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

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

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Abstract

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

Keywords

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

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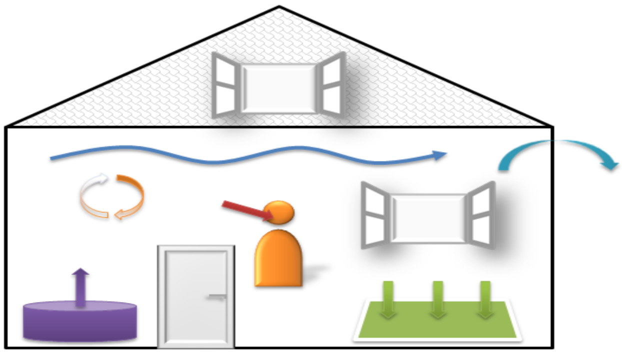
TOC/Abstract Art



Introduction

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

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

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

USEtox is a tool for calculation of comparative toxicity potentials (characterization factors) for human health and freshwater ecosystems, developed under the auspices of the UNEP/SETAC Life Cycle Initiative. It models a cause–effect chain that links emissions to impacts through three steps: environmental fate, exposure, and effects. It was developed as a methodology simple enough to be used on a worldwide basis and for a large number of substances while incorporating broad scientific consensus.[12,13] It is the recommended LCA (midpoint) toxicity characterization model of the European Union[14], endorsed by the UNEP/SETAC Life Cycle Initiative, and adopted by the US-EPA's life cycle impact assessment tool TRACI.[15] Therefore, it is regarded as the relevant basis to integrate indoor and outdoor exposure characterization into one consistent method for use in LCA, as also discussed by Hellweg et al.[6]

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The aims of this paper are 1) extending the USEtox model[13] to include the indoor environment as a compartment; 2) providing an overview of recommended parameter values to be used as default household model parameters for different geographical settings; 3) comparing intake fractions calculated with these recommended default parameters with intake fractions for outdoor exposure; and, 4) applying the new characterization factors for indoor exposure to a comprehensive case study on cooking worldwide. The scope of this paper is restricted to the LCA emitter perspective, i.e. the calculation of potential health effects from indoor emissions modeled as the cumulative impacts from indoor exposure and outdoor exposure due to indoor emissions only. The focus was on indoor emissions of volatile and semi-volatile organic compounds, because pollutants such as particles, ozone or NOx require specific model processes for transport and transformation and are currently not addressed by LCA toxicity models and not included in USEtox. In LCA their impacts on human health are assessed respectively in the separate impact categories "particulate matter formation" and "photochemical ozone formation".

Materials and Method

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

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

where iF is the population intake fraction of a chemical (-), IR is the daily inhalation rate of air of an individual (m^3/day), N is the number of people exposed (-),V is the volume of the exposure area (m^3), k_{ex} is the air exchange rate of the volume in the exposure area (-) and m is the mixing factor (-). The following sections describe how this has been implemented into the matrix-algebra framework of the USEtox model.[13]

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

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

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

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

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

$$\mathbf{FF} = \left(-\mathbf{K}\right)^{-1} \qquad (3)$$

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

1) The advective ventilation flow, parameterized as the air exchange rate k_{ex} [h^{-1}] (as in Equation 1b in [6]). The air exchange rate does not depend on the substance, but on the building characteristics, such as type and size of windows and doors, type of walls, and the number of cracks in the façades. Average values for several regions are given in Table S1 in the Supporting Information (SI). k_{ex} is not a loss, but an inter-media transport mechanism connecting indoor with outdoor compartments. Based on the average distribution of the global population between urban and rural areas of about 50% respectively[18],

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

2) The gas-phase (g) air-degradation rate $k_{g,\deg}$ [h^{-1}] is mainly related to reactions with ozone, hydroxyl radicals, and nitrate radicals (gas-phase degradation). The overall degradation rate in the indoor air is calculated as the average radical concentration ([OH], [O_3], [NO_3]) multiplied by the corresponding second order degradation rate constant: $k_{g,\deg} = k_{OH} \cdot [OH] + k_{O_3} \cdot [O_3] + k_{NO_3} \cdot [NO_3]$. Long-term averaged indoor concentrations of ozone ([O_3] = 8 ppb), hydroxyl radical ([OH] = 3×10^{-6} ppb) and nitrate radical ([NO_3] = 10^{-3} ppb) were taken from Wenger at al.[17] and second order degradation rate constants from the EPI Suite v4.1 software[19], which provides OH rate constants for most substances, but only few for O_3 and NO_3.

3) An equivalent removal rate by adsorption to indoor surfaces, k_s [h^{-1}] can be calculated as a net removal rate from the air, assuming steady-state conditions between the air and room surface without adding a separate compartment.[17] This approach is similar to the net removal rate calculated in USEtox from the freshwater outdoor environment to the sediments, which are not considered as separate compartments to limit the model complexity.[20] Since degradation on surfaces is not well characterized, this removal rate to surfaces is subject to high uncertainty. Surface removal in the current model is applied primarily to Semi-Volatile Organic Compounds (SVOCs), for which additional gaseous dermal exposure may also be relevant and may compensate this removal. If the model is eventually used for particulate matter (PM) and ozone, then surface removal could become more important and requires further assessment of the literature on indoor ozone and PM deposition including the work of Weschler[21] and Nazaroff[22]. We therefore do not include the sorption removal pathway in the default model, but only consider it for the sensitivity study together with the dermal gaseous exposure pathway. A more detailed description of the calculation of the equivalent removal rate to the surface k_s is given in SI (section S3).

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

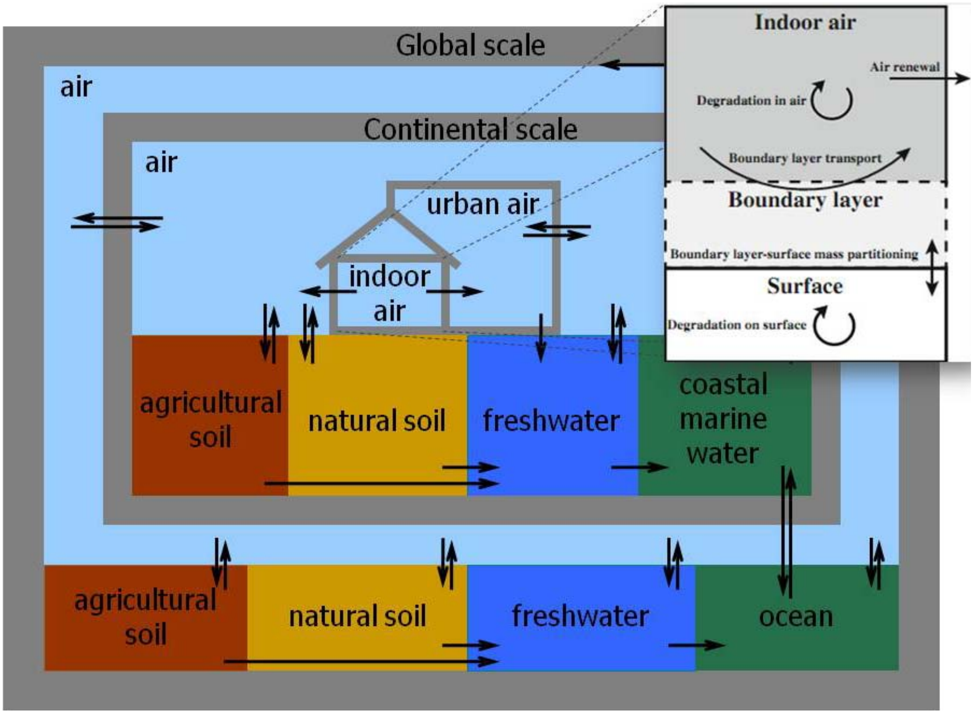

Figure 1: Schematic representation of the USEtox model with indoor compartment embedded; adapted from Rosenbaum et al.[13] and Wenger et al.[17]

Exposure: The exposure pathway considered in this paper is inhalation. The relevant parameters for inhalation exposure in households are the following: individual daily inhalation (breathing) rate (IR) [m^3/d], average number of people in the building N [dimensionless], building volume V [m^3], and daily time fraction spent indoors f_t [dimensionless]. The latter is the quotient of the time spent indoors and the total time of a day (24h). Recommendations, assumptions, and choices for these parameter values are further discussed below. The exposure factor XF [1/d] for the indoor exposure setting is then calculated based on Equation 1b in Hellweg et al.[6] (with mixing factor m = 1, assuming that complete mixing within the indoor volume is an inherent hypothesis of the indoor iF model):

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

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

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that the validity of equation (4) is restricted to VOCs.[23–25] In this paper the potential influence of the dermal gaseous uptake pathway is considered as a sensitivity study together with the influence of adsorption removal on indoor surfaces which competes with this exposure pathway. Existing approaches[17,26] were adapted to determine the convective transfer at body surface as a function of heat transfer coefficients[27], which might be added to USEtox in a later stage once data will be broadly available and the models further evaluated, in conjunction with the introduction of a dermal pathway within USEtox.

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

Model Parameterization

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

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

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

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

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

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

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

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

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

Sensitivity and variability analysis

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

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following parameters used to calculate indoor iF were included in the variability analysis using Monte Carlo simulation with 50,000 iterations and Latin Hypercube Sampling (Crystal Ball 11.1.2): 1) building volume V; 2) number of people in the building N; 3) air exchange rate k_{ex}; 4) individual daily inhalation rate (at home) IR, 5) daily time at home t_{home} (used to calculate the daily time fraction spent indoors f_t). For the values of V and N the sampling method has been adapted to reflect the dependency between these parameters: for each Monte Carlo run, a corresponding set of values for N and V for one country was selected out of their discrete distribution over all countries, with a probability-weighting based on its population. The average individual inhalation rate at rest for households was sampled from the reported interval of 0.44-1.04 m^3/h [36] assuming a beta distribution between these limits. The air exchange rate (k_{ex}) was sampled from a discrete distribution representing L-AER and H-AER buildings respectively from various countries using a probability-weighting based on their respective population. The daily time at home was assumed to be normally distributed with an assumed standard deviation of 2, resulting in a 95% confidence interval ranging from 10 to 18 hours per day at home. For further details and values the reader is referred to SI.

Case Study

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

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according to equation (2),[13] extended to endpoint results expressed as Disability Adjusted Life Years (DALY) using the following disability weights: 11.5 DALY/CTU_h for cancerous effects and 2.7 DALY/CTU_h for non-cancerous effects (CTU_h – Comparative Toxic Unit for humans[13] corresponding to cases of cancer or of non-cancer).[40] Respiratory inorganics impacts of $PM_{2.5}$, NO_x, SO_x and NH_3 were estimated using the effect and characterization factors from Gronlund et al.[41] The direct emissions are displayed in Table S2 of SI together with further details on the background processes given in section S2 of SI.

Results

Intake fractions and characterization factors

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

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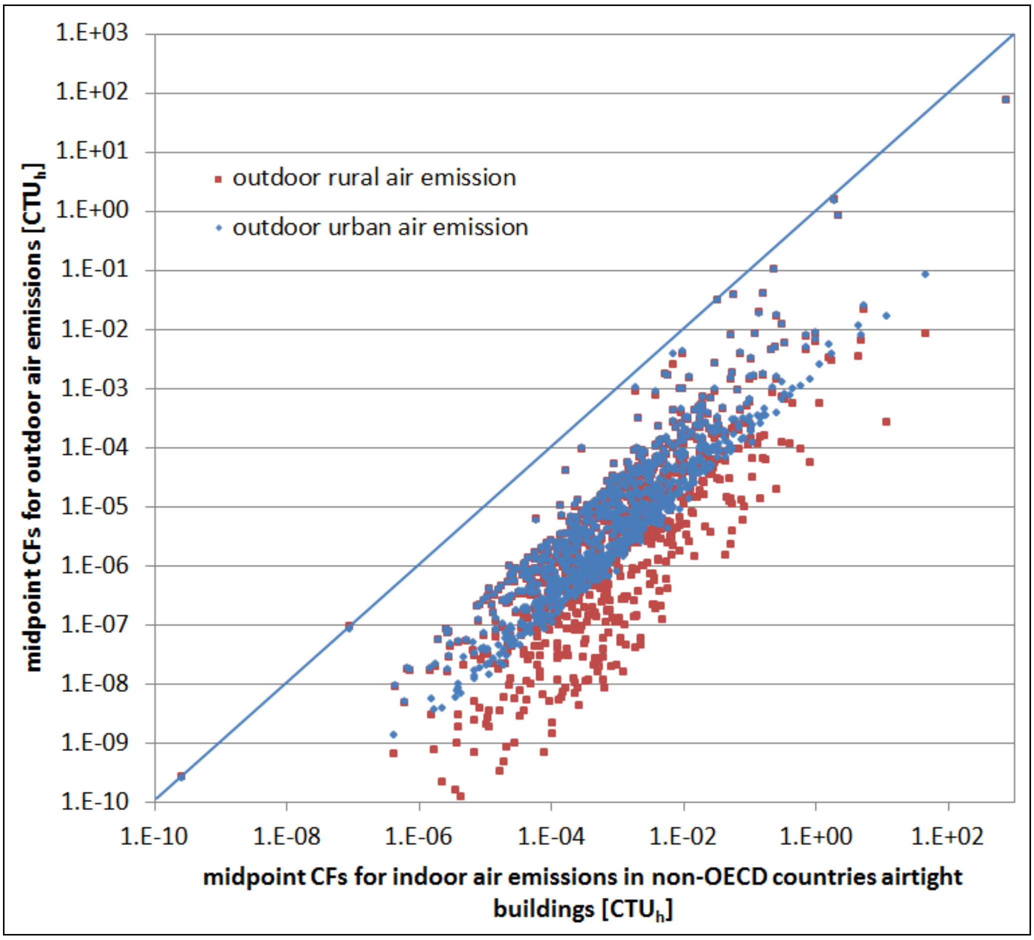

Figure 2: Comparison of characterization factors (CFs) for indoor emissions in non-OECD countries and L-AER buildings (x-axis) relative to CFs for continental urban and rural outdoor emissions (y-axis); the difference between indoor and outdoor iFs is smaller for the other regions

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

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

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

[a]single data point (US average) as we are using country averages and hence no variability assessed on sub-country level

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

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

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indoor air advective removal, degradation, and inhalation pathways in the default model at this stage. More details on the sensitivity study can be found in section S3 of SI.

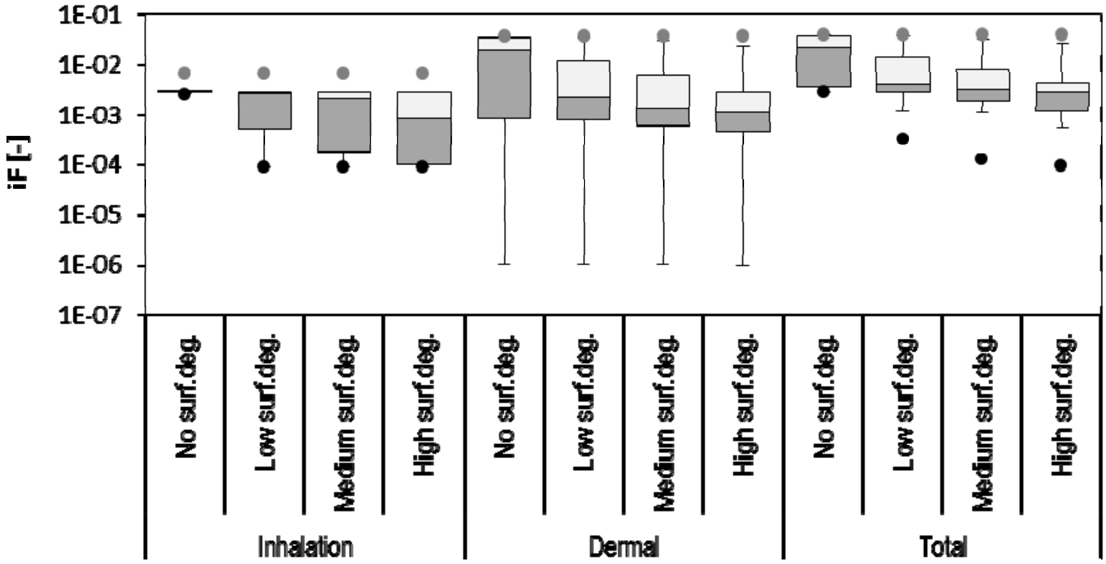

Figure 3: Variations in indoor intake fractions for the 3073 organic substances in the USEtox 1.01 database considering the inhalation, dermal gaseous and sum of these two exposure pathways with four assumptions: No, low, medium, and high surface degradation rates following sorption, respectively corresponding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

Case study: world cooking

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

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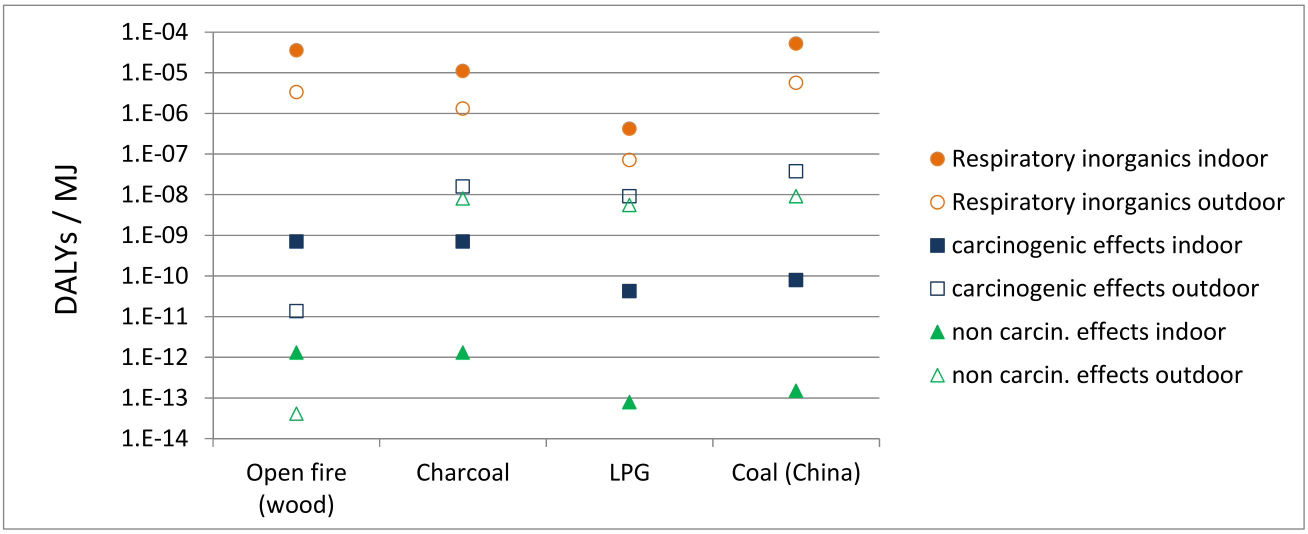

Figure 4: Human health impacts in DALY from indoor and outdoor exposure

Discussion

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

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

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

With the indoor exposure model implemented in USEtox and the resulting characterization factors, it is now operational to integrate household indoor exposure to substances into life cycle assessment studies. Both, iF and characterization factors calculated in this study are based on the still sparse data sources available, which highly influenced the number of regions that could be defined. When more data become available the definitions of regions should be revised in order to better represent global variability, and the iF and characterization factors should be updated. Meanwhile, the parameters in Table 1 for the OECD and non-OECD scenarios are recommended for LCA application of Hellweg et al.'s one-box indoor exposure model. Since the present intake fractions are based on average occupancy and continuous emission, further efforts are needed in the future to better assess emissions with non-continuous sources related to the nexus of occupant and source activity patterns (e.g. cooking), in particular emission patterns that involve near-person releases. Another refinement would be to account for substance removal by filters in centrally air-conditioned buildings, a region-specific removal rate that may be substantial in hot climate. Moreover, whereas degradation was not an important removal process we underline that impacts from the products of homogenous reactions in air or other degradation processes may have significant impacts[42–44] but are not taken into account in the CFs calculated by this research work. According to current practice, LCA practitioners can take them into account by adding the amount of reaction products generated from a parent compound to the life cycle emission inventory and characterize them with their corresponding characterization factors.

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

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

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

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

Supporting information

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

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References

(1) Hellweg, S.; Milà i Canals, L. Emerging approaches, challenges and opportunities in life cycle assessment. *Sci.* **2014**, *344* , 1109–1113.

(2) Keller, D.; Wahnschaffe, U.; Rosner, G.; Mangelsdorf, I. Considering human toxicity as an impact category in Life Cycle Assessment. *Int. J. Life Cycle Assess.* **1998**, *3*, 80–85.

(3) Jönsson, Å. Is it feasible to address indoor climate issues in LCA? *Environ. Impact Assess. Rev.* **2000**, *20*, 241–259.

(4) Meijer, A.; Huijbregts, M. A. J.; Reijnders, L. Human health damages due to indoor sources of organic compounds and radioactivity in life cycle impact assessment of dwellings. Part 2: Damage scores. *Int. J. Life Cycle Assess.* **2005**, *10*, 383–392.

(5) Meijer, A.; Huijbregts, M. A. J.; Reijnders, L. Human health damages due to indoor sources of organic compounds and radioactivity in life cycle impact assessment of dwellings - Part 1: Characterisation factors. *Int. J. Life Cycle Assess.* **2005**, *10*, 309–316.

(6) Hellweg, S.; Demou, E.; Bruzzi, R.; Meijer, A.; Rosenbaum, R. K.; Huijbregts, M. A. J.; McKone, T. E. Integrating Indoor Air Pollutant Exposure within Life Cycle Impact Assessment. *Environ. Sci. Technol.* **2009**, *43*, 1670–1679.

(7) Kikuchi, Y.; Hirao, M. Local risks and global impacts considering plant-specific functions and constraints: A case study of metal parts cleaning. *Int. J. Life Cycle Assess.* **2010**, *15*, 17–31.

(8) Skaar, C.; Jørgensen, R. B. Integrating human health impact from indoor emissions into an LCA: A case study evaluating the significance of the use stage. *Int. J. Life Cycle Assess.* **2013**, *18*, 636–646.

(9) Collinge, W.; Landis, A. E.; Jones, A. K.; Schaefer, L. A.; Bilec, M. M. Indoor environmental quality in a dynamic life cycle assessment framework for whole buildings: Focus on human health chemical impacts. *Build. Environ.* **2013**, *62*, 182–190.

(10) Demou, E.; Hellweg, S.; Wilson, M. P.; Hammond, S. K.; Mckone, T. E. Evaluating indoor exposure modeling alternatives for LCA: A case study in the vehicle repair industry. *Environ. Sci. Technol.* **2009**, *43*, 5804–5810.

(11) Chaudhary, A.; Hellweg, S. Including Indoor Offgassed Emissions in the Life Cycle Inventories of Wood Products. *Environ. Sci. Technol.* **2014**, *48*, 14607–14614.

(12) Hauschild, M. Z.; Huijbregts, M. A. J.; Jolliet, O.; MacLeod, M.; Margni, M.; Van de Meent, D.; Rosenbaum, R. K.; McKone, T. E. Building a model based on scientific consensus for Life Cycle Impact Assessment of Chemicals: the Search for Harmony and Parsimony. *Environ. Sci. Technol.* **2008**, *42*, 7032–7037.

(13) Rosenbaum, R. K.; Bachmann, T. M. K.; Gold, L. S.; Huijbregts, M. A. J.; Jolliet, O.; Juraske, R.; Koehler, A.; Larsen, H. F.; MacLeod, M.; Margni, M.; et al. USEtox - The UNEP/SETAC-consensus model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in Life Cycle Impact Assessment. *Int. J. Life Cycle Assess.* **2008**, *13*, 532–546.

(14) EC-JRC. *International Reference Life Cycle Data System (ILCD) Handbook - Recommendations for Life Cycle Impact Assessment in the European context*; First edit.; European Commission, Joint Research Centre, Institute for Environment and Sustainability: Ispra, Italy, 2011.

(15) Bare, J. TRACI 2.0: the tool for the reduction and assessment of chemical and other environmental impacts 2.0. *Clean Technol. Environ. Policy* **2011**, *13*, 687–696.

(16) Rosenbaum, R. K.; Margni, M.; Jolliet, O. A flexible matrix algebra framework for the multimedia multipathway modeling of emission to impacts. *Environ. Int.* **2007**, *33*, 624–634.

(17) Wenger, Y.; Li, D. S.; Jolliet, O. Indoor intake fraction considering surface sorption of air organic compounds for life cycle assessment. *Int. J. Life Cycle Assess.* **2012**, *17*, 919–931.

Author-produced version of the article published in Environmental Science & Technology, 2015, N°49(21), p. 12823-12831
The original publication is available at http://pubs.acs.org
Doi: 10.1021/acs.est.5b00890

(18) UN. *World Urbanization Prospects: The 2011 Revision*; New York, USA, 2011.

(19) US EPA. Estimation Programs Interface EPI Suite Version 4.11, 2012.

(20) Henderson, A.; Hauschild, M. Z.; Van de Meent, D.; Huijbregts, M. A. J.; Larsen, H. F.; Margni, M.; McKone, T. E.; Payet, J.; Rosenbaum, R. K.; Jolliet, O. USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties. *Int. J. Life Cycle Assess.* **2011**, *16*, 701–709.

(21) Weschler, C. J. Ozone in Indoor Environments: Concentration and Chemistry. *Indoor Air* **2000**, *10*, 269–288.

(22) Nazaroff, W. W. Indoor particle dynamics. *Indoor Air* **2004**, *14*, 175–183.

(23) Weschler, C. J.; Nazaroff, W. W. Dermal Uptake of Organic Vapors Commonly Found in Indoor Air. *Environ. Sci. Technol.* **2013**, *48*, 1230–1237.

(24) Gong, M.; Zhang, Y.; Weschler, C. J. Predicting dermal absorption of gas-phase chemicals: transient model development, evaluation, and application. *Indoor Air* **2014**, *24*, 292–306.

(25) Weschler, C. J.; Nazaroff, W. W. SVOC exposure indoors: fresh look at dermal pathways. *Indoor Air* **2012**, *22*, 356–377.

(26) Tibaldi, R.; ten Berge, W.; Drolet, D. Dermal Absorption of Chemicals: Estimation by IH SkinPerm. *J. Occup. Environ. Hyg.* **2013**, *11*, 19–31.

(27) Csiszar, S. A.; Ernstoff, A. S.; Fantke, P.; Jolliet, O. Stochastic modeling of near-field exposure to parabens in personal care products. *Submitt. Rev.*

(28) You, Y.; Niu, C.; Zhou, J.; Liu, Y.; Bai, Z.; Zhang, J.; He, F.; Zhang, N. Measurement of air exchange rates in different indoor environments using continuous CO2 sensors. *J. Environ. Sci.* **2012**, *24*, 657–664.

(29) Massey, D.; Kulshrestha, A.; Masih, J.; Taneja, A. Seasonal trends of PM10, PM5.0, PM2.5 & PM1.0 in indoor and outdoor environments of residential homes located in North-Central India. *Build. Environ.* **2012**, *47*, 223–231.

(30) Rotko, T.; Oglesby, L.; Künzli, N.; Jantunen, M. J. Population sampling in European air pollution exposure study, EXPOLIS: Comparisons between the cities and representativeness of the samples. *J. Expo. Anal. Environ. Epidemiol.* **2000**, *10*, 355–364.

(31) Hänninen, O. O.; Alm, S.; Katsouyanni, K.; Künzli, N.; Maroni, M.; Nieuwenhuijsen, M. J.; Saarela, K.; Srám, R. J.; Zmirou, D.; Jantunen, M. J. The EXPOLIS study: Implications for exposure research and environmental policy in Europe. *J. Expo. Anal. Environ. Epidemiol.* **2004**, *14*, 440–456.

(32) Schweizer, C.; Edwards, R.; Bayer-Oglesby, L.; Gauderman, W.; Ilacqua, V.; Jantunen, M.; Lai, H.; Nieuwenhuijsen, M.; Künzli, N. Indoor time-microenvironment-activity patterns in seven regions of Europe. *J. Expo. Sci. Environ. Epidemiol.* **2007**, *17*, 170–181.

(33) Klepeis, N. E.; Nelson, W. C.; Ott, W. R.; Robinson, J. P.; Tsang, A. M.; Switzer, P.; Behar, J. V; Hern, S. C.; Engelmann, W. H. The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. *J. Expo. Sci. Environ. Epidemiol.* **2001**, *11*, 231–252.

(34) Leech, J. A.; Nelson, W. C.; Burnett, R. T.; Aaron, S.; Raizenne, M. E. It's about time: A comparison of Canadian and American time-activity patterns[dagger]. *J. Expo. Sci. Environ. Epidemiol.* **2002**, *12*, 427–432.

(35) Smith, K. R. Looking for pollution where the people are. In *AsiaPacific issues no. 10*; East-West Center: Honolulu, Hawaii, USA, 1994.

(36) Nazaroff, W. W. Inhalation intake fraction of pollutants from episodic indoor emissions. *Build. Environ.* **2008**, *43*, 269–277.

(37) Lim, S. S.; Vos, T.; Flaxman, A. D.; Danaei, G.; Shibuya, K.; Adair-Rohani, H.; Amann, M.; Anderson, H. R.; Andrews, K. G.; Aryee, M.; et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of Disease Study 2010. *Lancet* **2012**, *380*, 2224–2260.

(38) Census of India. CensusInfo India 2011 http://www.devinfolive.info/censusinfodashboard/website/index.php/pages/kitchen_fuelused/Total/insidehouse/IND.

(39) ecoinvent Centre. ecoinvent data v2.1, 2007.

(40) Huijbregts, M. A. J.; Rombouts, L. J. A.; Ragas, A. M. J.; Van de Meent, D. Human-Toxicological Effect and Damage Factors of Carcinogenic and Noncarcinogenic Chemicals for Life Cycle Impact Assessment. *Integr. Environ. Assess. Manag.* **2005**, *1*, 181–192.

(41) Gronlund, C.; Humbert, S.; Shaked, S.; O'Neill, M.; Jolliet, O. Characterizing the burden of disease of particulate matter for life cycle impact assessment. *Air Qual. Atmos. Heal.* **2015**, *8*, 29–46.

(42) Terry, A. C.; Carslaw, N.; Ashmore, M.; Dimitroulopoulou, S.; Carslaw, D. C. Occupant exposure to indoor air pollutants in modern European offices: An integrated modelling approach. *Atmos. Environ.* **2014**, *82*, 9–16.

(43) Kim, S.; Hong, S.-H.; Bong, C.-K.; Cho, M.-H. Characterization of air freshener emission: the potential health effects. *J. Toxicol. Sci.* **2015**, *40*, 535–550.

(44) Rohr, A. C. The health significance of gas- and particle-phase terpene oxidation products: a review. *Environ. Int.* **2013**, *60*, 145–162.

(45) Banerjee, S. G.; Bhatia, M.; Azuela, G. E.; Jaques, I.; Sarkar, A.; Portale, E.; Bushueva, I.; Angelou, N.; Inon, J. G. *Global tracking framework: Sustainable energy for all*; Washington, DC, USA, 2013.

(46) Wilkinson, P.; Smith, K. R.; Joffe, M.; Haines, A. A global perspective on energy: health effects and injustices. *Lancet* **2007**, *370*, 965–978.