table 2);
[b] Effective population density approximated based on GIS analysis of locations distributed throughout continent.
[c] Linear population densities (LPD) are calculated based on urban data of recorded cities and supplemented by estimates of populations and sizes of unknown cities; we find that the LPD is sensitive to the recorded urban fraction of each region but less dependent on the assumed population of unrecorded cities.
[d] Assuming a square urban area.
[e] Corresponding emission-weighted average for the type of emission source.
[f] Assuming 10,000,000 people in remote area.
Note: 1) M = million, B = billion, 2) Because of rounding, values might not match perfectly.

2 Characterization of particulate matter (PM) emissions

2.1 Total emission by source type and emission height

Table S2 shows the total United States annual emissions (t y^{-1}) from different sources for different pollutants (*S13*). Each source is marked as *H*, *L*, *G*, or *T* to indicate that the emissions were assumed as, respectively, 'high-stack', 'low-stack', 'ground-level', and 'tailpipe', with the latter being treated the same as 'ground-level'. Wildfire and miscellaneous emissions are excluded from the inventory. Table S2 also

shows the fraction of emissions from high-stack, low-stack, and ground-level sources, based on these data and classifications.

TABLE S2. Total United States emissions of PM_{10}, $PM_{10-2.5}$, $PM_{2.5}$, SO_2, and NO_x, by source (*S13*) [a]

Source Sector	PM_{10} (t/y)	$PM_{10-2.5}$ (t/y)	$PM_{2.5}$ (t/y)	SO_2 (t/y)	NO_x (t/y)
Electricity generation (H)	616,801	116,543	500,258	10,411,906	4,668,962
Fertilizer & Livestock (L)	3,104	1,581	1,523		2,099
Fires (-)	1,452,085	221,565	1,230,520	102,028	161,029
Fossil fuel combustion (L)	361,228	170,352	190,876	2,028,611	2,419,027
Industrial processes (H)	1,203,048	711,524	491,524	1,233,856	1,158,549
Miscellaneous (-)	5,512,327	5,267,082	245,245	754	2,327
Non-road equipment (T)	329,578	27,697	301,881	515,081	4,517,275
On-road vehicles (T)	203,260	54,827	148,433	257,519	8,133,567
Residential wood combustion (L)	366,619	28,572	338,047	5,093	36,716
Road dust [b] (T)	10,253,451	9,398,548	854,903	0	0
Solvent use (L)	8,332	1,306	7,026	1,048	9,010
Waste disposal (H)	296,222	22,232	273,990	26,015	120,552
Total [c]	**13,641,643**	**10,533,182**	**3,108,461**	**14,479,129**	**21,065,757**
Total high-stack (H)	2,116,071	850,299	1,265,772	11,671,777	5,948,063
Total low-stack (L)	739,283	201,811	537,472	2,034,752	2,466,852
Total ground-level (G+T)	10,786,289	9,481,072	1,305,217	772,600	12,650,842
% from high-stack ($f_{e,high\text{-}stack}$)	16%	8%	41%	81%	28%
% from low-stack ($f_{e,low\text{-}stack}$)	5%	2%	17%	14%	12%
% from ground-level ($f_{e,ground\text{-}level}$)	79%	90%	42%	5%	60%

Fraction of PM_{10} smaller than 2.5 μm ($f_{<2.5,e}$) and PM_{10} within the range 2.5 μm and 10 μm ($f_{10\text{-}2.5,e}$)

	$f_{<2.5,e}$	$f_{10\text{-}2.5,e}$	
high-stack	60%	40%	
low-stack	73%	27%	
ground-level	12%	88%	$f_{<2.5,e}$ = 0.73 for tailpipe-only emissions from road transportation
indoor	92%	8%	based on residential wood combustion and solvent emissions

[a] NH_3 annual emissions in the Unites States are approximately 4 million tons, 95% from low-stack (mainly livestock) and 5% from transportation (i.e., ground-level) (*S13*).
[b] If the inventory database used does not contain road dust emissions, then this value should be removed from the calculations, based on values in Table S2 and Table 3.
[c] Excluding wildfire and miscellaneous emissions.

To estimate typical stack height values, we use a detailed list of stack heights from ten German states found in Pregger and Friedrich (*S14*). SO_2, and NO_x emission-weighted average stack heights from industrial point sources are, respectively, 144, 121, and 132 m. Assuming that German industrial point sources have relatively higher stacks, a value of 100 m for worldwide stacks, based on Van Zelm et al. (*S15*), appears plausible.

2.2 Fraction of PM_{10} smaller than 2.5 μm ($f_{<2.5,e}$)

Different values are suggested in the literature for the fraction of PM_{10} smaller than 2.5 μm ($f_{<2.5,e}$), from as low as 5% (embarkation of coal; *S16-18*), 8% for road dust (*S13*), 60% (*S16, S19*), 66% (average in Germany; *S20*), 89% (United States coal power plant average; *S21*), 95% (tailpipe; *S22*), 70% to 95% (coal fired power station with flue gas cleaning) and up to nearly 100% (mobile, internal combustion engine; *S16-S18*). As shown in Table S2, the emission-weighted average U.S. values for $f_{<2.5,e}$ vary with emission

height. These fractions are variable and can be adapted to specific situations, such as for older power plants.

3 Intake fraction

3.1 Intake fraction regressions

3.1.1 Intake fraction regressions from USEtox

Unknown stack height. For a given population density, the intake fraction of primary $PM_{2.5}$ modeled with USEtox (version 1.00, December 1, 2009; *S23*) can be approximated using the regressions below (Equations S7 - S10), where L is the length of the urban area (km) and d_{urban}, d_{rural} and d_{remote} are the respective population densities of the urban, rural, and remote areas (persons km^{-2}). These were calculated based on a series of 40 runs each, with densities ranging from $1/1000^{th}$ of recommended value to 100 times the value.

The total intake fraction for a remote emission (with an R^2 of 1.00) is:

$$iF_{remote} = 2.3 \cdot 10^{-8} \times d_{remote} + 8.6 \cdot 10^{-8} \quad \text{(S7)}$$

The total intake fraction for a rural emission (with an R^2 of 1.00) is:

$$iF_{rural} = 2.6 \cdot 10^{-8} \times d_{rural} + 7.9 \cdot 10^{-8} \quad \text{(S8)}$$

The intra-urban intake fraction for an urban emission (with an R^2 of 1.00) is:

$$iF_{intra\text{-}urban} = 1.8 \cdot 10^{-10} \times L \times d_{urban} \quad \text{(S9)}$$

The total intake fraction for an urban emission is the sum of Equation S9 and Equation S8:

$$iF_{urban} = iF_{intra\text{-}urban} + iF_{rural} \quad \text{(S10)}$$

If rural conditions are unknown, the intake fraction for a rural emission presented in Table 3 can be used as a default iF_{rural} in Equation S10. Note that the high R^2 in all of the equations is related to the dominating influence of population density relative to the effects of advection, deposition and degradation. A shorter-lived pollutant would not have as good a linear fit.

Differentiating stack heights. Intake fractions of primary $PM_{2.5}$ evaluated with USEtox (*S23*) are for unknown stack height emissions. The intake fraction for an unknown stack height ($iF_{unknown\text{-}stack}$) can be calculated as the emission-weighted average of the intake fractions for high-stack ($iF_{high\text{-}stack}$), low-stack ($iF_{low\text{-}stack}$) and ground-level ($iF_{ground\text{-}level}$):

$$iF_{\text{unknown-stack}} = f_{e,\text{high-stack}} \times iF_{\text{high-stack}} + f_{e,\text{low-stack}} \times iF_{\text{low-stack}} + f_{e,\text{ground-level}} \times iF_{\text{ground-level}} \quad \text{(S11)}$$

where $f_{e,\text{high-stack}}$, $f_{e,\text{low-stack}}$, and $f_{e,\text{ground-level}}$ are the respective fractions of total emissions from high-stack, low-stack and ground-level emissions (Table S2).

To consistently differentiate between stack heights, one can calculate the intake fraction ratios of ground-level to low-stack (X) and low-stack to high-stack (Y) emissions:

$$X = iF_{\text{ground-level}}/iF_{\text{low-stack}} \quad \text{(S12)}$$

$$Y = iF_{\text{low-stack}}/iF_{\text{high-stack}} \quad \text{(S13)}$$

RiskPoll (*S24*) provides these intake fractions for primary $PM_{2.5}$, with ground-level to low-stack ratios (X) of 2.9 for urban and 1.9 for rural conditions, and ratios of low-stack to high-stack (Y) of 1.3 for urban and 1.2 for rural conditions.

Combining Equations S11, S12 and S13:

$$iF_{\text{high-stack}} = iF_{\text{unknown-stack}}/(f_{e,\text{high-stack}} + Y \times f_{e,\text{low-stack}} + X \times Y \times f_{e,\text{ground-level}}) \quad \text{(S14)}$$

$$iF_{\text{low-stack}} = Y \times iF_{\text{unknown-stack}}/(f_{e,\text{high-stack}} + Y \times f_{e,\text{low-stack}} + X \times Y \times f_{e,\text{ground-level}}) \quad \text{(S15)}$$

$$iF_{\text{ground-level}} = X \times Y \times iF_{\text{unknown-stack}}/(f_{e,\text{high-stack}} + Y \times f_{e,\text{low-stack}} + X \times Y \times f_{e,\text{ground-level}}) \quad \text{(S16)}$$

To estimate the intake fractions of very high-stack emissions (>250 m), values from RiskPoll (*S35*) show that the high-stack (100 m) intake fraction can be multiplied by 0.40 and 0.77 for urban and rural emissions for $PM_{10\text{-}2.5}$, and by 0.54 and 0.79 for urban and rural emissions for $PM_{2.5}$, respectively.

3.1.2 Intake fraction regressions from Greco et al. (*S25*)

The intake fractions for secondary PM from SO_2 and NO_x are evaluated using the regressions of Greco et al. (*S25*):

$$iF(\text{PM}(\text{SO}_2)_{\text{urban and rural}}) = [(P_{<50\text{km}} \times 1.31 \bullet 10^{-13}) + (P_{50\text{–}100\text{km}} \times 3.11 \bullet 10^{-14}) + (P_{100\text{–}200\text{km}} \times 6.92 \bullet 10^{-15}) + (P_{200\text{–}500\text{km}} \times 4.04 \bullet 10^{-15}) + (P_{>500\text{km}} \times 8.35 \bullet 10^{-16})] \times (13/20) \quad \text{(S17)}$$

$$iF(\text{PM}(\text{NO}_x)_{\text{urban and rural}}) = [(P_{<50\text{km}} \times 1.56 \bullet 10^{-14}) + (P_{50\text{–}100\text{km}} \times 4.89 \bullet 10^{-15}) + (P_{100\text{–}200\text{km}} \times 6.44 \bullet 10^{-16}) + (P_{200\text{–}500\text{km}} \times 1.69 \bullet 10^{-16}) + (P_{>500\text{km}} \times 2.75 \bullet 10^{-16})] \times (13/20) \quad \text{(S18)}$$

where $P_{<50\text{km}}$, $P_{50\text{–}100\text{km}}$, $P_{100\text{–}200\text{km}}$, $P_{200\text{–}500\text{km}}$, $P_{>500\text{km}}$ are the populations, respectively, within a radius of 50 km from the location of emission, within a 'donut' of 50-100 km, within a 'donut' of 100-200 km, within a 'donut' of 200-500 km, and further than 500 km, respectively. The ratio at the end adjusts for a breathing

rate of 13 m^3 $person^{-1}$ day^{-1} (*S3*) used in the present study, whereas Greco et al. (*S25*) used originally 20 m^3 $person^{-1}$ day^{-1}. Table S3 presents the populations used in the regressions of Greco et al. (*S25*).

TABLE S3. Populations (in millions) used in the regressions of Greco et al. (*S25*)

	urban case	rural for 100 km, then continental case	remote
$P_{<50km}$	2.6	0.40	0.0078
$P_{50–100km}$	2.4	2.2	0.024
$P_{100–200km}$	9.4	9.4	0.094
$P_{200–500km}$	66	66	0.66
$P_{>500km}$	920	920	9.2
Total continent:	1,000	1,000	10

3.1.2 Intake fraction regressions from Heath et al. (*S26*)

Heath et al. (*S26*) developed two regressions to evaluate the intake fractions for urban and rural emissions of primary $PM_{2.5}$:

$$iF_{urban} = 5.8 \times (P_{100})^{0.5} \quad \text{(S19)}$$

$$iF_{rural} = 114.6 \times (H_E)^{-1.174} \times (P_{100})^{0.838} \quad \text{(S20)}$$

where P_{100}, in millions of persons, is the population within a radius of 100 km of the emitting facility and H_E is the stack height, in meters.

3.2 Intake fraction values

3.2.1 Intake fractions of $PM_{2.5}$ depending on the emission archetype

Table S4 presents the comparison of $PM_{2.5}$ intake fractions of different models. The last column indicates which of the values are reported in Fig. 2.

TABLE S4. Comparison of primary $PM_{2.5}$ intake fractions (ppm) of different models

Model	Urban	Rural	Remote	Emissions-weighted average	*Variation in intake fraction ratio between max and min intake fraction presented in this table)*	Comment	Shown in Fig. 2
Greco et al. (*S25*) counties	8.9 (max = 15 in N.Y.)	1.6	(min = 0.088 in Maine – cannot be considered remote)	2.4	*170*	US, BR adapted to 13 m^3/person·day (original is 20); urban > 2000 person/km2	X
Greco et al. (*S25*) regressions	4.0	2.7 with all 100 persons/km^2; 2.2 with 50 persons/km^2 for 100 km x 100 km	0.027		*130*	BR adapted to 13 m^3/person·day (original is 20)	X
Levy et al. (*S27*)		1.4 [a] (stationary) 5.9 [a] (mobile)				BR adapted to 13 m^3/person·day (original is 20)	X
Levy et al. (*S27*) regressions (stationary)	(2.2 – does not capture urban areas)	2.1	(0.61 – cannot be considered remote)		*>3*	BR adapted to 13 m^3/person·day (original is 20)	X
Levy et al. (*S27*) regressions (mobile)	(19 – does not capture urban areas)	18	(2.0 – cannot be considered remote)		*>8*	BR adapted to 13 m^3/person·day (original is 20)	X
Evans et al. (*S28*)	6.1 (mobile)	1.4 (0.16-4.1) (power plant); 5.7 (mobile)			*38*	based on Wolff (*S29*), US, original BR is 20 m^3/person·day	X
USEtox (*S23*)	26	2.6	0.11	15	*240*	Generic continent	X
RiskPoll (*S24, S30-32*), transportation	41.0	8.20	0.041		*1,000*		
RiskPoll (*S24, S30-32*), low stack (25 m)	14.1	4.32	0.041		*340*		
RiskPoll (*S24, S30-32*), high stack (100 m)	10.7	3.51	0.041		*260*		
RiskPoll (*S24, S30-32*), very high stack (250 m)	5.77	2.78	0.041		*140*		
RiskPoll (*S24, S30-32*), emission-weighted average stack height (using transportation, low and high stacks)	24	5.62	0.041	16	*590*		X
Krewitt (*S33*) (for the reference year 2010)		1.7				Europe	X
Hofstetter (*S34*)		5.7				Europe	X
Van Zelm et al. (*S15*)				4.9		Europe, as PM_{10}, 90 persons/km^2	X
Marshall et al. (*S8*) (intra-urban only)	37					Los Angeles	
Marshall et al. (*S8*) regressions (intra-urban only)	14					BR adapted to 13 m^3/person·day (original is 12.2) and population to $1.6 \cdot 10^6$ persons	X
Heath (*S26*)	18	0.78			*23*	US, original values for California cities and rural areas	
Heath (*S26*) regressions (adapted with average world conditions)	13	1.3	0.031		*420*	Regression developed originally for US, sensitive to the stack height, BR	X

NEEDS (*S35-37*)				15.3	*17*	adapted to 13 m^3/person·day (original is 20) Adjusted European values for global conditions	X
Overall variation in intake fraction (ratio between max and min intake fraction presented in this table)	*15*	*18*	*3*	*30*			

[a] Although these values are weighted averages of intake fraction estimates using relative emissions in United States areas, these weights are not based on urban or rural differences and therefore cannot be considered to be the "average" intake fraction..

For urban areas, the breathing rate-corrected intake fraction for urban emissions provided by the regressions from Greco et al. (*S25*) is 4.0 ppm, assuming average global urban parameters (8,300 persons km^{-2} over 15.5 km x 15.5 km) surrounded by a continental region (100 persons km^{-2}). This intake fraction is lower than the 26 ppm found with USEtox (*S23*), assuming the same urban and continental conditions. This lower intake fraction is largely because the Greco et al. (*S25*) model is based on an arithmetic average dilution rate of 3,000 m^2 s^{-1}, which is 5 times higher than the urban harmonically averaged dilution rate of 610 m^2 s^{-1} used in USEtox (see Section 2.4 of the main text). When correcting for this factor, the model of Greco et al. (*S25*) obtains a more similar intake fraction of 20 ppm.

3.2.2 Intake fractions for other pollutants

Table S5 presents a summary of emission-weighted average intake fractions for primary PM_{10}, primary $PM_{10-2.5}$ and primary $PM_{2.5}$, secondary ammonium particles from SO_2, from NO_x, and from NH_3 from different models.

TABLE S5. Summary of intake fractions (ppm) for primary PM_{10}, primary $PM_{10-2.5}$, primary $PM_{2.5}$, secondary PM from SO_2, secondary PM from NO_x, and secondary PM from NH_3, adjusted for a breathing rate of 13 m^3/person·day

Model	Intake fractions for: PM_{10}	$PM_{10-2.5}$	$PM_{2.5}$	secondary PM from SO_2	secondary PM from NO_x	secondary PM from NH_3	Comment
Units	kg PM_{10}/ kg PM_{10}	kg $PM_{10-2.5}$/ kg $PM_{10-2.5}$	kg $PM_{2.5}$/ kg $PM_{2.5}$	kg $PM(SO_2)$/ kg SO_2	kg $PM(NO_x)$/ kg NO_x	kg $PM(NH_3)$/ kg NH_3	
Greco et al. (*S25*) counties (mobile)			1.6	0.38	0.055		**BR** adapted "continental" (median); US conditions
Greco et al. (*S25*) regressions (mobile)			2.9	0.85	0.18		**BR** adapted emission weighted average
			2.7	0.83	0.18		**BR** adapted "continental" (median)
Levy et al. (*S27*) (stationary)			1.4	0.14	0.023		
Levy et al. (*S27*) (mobile)			5.9	0.12	0.020		**BR** adapted to 13 m^3/person·day (original is 20); US conditions for the first two lines
Levy et al. (*S27*) regression (stationary)			1.4	0.14	0.011		
Levy et al. (*S27*), regression (mobile)			9.7	0.18	0.016		
Evans et al. (*S28*) (stationary)			1.4	0.10	0.018		
Evans et al. (*S28*) (mobile) (urban)			6.1	0.078	0.015		based on Wolff (*S29*); US conditions
Evans et al. (*S28*) (mobile) (rural)			5.7	0.091	0.017		
USEtox (*S23*)			15				
RiskPoll (*S24, S30-32*)	21 [a]	22 [b]	15 [c]	0.77	0.76		emission and stack weighted average
Krewitt (*S33*) (for the reference year 2010)	1.7			0.60	1.4		'continental'; EU conditions
Hofstetter (*S34*)	5.7			0.50	1.3	0.30	'continental'; EU conditions
Van Zelm et al. (*S15*)	4.9			0.93	1.0	1.5	emission-weighted average, 90 persons/km^2; EU conditions
Marshall et al. (*S8*) regressions (intra-urban only)			14				**BR** adapted to 13 m^3/person·day (original is 12.2) and population to $1.6 \cdot 10^6$; US conditions
NEEDS (*S35-37*)		6	15.3	0.94	0.76		emission-weighted average; EU conditions

[a] Intake fractions of PM_{10} (ppm): Urban: 4.37 (for 250 m), 9.28 (for 100 m), 13.4 (for 25 m), 38.5 (for transportation); Rural: 1.87 (for 250 m), 2.39 (for 100 m), 3.22 (for 25 m), 8.1 (for transportation); Remote: 0.027.

[b] Intake fractions of $PM_{10-2.5}$ (ppm): Urban: 3.25 (for 250 m), 8.22 (for 100 m), 12.3 (for 25 m), 37.3 (for transportation); Rural: 1.18 (for 250 m), 1.53 (for 100 m), 2.32 (for 25 m), 7.9 (for transportation); Remote: 0.017.

[b] Intake fractions of $PM_{2.5}$ (ppm): Urban: 5.77 (for 250 m), 10.7 (for 100 m), 14.1 (for 25 m), 41.0 (for transportation); Rural: 2.78 (for 250 m), 3.51 (for 100 m), 4.32 (for 25 m), 8.20 (for transportation); Remote: 0.041.

3.2.3 Size of secondary ammonium particulates

As described in the main text, we focus on fine secondary ammonium particulates ($PM_{2.5}$). For NH_3 emissions, we assume all resulting ammonium particulates are smaller than 2.5 μm based on the values of 1 in Liu et al. (*S43*) and Fine et al. (*S44*), and the value of 0.96 in Chow et al. (*S45*). Liu et al. measurements were taken in the Pearl River Delta of China during polluted periods, Fine et al. measurements were from the Pittsburgh summer and winter, and Chow et al. measurements were taken in Fresno and Angiola during the winter. For SO_2 and NO_x emissions, intake fractions are only available for fine PM, because Greco et al. (*S25*) use a model that calculates the incremental ammonium $PM_{2.5}$ concentration changes given incremental emission changes.

In some circumstances, the coarse fraction of secondary ammonium particulates can be non-negligible (*S42*). Clarke et al. (*S39*) reported average fine fractions of 0.88 for ammonium sulfates and 0.81 for ammonium nitrates, but based on a cutoff of 2.1 instead of 2.5 μm, and only represent locations near Leeds.

Coarse ammonium particulates are not addressed in the main paper. However, the attributable health damage from ammonium PM mass is expected to be much smaller for secondary $PM_{10-2.5}$ than for secondary $PM_{2.5}$, owing to the small coarse fraction of ammonium particulates and the attribution of most PM health effects to fine particles (*S46, S34, S47, S48*).

3.3 Recommended intake fractions

Table S6 summarizes the models and assumptions used for the recommended intake fractions presented in Table 3. In summary, USEtox (*S23*) is used for primary PM, Greco et al. (*S25*) for secondary PM from SO_2 and NO_x, Van Zelm et al. (*S15*) for secondary PM from NH_3 and RiskPoll (*S24*) to differentiate between high-stack, low-stack, and ground-level emissions of primary PM for urban and rural conditions, respectively.

TABLE S6. Summary of models or assumptions used for the recommended intake fractions of $PM_{10-2.5}$, $PM_{2.5}$, SO_2, NO_x, and NH_3

Pollutant emitted	Type of source for the PM emission :	Urban	Rural	Remote	Population-weighted average
$PM_{10-2.5}$	High-stack Low-stack Ground-level	= $iF(PM_{2.5})$× ratios of $iF(PM_{10-2.5})/iF(PM_{2.5})$ from RiskPoll			Weighted average among urban, rural and remote emissions
	Emission-weighted average	Weighted average among high-, low-stack, and ground-level (based on Table S2)			
$PM_{2.5}$	High-stack Low-stack Ground-level	Re-derived from unknown intake fraction, using i) urban ratio from RiskPoll, and ii) weighted average among high-stack, low-stack and ground-level	Re-derived from unknown intake fraction, using i) rural ratio from RiskPoll, and ii) weighted average among high-stack, low-stack and ground-level	USEtox (with parameters adapted), no difference among high-stack, low-stack, and ground-level	Weighted average among urban, rural, and remote emissions
	Emission-weighted average	USEtox (with urban parameters adapted)	USEtox (with rural parameters adapted)	Weighted average among high stack-, low-stack, and ground-level (based on Table S2)	
SO_2	All (assume all heights the same based on Levy et al. (*S27*))	Greco et al. (*S25*) for urban (with parameters adapted)	Greco et al. (*S25*) for rural (with parameters adapted)	based on rural iF, using same ratio as for $PM_{2.5}$ between rural and unknown	Weighted average among urban, rural and remote emissions
NO_x	All (assume all heights the same based on Levy et al. (*S27*))	Greco et al. (*S25*) for urban (with parameters adapted)	Greco et al. (*S25*) for rural (with parameters adapted)	based on rural iF, using same ratio as for $PM_{2.5}$	Weighted average among urban, rural and remote emissions
NH_3	All	= rural intake fraction, assuming no difference in intake fraction between rural and urban emissions	Van Zelm et al. (*S15*) corrected by difference in pop density, assuming no difference among high-stack, low-stack and ground-level	based on rural intake fraction, using same ratio as for $PM_{2.5}$	Weighted average among urban, rural and remote emissions

Legend for the cell colors

Based on (re-parameterized) model	*Equalized or derived from other values, based on strong evidence or models*	*Weighted average*	*Equalized or derived from other values, based on weak evidence or models*

Table S7 provides the equations behind each of the intake fractions provided in Table 3.

TABLE S7. Summary of equations used for the recommended intake fractions of $PM_{10-2.5}$, $PM_{2.5}$, SO_2, NO_x, and NH_3

Pollutant emitted	Type of source for the PM emission :	Urban	Rural	Remote	Population-weighted average
$PM_{10-2.5}$	High-stack Low-stack Ground-level	$= iF(PM_{2.5\ urban}) \times \{iF(PM_{10-2.5})/iF(PM_{2.5})$ from RiskPoll for urban, for the respective source$\}$	$= iF(PM_{2.5\ rural}) \times \{iF(PM_{10-2.5})/iF(PM_{2.5})$ from RiskPoll for rural, for the respective source$\}$	$= iF(PM_{2.5\ remote}) \times \{iF(PM_{10-2.5})/iF(PM_{2.5})$ from RiskPoll for remote, for the respective source$\}$	*= Eq. 6*
	Emission-weighted average	*= Eq. S11*			
$PM_{2.5}$	High-stack Low-stack Ground-level	*= Eq. S14* *= Eq. S15* *= Eq. S16*	*= Eq. S14* *= Eq. S15* *= Eq. S16*	*= Eq. S7*	*= Eq. 6*
	Emission-weighted average	*= Eq. S10*	*= Eq. S8*	*= Eq. S11*	
SO_2	All (assume all heights the same based on Levy et al. (*S27*))	*= Eq. S17*	*= Eq. S17*	$= iF(SO_{2\ rural}) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	*= Eq. 6*
NO_x	All (assume all heights the same based on Levy et al. (*S27*))	*= Eq. S18*	*= Eq. S18*	$= iF(NO_{x\ rural}) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	*= Eq. 6*
NH_3	All	$= iF_{rural}$	Van Zelm et al. (15) × (100/90), assuming no difference between high-stack and low-stack [a]	$= iF(NH_{3\ rural}) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	*= Eq. 6*

Legend for the cell colors

Based on (re-parameterized) model	*Equalized or derived from other values, based on strong evidence or models*	*Weighted average*	*Equalized or derived from other values, based on weak evidence or models*

[a] As a first approximation, this value can be adapted to a specific 'rural' situation by multiplying it by (x/100), with x being the population density (persons/km^2) of the rural area under consideration and 100 (persons/km^2) being the population density of the generic rural area.

4 Variability and uncertainty

Accounting for the emission-specific population density reduces the variability (not the uncertainty) of the estimated intake fraction, which in turn reduces the uncertainty of the characterization factors and the life cycle assessment (LCA) results. Variability can become a source of uncertainty if it is not accounted for in the calculation. One of the main constraints in life cycle assessment regarding regionalization is that most of the inventories of background processes do not give information (nor even the option to give information) on the country of emission, providing only information on the archetype (e.g., high or low population density) where emissions occur. Certain specific processes may include the country of origin, but life cycle assessment software typically do not retain that information when aggregating inventories to perform the impact assessment. Since aggregated inventories are still

distinguished by archetype, the recommended intake fractions presented here can be directly used within current life cycle assessment constraints (including life cycle assessment software).

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