**A Modeling Enterprise for Chemistry of Indoor Environments (CIE)**

**Why do we need a CIE modeling enterprise?**

On November 7-8, 2016, we convened a workshop in Washington, D.C. to identify how best to define a modeling enterprise for studying the chemistry of indoor environments. The workshop included presentations and discussions organized around four themes: (1) Framework for an indoor air chemistry modeling enterprise, (2) From outdoor to indoor chemistry models, (3) Intersection of indoor chemistry models with building data and models, and (4) Outreach and community building. This editorial reports the major findings and recommendations from the workshop, as distilled by the three workshop organizers. To reflect the workshop conversations, the term “indoor chemistry” is used in this editorial in a broad sense: chemical phenomena that influence the composition of indoor air including transport, partitioning, phase change, and chemical transformations.

The roughly seven billion people in the world today spend most of their time exposed to air inside buildings. This air, although originating from outdoors, is altered by means of physical and chemical processes specific to each indoor microenvironment. For many pollutants, the differences between indoor and outdoor air are large due to indoor sources; for some pollutants, chemical transformations play a dominant role. The extent to which indoor chemistry influences human health is unknown, but at the very least, epidemiological observations of adverse effects of outdoor air pollution are suspected to be influenced by indoor exposure to primary and secondary pollutants resulting from these processes1,2.

One of the challenges with understanding the chemistry of indoor environments is the scale and complexity of the challenge. As outlined in a recent editorial3, in the US alone, there are around 320 million people4, 131 million residential units5, 5.6 million commercial buildings6, and around 100,000 chemicals in the atmosphere7. Consequently, there are potentially 1021 unique combinations of people, places, and pollutants at any given time! This number increases when we consider that each building or residential unit comprises multiple room-scale or smaller microenvironments. Aiming to reveal the true breadth of indoor chemistry and personal exposure through direct extrapolations of measurements from a handful of buildings is clearly unrealistic.

Given that fewer measurements of air pollutants and related parameters exist indoors compared to outdoors and the heterogeneity of indoor chemistry microenvironments, models must be used to advance understanding. Such models must be appropriately framed to the scientific questions being posed, and designed to address appropriate temporal and spatial scales and populated with scientifically rigorous mechanisms, so that they can be applied to predict indoor conditions across the vast and varied landscape of building environments.

**Modeling domains and connections**

The composition of indoor air depends strongly on indoor sources (e.g. cooking, cleaning, material emissions), infiltration of outdoor pollution, chemistry, and transport among indoor zones. Figure 1 shows the physical interconnections and major zones in buildings that should be included in robust indoor chemistry models. The building shell is surrounded by a peribuilding environment that is influenced by the ambient atmosphere and by factors specific to each building and its surroundings. This air penetrates the building shell, intentionally or unintentionally, and passes among occupied rooms, interstitial spaces, ducts and HVAC systems. The zone very near the occupants, the perihuman environment, can have attributes that differ from the other indoor zones. Within each zone, gases and particulate matter are influenced by processes that include emissions, deposition, diffusion, adsorption, oxidation, hydrolysis, and so forth. The “time” axis emphasizes the dynamic nature of the constituents of each zone, flow between zones, and processes taking place.

The conceptual model shown in Figure 1 indicates how indoor models have typically been developed and are likely to be developed in the future: zones that are reasonably homogenous are modeled separately and connected to other zones via flow of energy, air mass, and species. More complex models that account for spatial variation within zones (e.g. those that include computational fluid dynamics) may also be necessary. Some modeling domains, such as building physics models, may be considered in isolation or in tandem since they are not strongly influenced by processes such as homogeneous chemistry. Other systems are strongly interconnected, such as surface phenomena, homogeneous chemistry, and indoor aerosol chemistry.



**Figure 1**. Conceptual model of zones and interconnections that influence or are influenced by indoor chemistry. Within each zone, dynamic processes occur including chemical transformations, emissions, deposition, condensation or evaporation, and transport of mass and energy.

**Research needs**

We group research needs into three major categories: *Processes*, *Model Evaluation*, and *Building Data and Simulation*. Within the *Processes* category, we further divide into gas-phase, aerosol-phase, and interfacial mechanistic processes.

**Processes: Gas-phase mechanisms**

In recent decades, models have revealed major insights into the mechanisms and importance of indoor chemistry. Nazaroff and Cass8 used an abbreviated chemical scheme to suggest that the same type of reactive chemistry as occurs in photochemical smog could influence the composition of indoor air in some circumstances. Weschler and coworkers used a simple mass balance model to investigate potential nitrate and hydroxyl (OH, a key outdoor oxidant) radical formation9,10. Consequently, models of increasing chemical complexity have been developed to investigate OH and hydroperoxy (HO2) concentrations, HONO chemistry, and secondary organic aerosol (SOA) composition, amongst other things11–15.

Oxidation is a key process influencing indoor air gas-phase chemistry and indoor measurements of ozone, NOX, and VOC have been informative. Looking forward, combined modeling and measurements of OH and HO2 concentrations in environments, beyond the few existing studies in classrooms16, would be beneficial, as would studies of nitrate and chlorine radicals and speciated organic peroxy radicals (RO2). For instance, though it has been suggested that the nitrate radical may be important indoors17, some models predict that the indoor concentration is unlikely to exceed instrumental detection limits under typical conditions13,18, so measurements of NO3 could reconcile these differing expectations. Measurements of RO2 concentration and composition would provide information indicative of the dominant reactive VOCs indoors. Chlorine chemistry is also relatively understudied indoors, given its sources indoors through use of chlorinated water supplies and cleaning activities. Measurements of stable secondary pollutants from oxidation (e.g. carbonyls, organic nitrates, peroxides, and carboxylic acids) would enable insight into indoor oxidation pathways, particularly if made simultaneously outdoors.

The detailed chemical mechanisms that form the backbone of indoor air chemistry models, such as the Master Chemical Mechanism20, were originally constructed for outdoor chemistry. As such, there are areas where the mechanisms can be developed to more effectively represent indoor processes. New terpene schemes would be beneficial, as would those of the species found in skin oil and emitted from the surfaces of many indoor materials. Also, these models make assumptions about photolysis rates indoors, yet we know little about the propagation of light indoors or the spectral frequencies of indoor light sources, as the few available measurements show wide variation8,21, 21. Similarly, the deposition rates assumed for oxidative gases and the consequent emission rates of secondary pollutants can impact indoor air chemistry to a significant degree, though such processes are currently highly uncertain and specific knowledge is largely limited to ozone.

**Processes: Aerosol-phase mechanisms**

Airborne particulate matter of differing composition is present indoors owing to outdoor-to-indoor transport, indoor primary emissions, and indoor formation of SOA following chemical reactions. Nazaroff and Cass22 first developed a model to predict size-resolved particle concentrations and fate indoors, considering ventilation, filtration, deposition onto surfaces, direct emission, and coagulation. Later models focused on chemistry and indoor SOA formation11,23–26.

Indoor aerosol modeling can benefit from advances in the formation and transformation of ambient aerosols. For example, model treatment of outdoor particles as having individual component-based thermodynamic partitioning and water uptake behavior should be valid for indoor models. Specifically, ammonium sulfate ((NH4)2SO4), soil, and black carbon particles will undergo mechanical losses without volatility changes, but ammonium nitrate (NH4NO3) and organic aerosol (OA) can also undergo further changes based on thermodynamic partitioning27. Measured OA can be subdivided into factors using positive matrix factorization (PMF), such as hydrocarbon-like OA and semi- and low-volatility oxygenated OA. Such factor-based partitioning can then be parameterized with the 1- or 2-D volatility basis set (VBS)28–30. Water uptake for different particulate matter composition can also be parameterized, for example using Köhler theory31. A recent field study32 demonstrated component-based partitioning as a function of outdoor-to-indoor temperature differences using measurements with an online, real-time aerosol mass spectrometer (AMS).

Thermodynamic and water uptake models typically assume equilibrium, and the validity of that assumption relative to indoor residence time scales must be explored. For instance, one location in buildings with very short residence times, large temperature swings, and sudden changes in particle concentrations is the air handling system, commonly fitted with both heating or cooling coils and particle filters; equilibrium may not be attained here. Future models for predicting particle size distribution changes due to mechanical losses need to be combined with partitioning and uptake models for size-resolved aerosol predictions.

Indoor aerosol models must account for the large variety of possible indoor emissions. For fine-mode particles, dominant primary emission sources are combustion related, with cooking having the greatest potential to increase indoor aerosol fine mass and number35–37, when indoor smoking is absent. Also, a sometimes important source of indoor aerosol is SOA generated from chemical reactions. The oxidation of VOC by ozone, OH, or NO3 results in products of lower volatility that partition into the condensed phase and form SOA39. SOA formation can be predicted with approaches that use detailed partitioning modeling or empirical aerosol mass fractions (AMF) within the VBS framework. Certain terpenoids in cleaning products, such as α-terpinene and terpinolene, have not had their formation potential well characterized. Questions also remain as to the total ozone reactivity in buildings (i.e., whether enough indoor VOCs are accounted for), which would impact SOA formation.

**Processes: Interfacial mechanisms**

Indoor materials and their “surfaces” are among the least understood domains for making progress towards the goal of developing comprehensive indoor chemistry models. The available surface area in a building, relative to the volume of air in contact with those surfaces, is much greater than outdoors. Lower volatility molecules sorb to surfaces and, in turn, persist in buildings with more time to react with other molecules. Indoor materials provide time, opportunity, and a distinctive microenvironment that can substantially alter the composition of indoor air.

Many types of reactions may be occurring at indoor surfaces, but reactions of ozone at indoor surfaces are well known (if not mostly understood) and clearly impact exposure. For example, population exposure to ozone would be approximately 5 to 7 times greater if not for its reactive loss on indoor surfaces1,43. Simultaneously, this chemistry generates a host of oxidized organics and thereby increases inhalation intake of aldehydes, ketones, carboxylic acids44, and ultrafine SOA45,46.

Models that predict molecular transport (uptake and emissions) exist for building surfaces like painted drywall, vinyl flooring, concrete, and carpet. Relatively simple models have adequately matched experimental results probing primary emissions47, sorption/ desorption48–50 and ozone deposition51,52. Building upon emission models, advanced models must necessarily better account for porosity, internal kinetic limitations to transport, interfacial phenomena including sorption and heterogeneous chemistry, the presence of nonuniform surface properties, and so forth. We need to better model transport, chemistry at and within the skin, in clothing, bedding and upholstery, in layers of dust, and on hot and cold surfaces53. Currently, yields and formation rates of some oxidation reaction products at surfaces are reported, but are not yet predictable. We need models that account for the time history of surfaces that result in phenomena such as ozone aging54,55 or result in coatings that oligomerize and become more impermeable over time56. Identifying the “most important” surfaces to study is necessary to populate models most effectively.

**Model evaluation**

One advantage toward understanding chemistry in buildings rather than in the outdoor environment is that buildings are more controllable. Yet, the vast heterogeneity within and among buildings makes evaluating models by making measurements in real buildings a great challenge. The level of detail required to parameterize, for example, a detailed mechanistic gas chemistry model, will be large and may require information from building areas that are impossible to access. Instead, comprehensive models might be best compared against measurements from laboratory chambers or well-defined test buildings. Similarly, a detailed model of transport and chemistry taking place on and inside materials should be tested with model systems representative of the vast number of materials present in buildings. The process is necessarily iterative: models help design experimental systems, experiments challenge and improve models, models help define the temporal resolution of measurements, large scale systems inform small scale systems, and so forth.

Whether at laboratory or full scale, these physical representations should be sufficiently characterized to help parameterize the model and also test the model by perturbing the system. The composition and other characteristics of materials can be defined, rather than imposed on the researchers as they are in a real, occupied building. An advantage of highly controlled and instrumented systems is the ability to carefully perturb the system to probe the impact of individual parameters on the resulting chemistry.

In addition to championing the use of controlled test facilities to evaluate models, we also believe that field experiments in real buildings are necessary, since in controlled experiments there are limited opportunities for discovery of new phenomena or species. For example, real and simulated human activities may have vastly different impacts on indoor environments as revealed by detailed chemical speciation over time. Field measurements also help complete the model framework by revealing new mechanisms, system inputs, and species. To make the best use of field results and improve models, it may be necessary to develop a list of “must measure” parameters. These could include common environmental parameters (e.g. light and temperature) or more challenging variables, like a specific radical species central to the chemistry of interest.

**Building data and simulation needs and opportunities**

Of course, modeling chemistry in indoor environments requires numerous building-oriented inputs. Variability exists for many parameters: air exchange rates (both with outdoor air and through recirculation pathways), building leakiness, mechanical system runtimes, filtration efficiencies, surface deposition rates, VOC sorption rates, particle and gaseous emission rates, and human presence and activity rates, among others. Misrepresenting certain variables due to parameter uncertainty or misplaced assumptions could have large impacts on results. Monte Carlo simulations, which represent inputs as probability distributions based on the variability and uncertainty of underlying knowledge, and which produce probability distributions of outcome values, are well suited to explore the sensitivity of developed models to unit changes in input parameters34,57.

Mixing time scales for air in built environments should be compared to the characteristic time scale (*τ*) of the process in question when setting model assumptions. For processes with longer characteristic times, such as monoterpene reactions with ozone (e.g. *τ* ~ 30 to 60 min), considering air as well mixed (e.g. *τ* ~ 5 min) results in appropriate bulk concentration approximations. Thus, these slower processes are sufficiently modeled by assuming a well-mixed room, mechanical system zone, or even an entire building. However, for certain processes, such as total VOC reactions with OH (e.g. *τ* ~ 0.1 s) produced by photolysis of HONO near a window, considering the room air as well mixed could result in substantial error. For faster processes, modeling must account for spatial variability, such as is done in computational fluid dynamics (CFD), which uses numerical methods to solve state equations for discrete room air volumes. Moreover, building transport time scales, which describe the transfer of air among zones due to mechanical flow or pressure differences between zones, must also be compared to process characteristic times when setting assumptions.

There is great opportunity to use existing models that describe certain aspects of building operation, either to enumerate the variability of building-related inputs for or through direct coupling with indoor chemistry models. As an example of parameter variability, the impact of light penetration through fenestration and its contribution to the dissociation of HONO may be understood using lighting models, such as Radiance (https://radiance-online.org/), a validated software tool that can predict spectral radiance and irradiance. Also, WUFI (https://wufi.de/en/) predicts transient coupled heat and moisture transport in exterior walls, so it can set water availability boundary conditions in chemical models. Building models that are good for coupling include whole building simulations such as EnergyPlus (https://energyplus.net/), which simulates building energy use and zonal mechanical system flows, or CONTAM (http://www.bfrl.nist.gov/IAQanalysis/index.htm), which simulates zonal mechanical and pressure driven flows.

**Community building in indoor chemistry modeling**

Unlike the ambient air chemistry community, there are no existing centers of excellence for indoor air chemistry, either at national or international levels. There is a clear need to improve connections among people motivated to model the chemistry of indoor environments. The new Alfred P. Sloan Foundation Program for Chemistry of Indoor Environments aims to do that, with the development of a new modeling enterprise being one of the main program aims. To that end, a MOdeling Consortium for Chemistry of Indoor Environments (MOCCIE) has recently been organized.

The aforementioned workshop on modeling indoor chemistry provided us with an enthusiastic network of people keen to participate in a new community for indoor air chemistry research. To build momentum and strengthen and enhance this community, we recommend applying networking tools both to maintain an online directory of skills and to provide a discussion area to facilitate brainstorming new projects. As a community, we should also plan for gatherings at large international meetings as well as additional meetings in between these events and, in particular, encourage early career investigators to become involved.

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