Integrating Human Indoor Air Pollutant Exposure within Life Cycle Impact Assessment

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Neglecting health effects from indoor pollutant emissions and exposure, as currently done in Life Cycle Assessment (LCA), may result in product or process optimizations at the expense of workers' or consumers' health. To close this gap, methods for considering indoor exposure to chemicals are needed to complement the methods for outdoor human exposure assessment already in use. This paper summarizes the work of an international expert group on the integration of human indoor and outdoor exposure in LCA, within the UNEP/ SETAC Life Cycle Initiative. A new methodological framework is proposed for a general procedure to include human-health effects from indoor exposure in LCA. Exposure models from occupational hygiene and household indoor air quality studies and practices are critically reviewed and recommendations are provided on the appropriateness of various model alternatives in the context of LCA. A single-compartment box model is recommended for use as a default in LCA, enabling one to screen occupational and household exposures consistent with the existing models to assess outdoor emission in a multimedia environment. An initial set of model parameter values was collected. The comparison between indoor and outdoor human exposure per unit of emission shows that for many pollutants, intake per unit of indoor emission may be several orders of magnitude higher than for outdoor emissions. It is concluded that indoor exposure should be routinely addressed within LCA.

Introduction

Indoor concentrations of chemicals and resulting human exposures often substantially exceed corresponding outdoor

concentrations, mainly because there are significant indoor emission sources and with much lower dilution volumes. For example, typical concentrations measured for tetrachloroethylene and formaldehyde in the ambient environment are smaller than 9 (1) and 24.6 μ g/m³ (2), respectively, whereas they are several orders of magnitude higher in many industrial or household settings (2, 3). Moreover, people spend most of their time indoors, which for industrial countries amounts to more than 20 h a day on average when considering both time spent at home and at the workplace or school (4). Both aspects often give rise to indoor emission intakes of up to several orders of magnitude higher than outdoor emission intakes (4-6). Nevertheless, health effects from indoor exposure are generally neglected in Life Cycle Assessment (LCA). Such an omission is an important shortcoming, as it may result in product or process optimizations at the expense of workers' or consumers' health.

Recently, there have been significant efforts to integrate indoor exposure models within environmental models commonly applied to LCA. For instance, Meijer et al. (7, 8) developed a model for the assessment of household exposure to chemicals and radiation emitted to indoor air. Hellweg et al. (9) used bulk-mixing models for occupational exposure in conjunction with multimedia models for the assessment of cumulative chemical exposure from ambient and indoor environments. Both studies illustrate that indoor exposure models are compatible with environmental models used in LCA. Moreover, they reveal the significance of health effects associated with occupational and household exposure in comparison to the total human-toxicity potential from all pathways. To capture potentially relevant effects to human health, indoor exposure to pollutants should be considered within LCA.

This paper summarizes the work of an international expert group on the integration of indoor and outdoor exposure in LCA, within the UNEP/SETAC Life Cycle Initiative (http://lcinitiative.unep.fr), which is taking up recommendations and conclusion toward the enhancement of the current LCA framework.

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The goal of this paper is to develop a general methodological framework that allows for the assessment of indoor emissions and human exposure in combination with commonly used outdoor fate and exposure models in LCA. To achieve this objective, (a) we review and evaluate existing indoor exposure models for households and occupational settings concerning their use in LCA, (b) we use these models to provide a methodological framework for the routine assessment of indoor exposure to chemicals within LCA studies, (c) we gather a sample set of model parameter values and illustrate the application of the methodological framework, and (d) compare examples of human intake fractions of indoor and outdoor exposure for a number of chemicals. Intake fractions are defined here as the mass of pollutant inhaled by humans per mass unit of chemical emitted (10).

Methods

Screening Assessment of Indoor Air Exposure Models. In the field of Indoor Air Quality for both residential and occupational environments, a large number of models has been developed for assessing exposure to indoor pollutants, ranging from semiquantitative models (11-15) to physicalprocess models (16-18). In this paper, we address only the latter type of models, as these are also commonly used for outdoor fate and exposure models, to allow for quantitatively comparing assessments of exposure. A model review is performed to enable the LCA practitioner to make an informed choice among suitable model alternatives for use in LCA, depending on the level of detail needed. The first column of Table 1 displays an overview of the model types available for indoor exposure assessments. These models range from simple bulk mixing models (zero-ventilation model, one-box model, two-zone model) to diffusion-based models (eddy diffusion model, Gaussian plume dispersion model) and complex computational fluid dynamics models. A brief description of the model principles is presented in column 2 of Table 1, together with some selected references for in-depth explanations in column 3 (a detailed overview is also presented in 16, 19).

Model performance was evaluated qualitatively, with regard to the capabilities and limitations of each alternative and its appropriateness in the context of LCA. The following criteria were set in assessing the models: accuracy and precision (reliability of the model), transparency (the ability of the model to communicate emissions/exposure relationships), data requirements, and ease of use. In addition, a literature survey was performed on case studies, mainly from occupational hygiene, that compared model calculations to measured concentrations (see the Supporting Information). The purpose of this screening assessment was to narrow down the choice of models to those that are compatible with the environmental fate and exposure models used in LCA. The screening assessment was performed by a team consisting of LCA and risk assessment experts as well as occupational hygienists (UNEP/SETAC working group on the Integration of Indoor Exposure Assessment within LCA, with the authors of this paper and the persons mentioned in the acknowledgment as members).

Equation 1a shows the calculation of intake fractions resulting from indoor pollutant inhalation using the example of the one-box model without eq 1a and with eq 1b correction for incomplete mixing, assuming a constant exposure time.

$$iF = \frac{\partial I_x}{\partial G_x} N = \frac{\partial C_x IR}{\partial G_x} N = \frac{\partial (G_x/Q) IR}{\partial G_x} N = \frac{IR}{Q} N = \frac{IR}{Vk_{\text{ex}}} N \quad \text{(1a)}$$
$$iF = \frac{IR}{Vmk_{\text{ex}}} N \quad \text{(1b)}$$

where iF is the population intake fraction of a chemical (–), I_x is the daily intake of a chemical by an individual (kg/day),

IR is the daily inhalation rate of air of an individual (m³/day), N is the number of people exposed, C_x is the chemical concentration in air (kg/m³), G_x is the emission rate of chemical x (kg/day), Q is the ventilation rate in the exposure area (m³/day), V is the volume of the exposure area (m³), V is the air exchange rate of the volume in the exposure area, and V is the mixing factor.

With regard to the models that were considered appropriate to calculate human intake fractions, a decision tree (Figure 1a) was elaborated based on the model assessment, providing rough guidance for model choice in specific situations.

Outdoor Exposure Model. The great variability between results of current toxicity models often impedes a wide consideration of toxicity impacts in comparative studies. To provide an agreed, consistent, and stable toxicity assessment method for comparative environmental assessments, the USEtox model (20, 21) was developed by an international expert group. USEtox is based on a scientific consensus and thus parsimoniously built from only the most influential model elements identified via an extensive model comparison (20, 21). It is currently being reviewed for endorsement by UNEP/SETAC as the globally recommended model and source to address human-toxicity impacts within LCA. As shown in Figure 1b, the model covers an urban and continental scale that is nested into a global scale accounting for impacts outside the continental scale. The various compartments are interlinked by steady-state exchange flows. The human intake fractions include exposure through inhalation of air, and ingestion of drinking water, leaf crops, root crops, meat, milk, and fish from freshwater and marine aquatic compartments, for the total human population.

Combining Indoor and Outdoor Exposure Assessments. The indoor exposure model was nested into the fate, exposure, and effects model USEtox (21), in order to quantify indoor and outdoor exposure on the same methodological basis. The indoor air and USEtox models communicate via air exchange (Figure 1b). The combined USEtox model with an indoor compartment embedded will be made publically available in 2009.

Intake fractions for indoor exposure were compared to those for outdoor exposure. Value ranges of parameters needed to run the models and calculate intake fractions were retrieved from a literature review and personal communications with authorities and building insurance offices (see the Supporting Information). Intake fractions from outdoor exposure were calculated with the USEtox model (20, 21). Intake fractions from both indoor and outdoor exposure are part of the characterization factor (CF), within the same impact category "human toxic effects", following the USEtox methodology (20, 21) (eq 2)

$$CF = iF \cdot EF$$
 (2)

Where iF is the intake fraction $[kg_{intake}/kg_{emitted}]$ and EF the effect factor $[cases/kg_{intake}]$. In the Life Cycle Impact Assessment phase, the characterization factors are multiplied to the emissions reported in the inventory phase to determine an overall impact score for potential human-toxic effects.

Results

Model Screening Assessment. As shown in Table 1, five of the existing indoor models (one-box model, one-box model with mixing factor, two-zone model, multibox model, and eddy-diffusion model) were considered compatible to the general principle of environmental exposure models, used for the assessment of human health effects in LCA. Therefore, these models could be connected to the environmental exposure models, in order to assess human-health effects from indoor as well as outdoor exposure, using the same methodological basis.

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TABLE

compatibility to environmental models used in LCA	worst-case assumption not adequate within LCA	compatible, as shown in ref <i>9</i>	compatible
ease of use	easy to use	easy to use	expert knowledge necessary to choose mixing factor; spatial distribution of workers needs to be known
data requirements for concentration quantification. further parameters needed to quantify intake fractions (eq 1a)	low: saturation pressure	ventilation rate; emission rate	ventilation rate; emission rate; mixing factors for the position of people exposed (spatial variation of concentration → expert knowledge)
transparency	Bulk-Mixing Models high transparency ig	transparent	medium
accuracy and precision	ixir lere	adequate for multiple sources and for good mixing conditions; not appropriate for near-field exposure to single sources, in particular in large rooms with bad mixing conditions	corrects for bad mixing conditions/ ventilation efficiencies; mixing factor is a predicted value that is not constant throughout the room
selection of references for model description	17, 22	17, 22°	24–26°
short description of model principle and basic equations ^b	calculates the worst-case concentration that would occur if there are no ventilation, no sinks and all of the mass of the chemical being considered enters the air instantaneously. $C = P_{Vap}/P_{atm} \times 10^{6}(ppm)$	concept of mass conservation and of concentration homogeneity throughout a single indoor volume; concentration at steady state is a function of emission and ventilation rate (adsorption also considered in cases (23); C = 200 (corrects for incomplete mixing with an empirical mixing factor $C = G/(mQ)$ (mg/m³)
model*	zero-ventilation model	one-box model	one-box model with mixing factor

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compatibility to environmental models used in LCA	compatible, as shown in ref <i>9</i>	compatible, as shown in refs 7, 8	compatible, but level of detail higher than for ambient fate and exposure models
ease of use	definition of various arbitrary parameter values necessary; spatial distribution of workers needs to be known	easy to use	steady-state version is easy to use, but value of D difficult to obtain; spatial distribution of workers needs to be known.
data requirements for concentration quantification. further parameters needed to quantify intake fractions (eq 1a)	ventilation rate; emission rate; size, geometry, and air exchange of inner zone; distance to source of people exposed	ventilation rate; emission rate; airflow between rooms; time fraction occupants spend inside rooms	eddy diffusitivity (empirical value); emission rate; distance to source
transparency	medium (inner zone is conceptual)	medium	Diffusion Models medium
accuracy and precision	good for near-field exposure assessment; assumption of ideal mixing within each box sometimes not accurate	adequate for multiple sources and for good mixing conditions; not appropriate for near-field exposure to single sources, in particular in large rooms with bad mixing conditions	Disable to describe concentrations as a function of space; not for unidirectional air draft
selection of references for model description	11, 27–31°	7, 8°	32–34°
short description of model principle and basic equations ^b	accounts for the higher intensity of exposure near the source by using two conceptual well-mixed compartments (near and far field); at steady state: $C_{NF} = G/Q + G/\beta C_{FF} = G/Q + G/\beta C_{FF} = G/Q + G/Q + G/B C_{FF} = $	accounts for transport to and exposure in multiple rooms; at steady-state with emission in room 1: $C_1 = (G + \beta_{21}C)/(\beta_{12} + Q_1) C_2 = \beta_{12}C/(\beta_{21} + Q_2)$ ($(\beta_{21} + Q_2)$ ($(\beta_{21} + Q_2)$ ($(\beta_{21} + Q_2)$	mass transport is driven by turbulent (or "eddy") diffusion, which is expected to dominate molecular diffusion; at steady state, concentrations are modeled as a function of distance from the emission source: $C_i = G/4\pi Dr$ (mg/m³)
mode!"	two-zone model	multibox model	eddy-diffusion model

ease of use	complicated	only for experts; large computational power needed
data requirements for concentration quantification. further parameters needed to quantify intake fractions (eq 1a)	high data demands: eddy diffusitivity (empirical value); emission rate; location of source and person exposed; direction and velocity of airflow	many parameters, such as physical, fluid dynamic, and heat transfer
transparency	wol	Numerical Analysis Models e and low lved
accuracy and precision	able to describe concentrations as a function of space, considers directional airflows	Numeric very accurate and highly resolved results
selection of references for model description	33, 35, 36	37
short description of model principle and basic equations ^b	Diffusion model that takes into account the direction of air currents	nonlinear set of equations for the conservation of mass, energy
TABLE 1. Continued model"	Gaussian plume dispersion model	computational fluid dynamics model (CDF)

compatibility to environmental models used in LCA

needs that are not available within a regular LCA study

many information

 a All models not marked in bold were considered to be compatible with conventional exposure models for the environment used in LCA (see last column for explanation). b Variables: R_{vap} is the saturation pressure [Pa], P_{atm} the ambient atmospheric pressure [Pa], C the indoor concentration [ppm or mg/m³], Q the ventilation flow [m³/s], Q the emission rate [mg/s], Q the mixing factor, $C_{\text{NF,FF}}$ the uniform concentration of the near and far field [mg/m³], Q the airflow rate between near and far field or between different rooms [m³/s], Q the eddy diffusivity [m²/s], and Q the distance from the emission source [m]. Q Further references for model applications and comparisons to measurements are provided in Tables S1 and S2 in the Supporting Information.

within regular LCA studies

variables

and momentum (Navier-Stokes equations)

input information not available

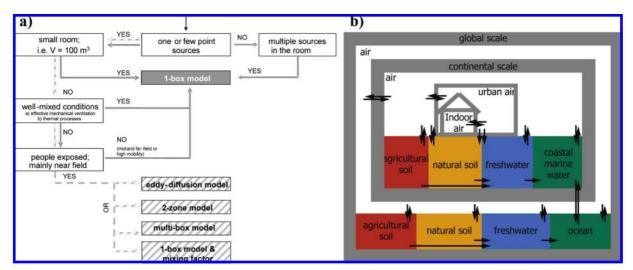


FIGURE 1. (a) Decision tree for indoor-model choice in LCA studies. All dashed lines lead to the models that allow for the assessment of near-field and far-field exposure (for model characteristics see Table 1); (b) Nesting the indoor model into the environmental fate and exposure model USEtox (adapted from ref 21).

Bulk-mixing models are particularly easy to integrate in the current LCA framework, because the models are conceptually very similar to the existing environmental models. The applicability of such models within LCA has already been demonstrated (7–9). The eddy-diffusion model represents another possible approach. Although not yet applied within the context of LCA, the steady-state version of this model could easily be integrated with environmental multimedia models. The eddy-diffusion model has the advantage that it can model spatial concentration profiles in indoor settings more accurately than bulk-mixing models. However, within the context of LCA it is open to discussion whether such level of detail is necessary or even possible. For instance, even if the information on the eddy-diffusitivity parameter is available (which is often not the case), the distance of all humans exposed to the emission source must be known to calculate intake fractions. This information is both highly variable and only rarely available in LCA studies.

Three models were ruled out for use in LCA. The zero-ventilation model assumes complete saturation without considering any pollutant sinks, such as ventilation. Therefore, this model may be applicable for assessing worst-case scenarios, for instance within a risk assessment, but not within the framework of LCA. On the other hand, the Gaussian plume and computational fluid dynamics models require too much input information, e.g. the specific airflows within the room (Table 1), which is not available within standard LCA studies. Thus, these models were not further considered.

The one-box model can be used as default model, as it matches the model principles and the level of detail of exposure models for the environment (Figure 1b). Thus, the focus of the current paper will be put on the one-box model. However, in some situations, a higher level of detail is necessary, and a more sophisticated model may be needed to refine the analysis and model spatial concentration patterns. In order to outline in which exposure situation which model is most appropriate, a decision tree was set up by the expert team (Figure 1a). This tree helps to choose an adequate model for a specific exposure situation. For instance, if there are multiple sources in the room or if the ventilation conditions are very good, the one-box model is often a valid choice. On the contrary, if the room volume is big and if the ventilation conditions are poor, other models that allow for the assessment of near-field exposure are more appropriate. This decision tree provides a rough guideline, with mostly qualitative criteria.

Model Parameter Values. The models shown in Figure 1 require information on parameter values with varying

extends (see eq 1a and equations in Table 1). Table 2 displays the results of a literature review concerning value ranges for a selection of exposure parameters.

Comparison of Indoor and Outdoor Intake Fractions. Figure 2 shows a comparison of outdoor and indoor intake fractions for households and for several industrial settings and chemicals. The examples shown focus on substances that are primarily emitted from indoor sources. The intake fraction is independent from the amount emitted (as opposed to, for example, concentration or dose) and expresses the marginal increase in exposure due to an increase in emission. It can be seen that the amount taken in by humans per kilogram of emission is several orders of magnitude higher for indoor emissions at workplaces and in residential settings than for outdoor emissions. Therefore, if, for example, a substance is emitted in an indoor setting and is eventually transferred to outdoor air by ventilation (neglecting for degradation), the major part of the impact is likely to occur in the indoor setting.

Discussion

Life Cycle Inventory Analysis. The assessment of indoor exposure needs to be facilitated by including emission factors, intake fractions and human-toxicological effect factors for indoor air sources into existing LCA software tools and databases. The present study helps by providing the methodological framework to estimate intake fractions in indoor settings in a structured, transparent and consistent manner. However, indoor emission data also need to be provided in inventory databases, similar to those available for outdoor emissions. This could be achieved by including an indoor air compartment, in addition to the existing air, water, and soil compartments in life cycle inventory databases, e.g., Ecoinvent (51). Further work is planned to establish a readyto-use list of relevant emission factors that can be incorporated in the life cycle inventory analysis.

Model Choice and Parameter Values. In LCA, information about specific exposures will not always be available. This will often restrict the choice of the model to the one-box model. The one-box model seems to be a good default choice, as the level of detail matches that of environmental models used in LCIA. More sophisticated models with indoor spatial differentiation may be used as well, if specific information, for instance on the spatial distribution of sources and people in the room, is available (Table 1). However, in this case, the level of detail would deviate from the environmental fate

TABLE 2. Model Parameters and Empirical Value Ranges

parameter	value ranges	references
inhalation rate of humans ^a , <i>IR</i>	0.44–1.04 m³/h (average 0.5 m³/h at rest) for households	6
numans , m	0.375-4.75 m ³ /h for occupational exposure ^b (average: 2.5 m ³ /h for a male worker)	38-42
	0.55 m ³ /h for environmental exposure (breathing rates of adults at rest); 0.62 m ³ /h for 10-year-old children	40, 43
air exchange per hour, <i>k</i>	U.S. residential buildings: geometric mean: 0.5 exchanges/h (Stdev = 2.1)	44
po	Dutch recent single-family dwellings (living room): 0.9 exchanges/h (Stdev = 0.7)	45
	occupational setting without mechanical system: 1 exchange/h or less	4, 46
	occupational setting with a mechanical system: 3-20 exchange/h.	47–49
ventilation flow, Q/N	households: U.S. household: GM = 80 m³/h per person, GSD = 2.7; arithmetic mean: 130 m³/h per person	4, 6
	average = 85.9 m ³ /h, standard deviation = 45.9 m ³ /h	45
building volume and number of people exposed	households: U.S. residences: GM = 160 m ³ /person; GSD = 1.9	6
	households Europe: median household size: 75–99 m²/household (~225–297 m³) for Austria, Denmark, Finland, France, Germany, and Greece; 300–447 m³/household for Norway; 150–222 m³/household for Albania, Croatia, Czech Republic, Estonia, Hungary, Iceland, Italy, Poland, Slovenia, and Turkey; Average size of household: 2–3 persons/household for all these countries but Albania (4.2 persons) and Turkey (4.4 persons) industry: up to several 1000 m³/personc	Figure S1
mixing factor, m	0.1-1	in the Supporting Information 49
air exchange rates between zones, β	3–30 m³/min	11
diffusitivity, D	$0.05-11.5~\mathrm{m^2/min}$ (0.1 $-$ 0.6 includes 70% of observed values	22

^a To avoid double counting, the consideration of exposure time in all compartments is necessary when assessing exposure in indoor and outdoor settings. ^b Depending on physical activity and human characteristics (e.g., sex). ^c The number of people exposed, *N*, and the building volume vary throughout and within industrial sectors (see the Supporting Information).

and exposure models commonly used, as the latter do not consider inhomogeneous mixing within the environmental media.

Setting up a list of recommended values for exposure parameters may be, in general, difficult. In this paper, we tried to provide ranges of parameters for the models that were considered suitable for use in LCA (Tables 1 and 2). However, these ranges are very broad. Especially with regard to the more abstract, but very sensitive parameters, such as the air exchange rates between the conceptual inner and outer boxes in a two-zone model or the eddy-diffusion constant, it is often difficult to find representative values. Contaminant dispersion phenomena within the rooms can be influenced by complex interactions between variables such as the room geometry, the direction of the principal air flows, and the presence and movements of occupants (52-57). The models identified suitable for use in LCA (Table 1) do not take into account such detailed information. Further, the intake fraction in indoor environments was assumed to not depend on the chemical. This is a valid assumption if removal by ventilation is large in comparison to adsorption to surfaces and degradation (which is often the case in household and occupational settings). However, intake fractions may be reduced significantly by sorption to indoor surfaces (58), especially for strongly sorbing substances in furnished residential homes.

For generic LCAs, a good approach is to calculate intake fractions for several generic workplace and household environments, which are characterized by air exchange rates, volumes, and numbers of people exposed. The parameter values for the indoor models (i.e., room volumes, air exchange rates, etc.) may vary geographically, e.g., because of climate conditions, cultural aspects, or different ventilation practices. For instance, the number of people per cubic meter working in the chemical industry in countries with cheap labor costs, such as China or India, is probably much higher than in industrial countries with a high degree of automation. Therefore, an important requirement for the final implementation of the model within the USEtox model is that the user can adapt the parameter values to his or her specific circumstances and that default parameter lists for various workplace settings and geographical regions are provided to facilitate the application.

The results assessed with the generic characterization factors based on the one-box model calculations will give only an indication of whether indoor exposure may be important. In such cases, it is advisable to refine the model parameter values or even change the model according to Figure 1a. The implementation of various indoor model options into the USEtox model, among which the user can choose, will make this recommendation feasible as well for LCA practitioners with limited time availability.

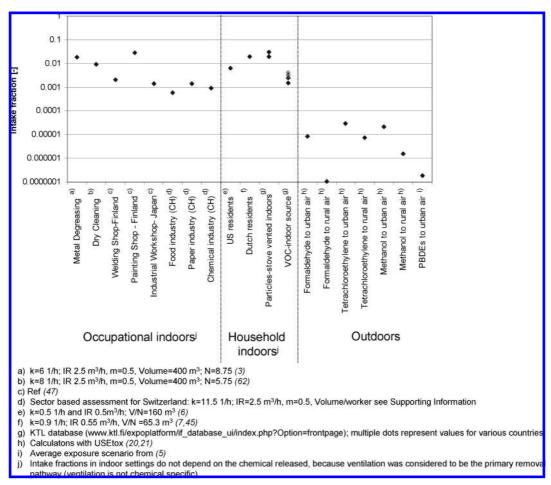


FIGURE 2. Examples for intake fractions found in indoor industrial and residential settings and in the ambient environment. In contrast to outdoor intake fractions, intake fractions related to indoor environments do not depend on the chemical, as ventilation was assumed to be the primary removal pathway, neglecting substance-specific degradation and adsorption (see Discussion).

The use of a model can be circumvented in the case that monitored concentration values and production volumes are available. Instead of multiplying the emissions to the intake fractions, as usually done in LCA, the amounts taken in by the people exposed would be directly calculated from the monitored concentrations and the number and inhalation rates of people. This approach requires that pollutant concentrations can be directly linked to the functional unit (source appointment), which is possible in some indoor settings.

In a later stage, indoor exposure to radioactive gases such as radon can also be incorporated within the impact category "radiation" in LCIA methods such as Eco-Indicator 99, similarly to the framework shown in this paper. This is especially important for household settings, where radon can be an important factor for the total health damage as a result of indoor exposure.

Routine Assessment of Indoor Exposure within Life Cycle Assessment. The framework suggested in this paper is the first in putting forward a general procedure for indoor exposure assessment within LCA. From a practical point of view this is relevant, as the model results suggest that intake fractions from indoor emissions are often larger than intake fractions from outdoor emissions. This finding is confirmed by previous studies (6, 59) showing that indoor chemical concentrations often surpass outdoor concentrations by many orders of magnitude. This stresses the need to consider indoor exposure in LCA. It could even lead to human toxicity becoming a dominant impact category for certain products such as paints, furniture, or

carpets. A routine assessment of indoor exposure in LCA will be facilitated by including the indoor model in the USEtox model (20, 21). Similar developments can be anticipated for the field of risk assessment, as European REACH legislation also calls for exposure scenarios, including worker or consumer exposure, for example. Such integrated assessment will point to the most important exposure pathways and improvement potentials, considering the whole life cycle of chemicals (60). Moreover, the past has witnessed several cases in which chemicals were banned for one reason, such as ecological impacts, but got substituted by chemicals with other problems, i.e., occupational health effects (e.g., the market introduction of n-hexane/acetone based brake cleaning products due to air quality rules in California in 1990 (61)). Such tradeoffs between the various possible effects of chemicals can be revealed when applying integrated models for indoor and outdoor exposure and, ultimately, such problem shifting may be avoided.

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Supporting Information Available

Literature survey comparing indoor model results to actual measurements and on the gathering of parameter values for three industrial sectors (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.

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