

Editorial

Indoor chemistry: research opportunities and challenges

Perspective

The mass of the earth's atmosphere is 5×10^{18} kg, which corresponds to more than 700 million kg for each of the current 7 billion human inhabitants. We lack a global census of built occupied space. Data for the United States indicate a total floor area in commercial (28%) and residential (72%) buildings of about 29 billion m² (USEIA, 2015a,b). The corresponding total mass of air in US buildings would be roughly 100 billion kg, corresponding to about 350 kg for each of the 320 million US residents. Hence, the indoor air mass per person in the US is about 0.5 millionths of the global atmosphere air mass per person. This comparison illuminates why atmospheric science research has strongly emphasized air outside of buildings.

However, when viewed through an anthropocentric lens, the indoor portion of the atmosphere gains prominence. Most of the air that humans encounter is indoor air. As is well known to readers of this journal, roughly 90% of the modern human time budget is spent indoors. Furthermore, most of that time is spent in one's own residence. Human emissions to the atmosphere also occur primarily indoors, and thus, humans have a disproportionate direct contribution to indoor air chemistry, through their metabolism, their skin and hair, their clothing, their use of personal care products, and their activities such as cooking.

Because buildings are leaky and deliberately ventilated, they continuously exchange air with their surroundings. To the extent that local outdoor air is polluted, ventilation will serve to introduce those pollutants into the indoor environment. Ventilation also serves to remove pollutants from indoor air, a process that is particularly important for those contaminants emitted directly from indoor sources.

The major impurities in indoor air are a combination of those introduced from outdoor air plus those emitted from indoor sources. However, even in the absence of indoor emissions, indoor concentrations may not be the same as in the local outdoor air. For example, in the case of ozone, reactions lead to an attenuation of indoor concentrations relative to those outdoors. On the other hand, source emissions cause indoor concentrations of many species to be substantially elevated above the corresponding outdoor levels. In addition, chemical transformations occur indoors, in the

gas-phase, on particles, and on indoor surfaces. The products formed from such reactions can materially influence the composition of indoor air.

Outdoors, much of the important reactive chemistry is driven by the hydroxyl radical. The primary source is photochemistry, and levels are substantially higher during the daytime than at night. Indoors, photon flux densities are much smaller than for daytime conditions outdoors. Consequently, indoor oxidative reactive chemistry is less dependent on the photochemically derived hydroxyl radical. Instead, ozone and possibly the nitrate radical play more important roles.

Another prominent feature of indoor environments is the large surface-to-volume ratio (S/V). Singer et al. (2007) reported nominal S/V values in the range 2.9–4.6 m² per m³ for a sample of ten residential rooms that they studied. The large quantity of exposed surfaces can influence indoor air chemistry through several mechanisms, including oxidative reactions, sorption, and acid–base chemistry.

Of order 10^4 – 10^5 different atmospheric organic species have been measured outdoors, and this number may be only a small fraction of those actually present (Goldstein and Galbally, 2007). Indoor air must contain a similarly broad spectrum of organic compounds. In addition to those entering from outdoors with ventilation, a variety of species are emitted from building materials, furnishings, cleaning products, the indoor microbiome, cooking, human metabolism, and personal care products among other sources. Among the broad classes of compounds found in indoor air are pesticides, plasticizers, and flame retardants. Exposure to elevated concentrations indoors can raise direct toxicity concerns for some organic species, such as formaldehyde. An additional emerging spectrum of concerns arises because of indoor exposures to endocrine disruptors, many of which are semivolatile organic compounds (SVOCs) (Rudel and Perovich, 2009). The physicochemical behavior of SVOCs indoors is challenging to understand and also likely to be of considerable significance for human health and well-being.

A grand challenge for studies of indoor chemistry is the huge number of distinct indoor environments. In the United States, for example, there are 114 million residential units and 5.6 million commercial buildings that serve a population of 320 million. Extrapolating to the global scale, one might expect in excess of one

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billion total indoor environments. Each may exhibit distinctive attributes. Very many chemical species are of potential interest. Concentrations of each species in each environment may vary with time to important extents. These circumstances strongly support a view that effective research on indoor chemistry should be oriented toward elucidating processes. We should seek to build a mechanistic understanding of the cause and effect relationships that facilitate the extrapolation from some finite set of specific investigations to the much larger pool of indoor environments to be characterized and understood.

The following paragraphs briefly describe frontiers of knowledge for four important themes in indoor chemistry. Significant research topics are highlighted, and key challenges are identified. Our aim is to be strongly illustrative rather than comprehensive.

Human occupants as agents influencing indoor chemistry

A common attribute of indoor environments is that most have human occupants. Perhaps surprisingly, relatively little effort has focused on the role of occupants influencing indoor chemistry. Weschler (2015) has reviewed progress in this area, most of which was achieved during the past decade. In addition to ozone-initiated chemistry on occupant surfaces, emissions of organic compounds from human occupants would certainly influence the chemical composition of air, especially in densely occupied spaces such as classrooms. Little work has been done with modern instruments to characterize the chemical emissions associated with human occupants of indoor spaces (Wang, 1975). It is clear that occupants' bioeffluents can adversely affect perceived air quality, but chemically specific understanding is lacking. Beyond the basic findings that organic chemical emissions can have material influence on indoor air composition and chemistry, a wide range of more detailed questions appear worthwhile to investigate. For example, to what extent are the emitted compounds associated with endogenous processes vs. originating from personal care products or from clothing? How variable are emissions among people? How variable are emissions from any individual as a function of time and metabolic activity? What important factors influence emissions? Do metabolic processes on the skin surface lead to the emission of secondary compounds, for example, following the use of personal care products? Modern analytical instruments based on techniques such as proton-transfer reaction mass spectrometry (Schripp et al., 2014) may be well suited for exploring many such issues.

Oxidative chemistry

The atmosphere is an oxidizing medium. Atmospheric chemistry is fueled by organic compounds, which enter

the atmosphere in a chemically reduced state. Over time, as the carbon is oxidized, the resulting products can have important environmental consequences. Among the significant intermediate products of atmospheric organic chemistry are aldehydes and organic acids that can contribute to adverse health and welfare consequences. Functionalization of organic compounds through oxidation leads to lower volatility products that can contribute to secondary organic aerosol. The formation of secondary organic aerosol has health significance, causes light scattering, and significantly influences other important atmospheric processes. In outdoor air, during day, direct photolysis (e.g., of aldehydes) and OH-induced oxidation of organic compounds drive much of the important chemistry. At night, the nitrate radical (NO_3) is an important catalyst.

Indoors, oxidative chemistry also can occur. Important oxidants indoors are three: ozone, the hydroxyl radical, and the nitrate radical (Gligorovski and Weschler, 2013; Waring and Wells, 2015). Although more remains to be learned, the role of ozone indoors is reasonably well studied (Weschler, 2000). On the other hand, remarkably little work has been done to assess the levels, causes, and consequences indoors of the hydroxyl radical (Gligorovski et al., 2014; Sarwar et al., 2002; Weschler and Shields, 1996, 1997; White et al., 2010). For the nitrate radical, even less is known, with only one published study reporting measured concentrations, in this case of the sum of NO_3 plus N_2O_5 (Nøjgaard, 2010).

Nitrate may be of particular importance in some indoor environments. The nitrate radical is formed by ozone reacting with nitrogen dioxide. Both species may be simultaneously present in some cases indoors at moderate to high concentrations. Nitrate radical photolyzes readily, so outdoors it is only important at night. Indoors, the normal level of lighting even in the daytime would not cause rapid photolytic destruction of nitrate. Hence, daytime nitrate levels indoors might sometimes exceed commonly occurring outdoor nitrate levels.

Because nitrate can be a relatively fast-acting oxidizing radical, because daytime nitrate levels indoors might be elevated, and because nitrate addition reactions can produce organic compounds associated with potentially high adverse health concerns, studies could usefully be undertaken to characterize nitrate levels indoors, to systematically assess the effect of important factors that influence indoor concentrations, and to investigate the production of organic nitrates in both the gas and particle phases. Studies focused on outdoor nighttime chemistry are beginning to elucidate the important role of the nitrate radical reacting with terpenes to produce secondary organic aerosol including alkyl nitrates (Rollins et al., 2012, 2013). Terpene levels are commonly elevated

indoors because of the use compounds such as d-limonene and alpha-pinene as solvents and scenting agents (Nazaroff and Weschler, 2004). To conduct studies of nitrate plus terpene chemistry directly relevant to indoor environments may require improved methods for measuring indoor nitrate radical levels. Nøjgaard (2010) used a flow tube in which nitrate-induced reactive chemistry was measured; he was unable to distinguish between nitrate and N_2O_5 . Excellent NO_3 and N_2O_5 measurement instruments are now being applied to outdoor atmospheric chemistry research, for example, using cavity ring-down spectroscopy (Dubé et al., 2006). Such methods might be adapted for use indoors.

Surface phenomena

There are at least three categories of important processes occurring on indoor surfaces that merit research attention: sorptive interactions, oxidative processes, and acid–base chemistry. A noteworthy feature of indoor surface chemistry is the long potential reactive time scales. For purely gas-phase processes, the reaction time scale of interest is limited by the air exchange rate to no more than a few hours. However, on surfaces, reactants may persist for much longer periods. Consequently, chemistry that is too slow to be important indoors in the gas-phase may be fast enough to be important if one or more of the reactants is surface bound.

Studies of sorptive interactions relevant to indoor air began a few decades ago, when it was found necessary to account for sorption in chamber studies characterizing the emissions of volatile organic compounds from building materials (Tichenor et al., 1991). Several subsequent chamber studies and field campaigns have elucidated short-term interaction processes in some important circumstances (Singer et al., 2004, 2007; Won et al., 2001). These studies have focused on the surficial aspects of sorptive interactions. Little is known, by contrast, about the ways in which the diffusion of sorbed compounds into the depth of indoor materials might influence indoor air composition. Most of the studies of sorptive interactions have focused on volatile organic compounds. Much less has been accomplished in studying sorptive partitioning of the stickier semivolatile organic compounds.

An interesting and poorly understood feature of oxidative chemistry on indoor surfaces is the potential to form secondary organic aerosol (Wang and Waring, 2014; Waring and Siegel, 2013). A surprising aspect is this: If ozone reacts with a surface bound molecule such as limonene and a more volatile compound is emitted as result, then how does that process trigger the formation of a nucleated particle? Conversely, if the reaction produces a less volatile compound than

limonene, how does that molecule ever escape the surface to form a new airborne particle?

Until now, almost all of the surface oxidative chemistry relevant to indoor environments has stressed ozone as the oxidizing agent. Are there important surface reactions in which the hydroxyl radical or the nitrate radical is the initiator? Are there differences in sorptive interactions of oxidized organics depending on whether they were produced through OH, NO_3 , or O_3 oxidation pathways?

Acid–base reactions on indoor surfaces are the least studied of these three categories. We know that there are important acidic and basic gases whose indoor concentrations can be substantial, such as carbon dioxide and ammonia. We also know that indoor surfaces can be covered with water molecules equivalent in quantity to several monolayers. Some organic compounds that sorb to indoor surfaces can act as an acid or a base in aqueous solution; a prominent example is nicotine for which the protonated form has no vapor pressure. One then should expect that gaseous reactants such as carbon dioxide and ammonia might influence the tendency of certain organic molecules such as nicotine to sorb to or desorb from indoor surfaces. Only a few investigations of this type have been reported (Ongwande and Morrison, 2008; Ongwande and Sawanyapanich, 2012).

Viewed broadly, a key challenge for research on indoor surface phenomena is to identify the most important processes to study from among the many possibilities. It may be necessary to undertake broad-ranging exploratory investigations as a basis for identifying the processes and parameters that are most interesting and important.

Semivolatile organic compounds

Under ordinary conditions, semivolatile organic compounds (SVOCs) are present to a significant extent in both the gas and condensed phases. As a class, SVOCs are diverse, prevalent, and understudied relative to their importance for both outdoor and indoor atmospheric chemistry. They exhibit complex physicochemical behavior and are probably quite important determinants of the healthfulness of indoor environment. Much remains to be learned.

There are moderate histories of scientific investigation that have focused on specific classes of SVOCs in the indoor environment, such as pesticides. Just over a decade ago, Rudel et al. (2003) published one of the first studies investigating SVOCs as a category, well beyond a single class. They sampled air and dust in 120 homes and analyzed for 89 organic chemicals, mostly SVOCs, which were believed to be endocrine disrupting compounds. In all, they detected 52 compounds in air and 66 in dust. The compounds they measured included phthalate plasticizers, a disinfectant, a

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detergent metabolite, an adhesive, flame retardant compounds, and pesticides.

Because of their low vapor pressure, SVOCs partition among many compartments in indoor environments, including airborne particulate matter, settled dust, surface films, and skin oils on occupants and on their clothing (Dodson et al., 2015; Weschler and Nazaroff, 2008, 2010). Factors such as temperature and particulate matter concentrations influence the partitioning, persistence, and fate of SVOCs. Evidence is emerging that such compounds can have very long persistence times indoors, even becoming legacy pollutants, as has been demonstrated for polychlorinated biphenyls (Frederiksen et al., 2012). Weschler and Nazaroff (2014) suggested that transport from air to the skin surface followed by transdermal permeation could constitute a meaningful exposure pathway for many SVOCs (and for some volatile organic compounds, too). Experimental evidence has just been published confirming this expectation for a few phthalates (Weschler et al., 2015).

There are many interesting, important, and worthwhile aspects of studying indoor SVOCs. An appropriate starting point would be to better characterize time-dependent concentrations of a suite of relevant compounds, including their phase partitioning. Useful studies could be performed at high time resolution in a small subset of buildings. Such sampling campaigns can reveal important insights about the underlying processes that are not evident from time-averaged sampling procedures. A second theme is to assess the roles that spatial and temporal variations in temperature and humidity play in the evolution of indoor SVOC concentrations. Temperatures can vary widely indoors, for example, with the intermittent operation of thermal conditioning systems. For compounds with vapor pressures that are highly sensitive to temperature, even modest temperature variation can strongly impact dynamic behavior and fate. A third topic of potential importance is the role of skin oils on indoor materials as a collection medium and transport vector for lipophilic SVOCs. It has been postulated (Weschler and Nazaroff, 2012) that skin oils on clothing may accumulate SVOCs while stored and then serve as a means for enhanced exposure when the clothing articles are subsequently worn.

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For all studies of SVOCs in indoor environments, a major challenge is analytical. Techniques for measuring these compounds in particle and gas-phases have historically been based on temporally averaged sampling with filters and adsorbent materials followed by offline laboratory analysis that is costly and time-consuming. Instrumentation capable of high time resolution continuous measurement of SVOCs including highly oxygenated species has recently been invented for use in the outdoor atmosphere (Isaacman et al., 2014; Zhao et al., 2013a,b). These advanced methods could now be fruitfully applied in studies of indoor chemistry.

Summary

In this editorial, we have highlighted key research opportunities and challenges in four topical themes for indoor chemistry: human occupants as agents influencing indoor chemistry; oxidative chemistry; surface phenomena; and semivolatile organic compounds. In each case, enough prior work has been done to demonstrate the importance of the theme and to create a foundation for future studies. Extensive achievements and ongoing progress in (outdoor) atmospheric chemistry—both in the analytical methods developed and in the scientific knowledge created—also contribute to a strong foundation from which to achieve rapid research progress in this exciting new domain.

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