

Integrating Human Indoor Air Pollutant Exposure within Life Cycle Impact Assessment

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ABSTRACT

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.

KEYWORDS

Indoor exposure; exposure models; Life Cycle Assessment; LCA; intake fraction

BRIEFS

Indoor air exposure can and should be assessed within Life Cycle Assessment, as indoor human intake fractions are often higher than outdoor intake fractions.

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 $9 \mu\text{g}/\text{m}^3$ (1) and $24.6 \mu\text{g}/\text{m}^3$ (2), respectively, while 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 hours 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. In order 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 towards the enhancement of the current LCA framework.

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. In order to achieve this objective, (a) existing indoor exposure models for households and occupational settings are reviewed and evaluated concerning their use in

LCA, (b) these models are used to provide a methodological framework for the routine assessment of indoor exposure to chemicals within LCA studies, (c) a sample set of model parameter values is gathered and the application of the methodological framework illustrated, and (d) examples of human intake fractions of indoor and outdoor exposure for a number of chemicals are compared. 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 semi-quantitative models (11-15) to physical-process 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 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 acknowledgement as members).

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

$$iF = \frac{\partial Intake}{\partial Emission} = \frac{\partial I_x}{\partial G_x} \cdot N = \frac{\partial C_x \cdot IR}{\partial G_x} \cdot N = \frac{\partial (G_x/Q) \cdot IR}{\partial G_x} \cdot N = \frac{IR}{Q} \cdot N = \frac{IR}{V \cdot k_{ex}} \cdot N \quad (1a)$$

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

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 (m^3 /day), N is the number of people exposed (-), C_x is the chemical concentration in air (kg/m^3), G_x is the emission rate of chemical x (kg/day), Q is the ventilation rate in the exposure area (m^3 /day), 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 (-).

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. In order to provide an agreed, consistent and stable toxicity assessment method for comparative environmental assessments, the USEtox model (22, 23) 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 (22, 23). 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 which 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 (23), 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.

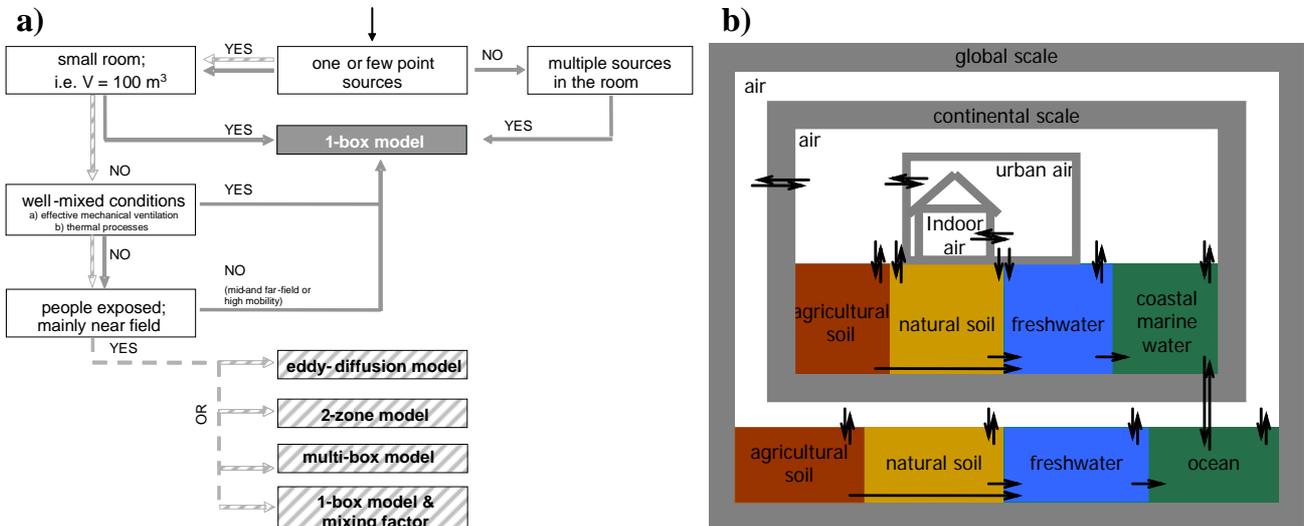


Figure 1: a) Decision tree for indoor-model choice in LCA studies. All dashed lines lead to the models that allow for the assessment of near-field and far-field exposure (for model characteristics see Table 1); b) Nesting the indoor model into the environmental fate and exposure model USEtox (adapted from (23)).

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 Supporting information). Intake fractions from outdoor exposure were calculated with the USEtox model (22, 23). 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 (22, 23) (equation 2):

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

Where iF is the intake fraction [$\text{kg}_{\text{intake}}/\text{kg}_{\text{emitted}}$] and EF the effect factor [$\text{cases}/\text{kg}_{\text{intake}}$]. 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, multi-box 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.

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-diffusivity 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.

Table 1: Screening assessment of indoor models by the UNEP/SETAC working group on the *Integration of Indoor Exposure Assessment within LCA*. All models not marked in grey were considered to be compatible with conventional exposure models for the environment used in LCA (see last column for explanation).

Model	Short description of model principle and basic equations ^a	Selection of references for model description	Accuracy and precision	Transparency	Data requirements for concentration quantification. Further parameters needed to quantify intake fractions (equ. 1)	Ease of use	Compatibility to environmental models used in LCA
Bulk-mixing models							
Zero-ventilation model	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. $C = \frac{P_{vap}}{P_{atm}} \cdot 10^6 [ppm]$	(17, 24)	Not accurate because: - ventilation neglected - complete saturation assumed - complete mixing assumed - no sinks considered No temporal evolution of concentrations considered. Worst-case estimate.	High transparency	Low: - saturation pressure	Easy to use	Worst-case assumption not adequate within LCA.
One-box model	Relies on the 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 (25)): $C = \frac{G}{Q} \left[\frac{mg}{m^3} \right]$	(17, 24) ^b	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.	Transparent	- ventilation rate - emission rate	Easy to use	Compatible, as shown in (9).
One-box model with mixing factor	Corrects for incomplete mixing with an empirical mixing factor.	(26-28) ^b	Corrects for bad mixing conditions/ventilation efficiencies. Mixing factor is a	Medium	- ventilation rate - emission rate - mixing factors for the position of	Expert knowledge necessary to choose mixing factor.	Compatible.

	$C = \frac{G}{m \cdot Q} \left[\frac{mg}{m^3} \right]$		predicted value that is not constant throughout the room.		people exposed (spatial variation of concentration → expert knowledge)	Spatial distribution of workers needs to be known.	
Two-zone model	Accounts for the higher intensity of exposure near the source by using two conceptual well-mixed compartments (near and far field). At steady state: $C_{NF} = \frac{G}{Q} + \frac{G}{\beta} \left[\frac{mg}{m^3} \right]$ $C_{FF} = \frac{G}{Q}$	(11, 29-33) ^b	Good for near-field exposure assessment. Assumption of ideal mixing within each box sometimes not accurate.	Medium (inner zone is conceptual)	- ventilation rate - emission rate - size, geometry and air exchange of inner zone - distance to source of people exposed	Definition of various arbitrary parameter values necessary. Spatial distribution of workers needs to be known.	Compatible, as shown in (9),
Multi-box model	Accounts for transport to and exposure in multiple rooms. At steady-state with emission in room 1: $C_1 = \frac{G + \beta_{21}C_2}{\beta_{12} + Q_1}$ $C_2 = \frac{\beta_{12}C_1}{\beta_{21} + Q_2}$ $\left[\frac{mg}{m^3} \right]$	(7, 8) ^b	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.	Medium	- ventilation rate - emission rate - airflow between rooms - time fraction occupants spend inside rooms	Easy to use	Compatible, as shown in (7, 8).
Diffusion models							
Eddy-diffusion model	Assumes that 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: $C_r = \frac{G}{4\pi Dr} \left[\frac{mg}{m^3} \right]$	(34-36) ^b	Able to describe concentrations as a function of space. Not for unidirectional air draft.	Medium	- eddy diffusivity (empirical value) - emission rate - distance to source	Steady-state version is easy to use, but value of D difficult to obtain. Spatial distribution of workers needs to be known.	Compatible, but level of detail higher than for ambient fate and exposure models.
Gaussian plume dispersion model	Diffusion model that takes into account the direction of air currents.	(35, 37, 38)	Able to describe concentrations as a function of space, considers directional airflows.	Low	High data demands: eddy diffusivity (empirical value) - emission rate - location of source and person exposed - direction and velocity of airflow	Complicated	Many information needs that are not available within a regular LCA study
Numerical analysis models							
Computational fluid dynamics model (CDF)	Non-linear set of equations for the conservation of mass, energy and momentum (Navier-Stokes equations).	(39)	Very accurate and highly resolved results.	Low	Many parameters, such as physical, fluid dynamic and heat transfer variables	Only for experts; large computational power needed	Input information not available within regular LCA studies

^a Variables: P_{vap} is the saturation pressure [Pa], P_{atm} the ambient atmospheric pressure [Pa], C is the indoor concentration [ppm or mg/m^3], Q the ventilation flow [m^3/s], G the emission rate [mg/s], m the mixing factor [-], $C_{NF,FF}$ is the uniform concentration of the near and far field [mg/m^3], β airflow rate between near and far field or between different rooms [m^3/s], D the eddy diffusivity [m^2/s], and r the distance from the emission source [m].

^b Further references for model applications and comparisons to measurements are provided in Tables S1 and S2.

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

Table 2: Model parameters and empirical value ranges.

Parameter	Value ranges	References
Inhalation rate of humans ^a , IR	0.44 – 1.04 m ³ /h (average 0.5 m ³ /h at rest) for households 0.375 – 4.75 m ³ /h for occupational exposure ^b (average: 2.5 m ³ /h for a male worker) 0.55 m ³ /h for environmental exposure (breathing rates of adults at rest); 0.62 m ³ /h for 10 year-old children	(6) (40-44) (42, 45)
Air exchange per hour, k	US residential buildings: geometric mean: 0.5 exchanges/hour (Stdev = 2.1) Dutch recent single-family dwellings (living room): 0.9 exchanges/hour (Stdev = 0.7) Occupational setting: Without mechanical system: 1 exchange/hour or less With a mechanical system: 3 – 20 exchange/hour.	(46) (47) (4, 48) (49-51)
Ventilation flow, Q/N	Households: US household: GM = 80 m ³ /h per person,	(4, 6)

inventory databases, e.g. Ecoinvent (55). Further work is planned to establish a ready-to-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 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 (56-61). 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 (20,62), 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. due to climate conditions, cultural aspects or different

ventilation practices. For instance, the number of people per m³ 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 only give 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, also for LCA practitioners with limited time availability.

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, 63) which show 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 (22, 23). 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 (64). 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, for instance 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 (65)). Such trade-offs 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 PARAGRAPH

Supporting information on a literature survey comparing indoor model results to actual measurements and on the gathering of parameter values for three industrial sectors.

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