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### Title

Indoor chemistry: research opportunities and challenges

### Permalink

<https://escholarship.org/uc/item/7jj7b63b>

### Journal

Indoor Air, 25(4)

### ISSN

0905-6947

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### Publication Date

2015-08-01

### DOI

10.1111/ina.12219

Peer reviewed



## Indoor Chemistry: Research Opportunities and Challenges

Journal:	<i>Indoor Air</i>
Manuscript ID:	INA-15-04-093
Manuscript Type:	Editorial
Date Submitted by the Author:	27-Apr-2015
Complete List of Authors:	Nazaroff, William; University of California, Civil & Environmental Engineering Goldstein, Allen; UC Berkeley, Dept of Env Sci, Pol & Mngmt
Keywords:	human emissions, oxidants, sorption, acid-base reactions, secondary organic aerosol, semivolatile organic compounds



## Indoor Chemistry: Research Opportunities and Challenges

### *Perspective*

The mass of the earth's atmosphere is  $5 \times 10^{18}$  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  $\text{m}^2$  (USEIA 2015a, 2015b). 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 to 4.6  $\text{m}^2$  per  $\text{m}^3$  for a sample of ten residential rooms that they studied. The large quantity of exposed surfaces can influence

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3 indoor air chemistry through several mechanisms, including oxidative reactions, sorption, and  
4 acid-base chemistry.  
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7 Of order  $10^4$ – $10^5$  different atmospheric organic species have been measured outdoors, and this  
8 number may be only a small fraction of those actually present (Goldstein and Galbally, 2007).  
9 Indoor air must contain a similarly broad spectrum of organic compounds. In addition to those  
10 entering from outdoors with ventilation, a variety of species are emitted from building materials,  
11 furnishings, cleaning products, the indoor microbiome, cooking, human metabolism, and  
12 personal care products among other sources. Among the broad classes of compounds found in  
13 indoor air are pesticides, plasticizers, and flame retardants. Exposure to elevated concentrations  
14 indoors can raise direct toxicity concerns for some organic species, such as formaldehyde. An  
15 additional emerging spectrum of concerns arises because of indoor exposures to endocrine  
16 disruptors, many of which are semivolatile organic compounds (SVOCs) (Rudel and Perovich,  
17 2009). The physicochemical behavior of SVOCs indoors is challenging to understand and also  
18 likely to be of considerable significance for human health and wellbeing.  
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22 A grand challenge for studies of indoor chemistry is the huge number of distinct indoor  
23 environments. In the United States, for example, there are 114 million residential units and 5.6  
24 million commercial buildings that serve a population of 320 million. Extrapolating to the global  
25 scale, one might expect in excess of one billion total indoor environments. Each may exhibit  
26 distinctive attributes. Very many chemical species are of potential interest. Concentrations of  
27 each species in each environment may vary with time to important extents. These circumstances  
28 strongly support a view that effective research on indoor chemistry should be oriented toward  
29 elucidating processes. We should seek to build a mechanistic understanding of the cause and  
30 effect relationships that facilitate the extrapolation from some finite set of specific investigations  
31 to the much larger pool of indoor environments to be characterized and understood.  
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35 The following paragraphs briefly describe frontiers of knowledge for four important themes in  
36 indoor chemistry. Significant research topics are highlighted and key challenges are identified.  
37 Our aim is to be strongly illustrative yet not comprehensive.  
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#### 40 *1. Human Occupants as Agents Influencing Indoor Chemistry*

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

11 The atmosphere is an oxidizing medium. Atmospheric chemistry is fueled by organic  
12 compounds, which enter the atmosphere in a chemically reduced state. Over time, as the carbon  
13 is oxidized, the resulting products can have important environmental consequences. Among the  
14 significant intermediate products of atmospheric organic chemistry are aldehydes and organic  
15 acids that can contribute to adverse health and welfare consequences. Functionalization of  
16 organic compounds through oxidation leads to lower volatility products that can contribute to  
17 secondary organic aerosols. The formation of secondary organic aerosol has health significance,  
18 causes light scattering, and significantly influences other important atmospheric processes. In  
19 outdoor air, during day, direct photolysis (e.g., of aldehydes) and OH-induced oxidation of  
20 organic compounds drive much of the important chemistry. At night, the nitrate radical ( $\text{NO}_3$ ) is  
21 an important catalyst.  
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26 Indoors, oxidative chemistry also can occur. Important oxidants indoors are three: ozone, the  
27 hydroxyl radical, and the nitrate radical (Gligorovski and Weschler, 2013; Waring and Wells,  
28 2015). Although more remains to be learned, the role of ozone indoors is reasonably well  
29 studied (Weschler, 2000). On the other hand, remarkably little work has been done to assess the  
30 levels, causes, and consequences indoors of the hydroxyl radical (Weschler and Shields, 1996;  
31 Weschler and Shields, 1997; Sarwar et al., 2002; White et al., 2010; Gligorovski et al., 2014).  
32 For the nitrate radical, even less is known, with only one published study reporting measured  
33 concentrations, in this case of the sum of  $\text{NO}_3$  plus  $\text{N}_2\text{O}_5$  (Nøjgaard, 2010).  
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36 Nitrate may be of particular importance in some indoor environments. The nitrate radical is  
37 formed by ozone reacting with nitrogen dioxide. Both species may be simultaneously present in  
38 some cases indoors at moderate to high concentrations. Nitrate radical photolyzes readily, so  
39 outdoors it is only important at night. Indoors, the normal level of lighting even in the daytime  
40 would not cause rapid photolytic destruction of nitrate. Hence, daytime nitrate levels indoors  
41 might sometimes exceed commonly occurring outdoor nitrate levels.  
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45 Because nitrate can be a relatively fast-acting oxidizing radical, because daytime nitrate levels  
46 indoors might be elevated, and because nitrate addition reactions can produce organic  
47 compounds associated with potentially high adverse health concerns, studies could usefully be  
48 undertaken to characterize nitrate levels indoors, to systematically assess the effect of important  
49 factors that influence indoor concentrations, and to investigate the production of organic nitrates  
50 in both the gas and particle phases. Studies focused on outdoor nighttime chemistry are  
51 beginning to elucidate the important role of the nitrate radical reacting with terpenes to produce  
52 secondary organic aerosol including alkyl nitrates (Rollins et al., 2012; Rollins et al., 2013).  
53 Terpene levels are commonly elevated indoors well above those in outdoor air because of the use  
54 compounds such as d-limonene and alpha-pinene as solvents and scenting agents (Nazaroff and  
55 Weschler, 2004). To conduct studies of nitrate plus terpene chemistry directly relevant to indoor  
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3 environments may require improved methods for measuring indoor nitrate radical levels.  
4 Nøjgaard (2010) used a flow tube in which nitrate-induced reactive chemistry was measured; he  
5 was unable to distinguish between nitrate and  $N_2O_5$ . Excellent  $NO_3$  and  $N_2O_5$  measurement  
6 instruments are now being applied to outdoor atmospheric chemistry research, for example using  
7 cavity ring-down spectroscopy (Dubé et al., 2006). Such methods might be adapted for use  
8 indoors.  
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### 10 11 3. *Surface Phenomena*

12 There are at least three categories of important processes occurring on indoor surfaces that merit  
13 research attention: sorptive interactions, oxidative processes, and acid-base chemistry. A  
14 noteworthy feature of indoor surface chemistry is the long extension of potential reactive time  
15 scales. For purely gas-phase processes, the reaction time scale of interest is limited by the air-  
16 exchange rate to no more than a few hours. However, on surfaces, reactants may persist for  
17 much longer periods. Consequently, chemistry that is too slow to be important indoors in the gas  
18 phase may be fast enough to be important if one or both of the reactants is surface bound.  
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21 Studies of sorptive interactions relevant to indoor air began a few decades ago, when it was  
22 found necessary to account for sorption in chamber studies characterizing the emissions of  
23 volatile organic compounds from building materials (Tichenor et al., 1991). Several subsequent  
24 chamber studies and field campaigns have elucidated short-term interaction processes in some  
25 important circumstances (Won et al., 2001; Singer et al., 2004; Singer et al., 2007). These  
26 studies have focused on the surficial aspects of sorptive interactions. Little is known, by  
27 contrast, about the ways in which the diffusion of sorbed compounds into the depth of indoor  
28 materials might influence indoor air composition. Most of the studies of sorptive interactions  
29 have focused on volatile organic compounds. Much less has been accomplished in studying  
30 sorptive partitioning of the stickier semivolatile organic compounds.  
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34 An interesting and poorly understood feature of oxidative chemistry on indoor surfaces is the  
35 potential to form secondary organic aerosol (Waring and Siegel, 2013; Wang and Waring, 2014).  
36 A surprising aspect is this: if ozone reacts with a surface bound molecule such as limonene and a  
37 more volatile compound is emitted as result, then how does that process trigger the formation of  
38 a nucleated particle? Conversely, if the reaction produces a less volatile compound than  
39 limonene, how does that molecule ever escape the surface to form a new airborne particle?  
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43 Until now, almost all of the surface oxidative chemistry relevant to indoor environments has  
44 stressed ozone as the oxidizing agent. Are there important surface reactions in which the  
45 hydroxyl radical or the nitrate radical is the initiator? Are there differences in sorptive  
46 interactions of oxidized organics depending on whether they were produced through OH,  $NO_3$ ,  
47 or  $O_3$  oxidation pathways?  
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51 Acid-base reactions on indoor surfaces are the least studied of these three categories. We know  
52 that there are important acidic and basic gases whose indoor concentrations can be substantial,  
53 such as carbon dioxide and ammonia. We also know that indoor surfaces can be covered with  
54 water molecules equivalent in quantity to several monolayers. Some organic compounds that  
55 sorb to indoor surfaces can act as an acid or a base in aqueous solution; a prominent example is  
56 nicotine for which the protonated form has no vapor pressure. One then should expect that  
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3 gaseous reactants such as carbon dioxide and ammonia might influence the tendency of certain  
4 organic molecules such as nicotine to sorb to or desorb from indoor surfaces. Only a few  
5 investigations of this type have been reported (Ongwandee and Morrison, 2008; Ongwandee and  
6 Sawanyapanich, 2012).  
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9 Viewed broadly, a key challenge for research on indoor surface phenomena is to identify the  
10 most important processes to study from among the many possibilities. It may be necessary to  
11 undertake broad-ranging exploratory investigations as a basis for identifying the processes and  
12 parameters that are most interesting and important.  
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#### 14 15 4. *Semivolatile Organic Compounds*

16 Under ordinary conditions, semivolatile organic compounds (SVOCs) are present to a significant  
17 extent in both the gas and condensed phases. As a class, SVOCs are diverse, prevalent, and  
18 understudied relative to their importance for both outdoor and indoor atmospheric chemistry.  
19 They exhibit complex physicochemical behavior and are probably quite important determinants  
20 of the healthfulness of indoor environment. Much remains to be learned.  
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23 There are moderate histories of scientific investigation that have focused on specific classes of  
24 SVOCs in the indoor environment, such as pesticides. Just over a decade ago, Rudel et al.  
25 (2003) published one of the first studies investigating SVOCs as a category, well beyond a single  
26 class. They sampled air and dust in 120 homes and analyzed for 89 organic chemicals, mostly  
27 SVOCs, which were believed to be endocrine disrupting compounds. In all, they detected 52  
28 compounds in air and 66 in dust. The compounds they measured included phthalate plasticizers,  
29 a disinfectant, a detergent metabolite, an adhesive, flame retardant compounds, and pesticides.  
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33 Because of their low vapor pressure, SVOCs partition among many compartments in indoor  
34 environments, including airborne particulate matter, settled dust, surface films, and skin oils on  
35 occupants and on their clothing (Weschler and Nazaroff, 2008; Weschler and Nazaroff, 2010;  
36 Dodson et al., 2015). Factors such as temperature and particulate matter concentrations  
37 influence the partitioning, persistence and fate of SVOCs. Evidence is emerging that such  
38 compounds can have very long persistence times indoors, even becoming legacy pollutants, as  
39 has been demonstrated for polychlorinated biphenyls (Frederiksen et al., 2012). Weschler and  
40 Nazaroff (2014) suggested that transport from air to the skin surface following by transdermal  
41 permeation could constitute a meaningful exposure pathway for many SVOCs (and for some  
42 volatile organic compounds, too). Experimental evidence has just been published confirming  
43 this expectation for a few phthalates (Weschler et al., in press).  
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47 There are many interesting, important and worthwhile aspects of studying indoor SVOCs. An  
48 appropriate starting point would be to better characterize time-dependent concentrations of a  
49 suite of relevant compounds, including their phase partitioning. Useful studies could be done at  
50 high time resolution in a small subset of buildings. Such sampling campaigns can reveal  
51 important insights about the underlying processes that are not evident from time-averaged  
52 sampling procedures. A second theme is to assess the roles that spatial and temporal variations  
53 in temperature and humidity play in the evolution of indoor SVOC concentrations. Temperatures  
54 can vary widely indoors, for example with the intermittent operation of thermal conditioning  
55 systems. For compounds with vapor pressures that are highly sensitive to temperature, even  
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3 modest temperature variation can strongly impact dynamic behavior and fate. A third topic of  
4 potential importance is the role of skin oils on indoor materials as a collection medium and  
5 transport vector for lipophilic SVOCs. It has been postulated (Weschler and Nazaroff, 2012) that  
6 skin oils on clothing may accumulate SVOCs while stored and then serve as a means for  
7 enhanced exposure when the clothing articles are subsequently worn.  
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10 For all studies of SVOCs in indoor environments, a major challenge is analytical. Techniques  
11 for measuring these compounds in particle and gas phases have historically been based on  
12 temporally averaged sampling with filters and adsorbent materials followed by offline laboratory  
13 analysis that is costly and time consuming. Instrumentation capable of high time resolution  
14 continuous measurement of SVOCs including highly oxygenated species has recently been  
15 invented for use in the outdoor atmosphere (Zhao et al., 2013a; Zhao et al., 2013b; Isaacman et  
16 al., 2014). These advanced methods could now be fruitfully applied in studies of indoor  
17 chemistry.  
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### 20 21 *Summary*

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

46 The authors gratefully acknowledge research support from the Alfred P. Sloan Foundation to  
47 study how emissions from humans affect indoor air chemistry. An expanded version of this  
48 editorial was provided to the Sloan Foundation as a deliverable of that research grant.  
49

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