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Indoor Environmental Chemistry and Its Relevance to Risk Management

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ABSTRACT

Risk management solutions resulting from research on indoor air quality can lead to improved human health, worker productivity, student performance, and quality of the common environment. Effective management of indoor health risks requires developing scientific and engineering information for decision making, developing and promoting technologies to protect and improve indoor environmental quality, and providing technical support and information transfer to stake holders to promote pollution prevention and sustainability. In the past, indoor environmental risk management has, for the most part, focused on control of primary pollutants, which enter the indoor environment directly from outdoor and indoor sources. Recent developments in indoor chemistry suggest that secondary pollutants, generated by chemical reactions in indoor air and on interior surfaces, may be more important than the primary pollutants, and that many types of chemically based air cleaning and decontamination methods have been developed but some have unwanted consequences. These new findings present both challenges and opportunities to indoor environmental risk management. This paper provides a brief overview of the recent advances in indoor chemistry and its relevance to indoor environment risk management.

INTRODUCTION

Indoor pollution is one of the major environmental factors affecting human health in both developing and developed countries. The public awareness of building related health problems, such as asthma, and the desire for acceptable indoor environmental quality (IEQ) are greater today than ever before. However, solving IEQ problems has proven challenging, and requires a joint effort by many types of professionals.

The ultimate goal of indoor environmental quality research is to protect human well-being by reducing the health risks and improve the quality of indoor environments. Effective management of indoor health risks requires developing scientific and engineering information for decision making, developing and promoting technologies to protect and improve indoor environmental quality, and providing technical support and information transfer to stake holders to promote pollution prevention and sustainability.

The rapid development of indoor environmental chemistry in recent years has added new dimensions to indoor environmental risk management. This paper provides a short summary of the advances in this field and discusses their implications to risk management.

RECENT ADVANCES IN INDOOR ENVIRONMENTAL CHEMISTRY

Indoor environmental chemistry, or indoor chemistry for short, utilizes the theory and methods of chemistry to study pollutant generation, transport, transformation, and fate indoors, and to improve indoor environmental quality. While studies in this field have continued for decades, major advances did not occur until the 1990s. After the turn of the century, a multitude of reviews, editorials, feature articles, summaries, and commentaries were published,[1-10] symbolizing the establishment of a new science discipline under indoor environmental science.

The major advances in indoor environmental chemistry can be broken down into seven areas: (1) chemicals in the indoor environment, (2) chemical reactions in indoor sources, (3) chemical reactions in indoor air, (4) chemical reactions on interior surfaces, (5) chemically based air cleaning, (6) chemically based decontamination, and (7) indoor analytical chemistry. Brief discussions on the first six areas are provided below. The last topic is ignored because of insufficient collection of information. It is recommended that it be discussed by others in the near future.

Chemicals in the Indoor Environment

The U.S. EPA and U.S. Consumer Product Safety Commission (CPSC) identified 11 major indoor air pollutants in homes: radon, environmental tobacco smoke, biologicals, carbon monoxide, nitrogen dioxide, organic gases, respirable particles, formaldehyde, pesticides, asbestos, and lead[11]. However, many more chemicals can be found in the indoor environments. U.S. EPA's Source Ranking Database[12] complies formulation data for about 12,000 potential indoor pollution sources, and lists 1377 chemical ingredients. The Household Products Database developed by the National Institute of Health[13] contains 2512 chemical ingredients used in 5011 household products. In both databases, each chemical ingredient is associated with a unique CAS Registry number. Although many of those chemicals are not of immediate or direct health concerns, some are potentially harmful. Thus, understanding their occurrence, concentrations, and distribution is one of the first steps to identify potential pollutants and their sources.

Indoor pesticides are a special group of consumer products that include biocides, fungicides, and insecticides. According to Godish[14], there are approximately 20,000 household pesticide products used in the United States and they include 300 active ingredients and 1700 inert ingredients. Butte[15] compiled recent studies on pesticides concentrations in non-occupational indoor environments in indoor air and house dust.

There are two major sources of combustion-generated chemicals: fuel combustion and environmental tobacco smoke. In addition to carbon dioxide and water, fuel combustion (including burning candles and incense) also generates nitrogen oxides, carbon monoxide, particulate matter, aldehydes, volatile organic compounds, and polycyclic aromatic hydrocarbons. Fuels containing sulfur will produce sulfur dioxide.[14] Combustion of biofuels in homes in developing countries is one of the most serious indoor air pollution problems in the world.

Over 4000 compounds have been identified in laboratory-based studies of mainstream tobacco smoke.[16,17] Major chemicals in gas and particle phases of tobacco smoke can be found in refs 18 and 19.

Biologically generated chemicals include microbial volatile organic compounds (MVOCs), bacterial and fungal toxins, and allergens. Over 100 MVOCs have been identified, including alcohols, aldehydes, ketones, esters, ethers, terpenes, furans, and aromatic compounds.[20]

Outdoor contaminants can enter the building through penetration, intrusion, and tracking-in. A useful list of major air pollutants is the 189 original hazardous air pollutants (HAPs) published by U.S EPA under the Clean Air Act Amendment of 1990 (http://www.epa.gov/ttn/atw/ orig189.html). A few modifications have been made since the initial publication of the list (http://www.epa.gov/ttn/atw/pollutants/atwsmod.html).

Chemical Reactions in Indoor Sources

Pollutant emissions from indoor sources may involve physical, chemical, and biological processes. The presence of chemical reactions often makes the emission pattern complex and difficult to predict. For instance, urea-formaldehyde resins, which are widely used as binding agents in manufacturing engineered wood, are a well-known source of indoor formaldehyde. In addition to residual formaldehyde, hydrolysis of the methylol end groups and, less commonly, methylene bridges of the resin molecules also generate formaldehyde.[14] It is the hydrolytic reactions that make the emission rate sensitive to moisture content in the material and in indoor air. Thus, understanding of the chemical reactions occurring in indoor sources is essential not only for indoor source characterization but also for pollution prevention and risk management. Table 1 lists major types of chemical reactions in indoor sources that have been studied in recent years.

Table 1. Summary of chemical reactions in indoor sources

Source Type	Chemical Class	Reaction Type	Reaction Products	References
Water-based cleaners	Ethoxylated alcohols	autoxidation	peroxides, formaldehyde, ethoxylated aldehydes	21-24
Alkyd paint	Unsaturated fatty acids	autoxidation	hydroperoxides and aldehydes	25-29
Engineered wood	Formaldehyde resins	hydrolysis	hydroperoxides and aldehydes	14, 30, 31
Concrete	Alkalinity*	alkaline hydrolysis	alcohols, etc.	32-34
Fluoropolymer treated articles (i.e, non-stick cookware)	fluoropolymers	oxidative pyrolysis	fluorinated acids, small fluorocarbons	35-36
SF_6 tracer gas	--	oxidative pyrolysis	SO_2, HF, H_2SO_4	37
UV-curing coating materials	photoinitiators	photolysis	benzaldehyde, cyclohexanone, and benzophenone, and monomers	38
Ozone generator	oxygen in air	ozonization	ozone	39
Dry-process photocopier	oxygen in air	ozonization	ozone	40
Interior surface materials	many organic chemicals	biodegradation	MVOCs	20, 41-43

* Causing flooring components attached to the concrete (e.g., adhesives, plasters, carpet backing, etc) to decompose.

Chemical Reactions in Indoor Air

In most indoor environments, there is little or no direct solar irradiation that can initiate vigorous photochemical reactions. Therefore, "dark" reactions are more important in the indoor environment than many photo-induced reactions responsible for the urban smog. On the other hand, many indoor sources can emit chemically reactive compounds, and chemicals sensitive to photolysis tend to have longer lives indoors. In recent years, researchers have attempted to identify potentially important gas-phase reactions based on reaction mechanisms and kinetics, as well as potentially hazardous reaction products. Weschler and Shields[3] proposed five broad categories of reactions that may be important indoors: (1) reactions between ozone and unsaturated hydrocarbons, (2) reactions between ozone and nitrogen oxides, (3) thermal decomposition of peroxyacyl nitrates, (4) free radical reactions, and (5) heterogeneous reactions. The same authors also listed several categories of reaction products that may be important to indoor air quality, including aldehyde, ketones, carbonyl acids, peroxyacyl nitrates, and stabilized free radicals on aerosols. The role of free radicals in indoor air chemistry have been discussed by several authors.[7, 8, 44-46]

Many laboratory studies have been conducted to investigate the reactions that are potentially important to indoor air quality. Most of the studies focused on ozone initiated reactions, especially ozone-terpene reactions. Aldehydes, secondary organic aerosols, and hydrogen peroxide are among the most extensively studied reaction products.[47-62] Although still rare, determinations of rate constants for reactions relevant to indoor environments have also been reported.[48, 63]

Over a dozen field measurements of chemically reactive species indoors have been reported. These studies provide evidences that indoor chemistry may play an important role in affecting indoor environmental quality, at least under certain conditions. Measurements have been focused on chemically reactive species[47, 64-74] and OH radicals.[75]

Studies on indoor air chemistry models started over two decades ago.[76] Since then, several papers have been published.[46, 77-80] Overall, indoor chemistry models are far from mature. Representations of heterogeneous reactions are especially weak.

Chemical Reactions on Interior Surfaces

A unique feature of all indoor environments is the large and diverse interior surfaces. Therefore, interactions between airborne species and the surfaces are a critical part of indoor chemistry. As early as three decades ago, researchers started to notice that the decay of nitrogen dioxide concentrations inside buildings is faster than the theoretical prediction based solely on the ventilation rate.[81] Weschler[9] suggested that potentially important heterogeneous reactions at interior surfaces include ozone-initiated surface chemistry, surface chemistry on building filters, acid-base surface chemistry, and surface chemistry involving esters and damp.

One important research area of indoor heterogeneous reactions is the deposition rates of chemically reactive species on surfaces. Grøntoft and Raychaudhuri[82] compiled experimentally determined deposition velocities published in the past three decades for ozone, nitrogen dioxide, and sulfur dioxide on different surfaces. The data ranges were 0.0007 to 0.109 cm/s for ozone, 0.0006 to 0.0919 cm/s for nitrogen dioxide, and 0.002 to 0.01 cm/s for sulfur dioxide.

Another important research area is the formation of secondary pollutants by heterogeneous reactions. Spicer et al.[83] studied the interaction of NO_2 with 34 materials commonly used in homes. They found that the reaction products include HONO, HNO_3, and NO, and that the distribution of nitrogen among the three reaction products is material dependent. Interaction of ozone with carpet is among the most intensively studied indoor heterogeneous reactions so far, with aldehydes being the major products of concern.[1, 84, 85] The results suggest that ozone may react with the unsaturated volatile hydrocarbons from the carpets, carpet fibers, and vegetable oils coated on the carpet. Other types of surfaces that react with ozone include latex paint[86], laminated counter-top coated with oils and detergent[87], and simulated aircraft cabin.[88]

Chemically-Based Air Cleaning

In addition to studying indoor pollutant sources, transport, transformation, and fate, indoor environmental chemistry also plays a vital role in solving IEQ problems and improving IEQ. Chemically based air cleaning is one of such areas. According to Daniels,[89] air cleaning devices fall into six categories: (1) bipolar air ionization, (2) ozone generation, (3) electrostatic precipitation, (4) gas-phase filtration, (5) solid media filtration, and (6) catalytic oxidation. Apparently, chemical processes are involved in most of these air cleaning devices.

Air cleaning devices based on chemosorption have been used for a long time. In chemosorption, the gas molecules are held to the surface by relatively strong chemical bonds, and the molar enthalpy change ranges from -10 to -200 kcal/mole. Chemosorption media are typically produced by impregnating sorbents (activated carbon, activated alumina, silica gel, etc.) with chemically reactive compounds, such as bromine, metal oxides, iodine, potassium iodide, and sodium sulfide. Most of these chemicals are highly selective in removing air pollutants. For instance, elemental sulfur is effective only for mercury. Activated alumina impregnated with potassium permanganate ($KMnO_4$) is effective in removing low-molecular-weight gases (e.g., HCHO) and has been used in various types of industrial and commercial air cleaning systems.[90] However, like many other types of air cleaners that remove VOCs by oxidation, this method may result in incomplete oxidation, generating aldehydes, ketones, and acids. Overall, progress in this area has been slow and applications of chemosorption methods to residential buildings have been limited.

Air ionizers are widely available on the market. Air ions can be generated by corona discharge. Most air ion generators used indoors are designed to produce negative air ions

(NAIs), with superoxide (O_2^-) being the major and most stable ion.[91] When the ions adhere to particles, the latter can be attracted to interior surfaces or filters more easily. Therefore, air ion generators are used mainly to remove indoor particles.[92] Germicidal effects are also reported.[93] Air ions can also react with VOCs, but the removal rates vary.[91] Potential problems associated with this type of air cleaners include partial oxidation products (i.e., aldehydes)[91] and ozone generation.[92, 93] The latter can be minimized by design or destroyed after generation. Total elimination of ozone is difficult, however.

Ozone generators are sold as cleaning devices in many countries. They generate ozone by silent electric discharge. Concerns over these products include (1) ozone is an air pollutant itself, (2) low removal efficiency for indoor pollutants, and (3) potential formation of secondary air pollutants. Many studies have been conducted to evaluate both the positive and negative effects (e.g., ref 94). Most researchers agree that ozone generation is not a practical and effective means of improving indoor air quality.

Using catalytic methods to remove indoor air pollutants is a relatively new research area of environmental catalysis.[95] Interest in this area, especially in photocatalysis, has been growing rapidly. Photocatalysis is used as a non-specific term for any catalysis in which photons are implicated. For air cleaning, heterogeneous photocatalysis – a technology based on the irradiation of a semiconductor such as titanium dioxide (TiO_2) – is most commonly used. The basic principles and mechanisms of photocatalysis can be found in ref 96. Most researchers believe that the hydroxyl radicals generated at the surface of the catalyst are the primary oxidizing species. For indoor applications, this technology has been used to mineralize VOCs and inactivate microorganisms. A large body of publications is now available on this topic, including a literature review by Zhao and Young.[97] Although this technology is promising, studies show that some systems sold at the market cannot remove formaldehyde efficiently.[98] In addition, the potential adverse effects need to be addressed. For instance, formation of partially oxidized products (e.g., aldehydes) have been reported.[99, 100]

Chemically Based Decontamination

Indoor decontamination methods fall into two categories: surface decontamination (with liquids, foams, or gels) and building decontamination (with gases or vapors). Unlike air cleaning processes, which remove pollutants from indoor air, the goal of decontamination is to eliminate or modify pollution sources.

Surface cleaners are used in homes and offices on daily basis for floors, furniture, and other surfaces. Many of the cleaning products also serve as disinfectants. Potentially hazardous chemicals used in the formulations and potential formation of secondary pollutants are discussed by Wolkoff et al.[101], Nazaroff and Weschler[102], and Guo.[103] Brief discussions on cleaning methods (including duct cleaning) are given by Kildesø and Schneider.[104] Surface decontamination is also used to remove contaminants deliberately released into a building. Available methods are summarized in ref 105. Cleaning of unlined air duct and duct liners is a special type of indoor surface decontamination. A variety of antifungal chemicals have been used in polymeric sealants to prevent fungal contamination in the heating, ventilation, and air-conditioning systems.[106, 107]

Building decontamination is needed when the indoor pollutant sources (e.g., molds) are so widespread that air cleaning cannot solve the problem, or when hazardous chemical or biological agents are accidentally or deliberately released into the building. The most commonly used chemical method for building decontamination is gas or vapor fumigation.

Ammonia fumigation has been used to reduce the formaldehyde levels in mobile homes. Ammonia reacts with free formaldehyde to form a solid, fused-ring compound known as hexamethylene tetramine or urotropine. Ammonia is generated on site by heating ammonium bicarbonate (NH_4HCO_3) or ammonium carbamate ($NH_4CO_2NH_2$). In addition to reacting with free formaldehyde, ammonia is said to also react with, and thus stabilize, the methylol end groups on the urea-formaldehyde resin, making the resin less susceptible to hydrolysis. A concise summary of this method, including pros and cons, is given by Godish.[14]

Several chemical methods have been developed for control of biological contaminants in buildings. Fumigants include chlorine dioxide, hydrogen peroxide, formaldehyde, and methyl bromide (CH_3Br) as fumigant. An excellent review on this topic is given by ref 105**.**

RELEVANCE TO INDOOR ENVIRONMENTAL RISK MANAGEMENT

As a decision-making process, risk management weighs scientific evidence, political judgement, and health and economic interests of various stake holders in deciding how to incorporate risk assessment results.[108] Indoor environmental risk management can benefit from indoor environmental chemistry through better understanding of pollutant sources, transport and fate and through the development of effective pollution control technologies and pollution prevention strategies. In particular, the knowledge of indoor environmental chemistry helps define the problem, analyzing the risks, and examine the options. For instance, we now know that chemical reactions (e.g., hydrolysis and autoxidaton) within the sources may affect the emissions significantly. Indoor homogeneous and heterogeneous reactions may generate pollutants that are more harmful than their parent chemicals. Many new indoor sources found in recent years are direct results of indoor chemistry research (see Table 1). Understanding of the chemical processes in the urea-formaldehyde resins has led to the reduction of formaldehyde emissions from engineered wood in the last two decades. Such knowledge has also helped researchers understand why humidity affects formaldehyde emission and to what extent humidity control can reduce the emission. Results from studies on ozone deposition have made it easier to predict indoor ozone levels by mathematical modeling. There is no doubt that further research on indoor chemistry will solve more IEQ problems. For instance, identification of exthoxylated non-ionic surfactant that are less sensitive to autoxidation may reduce the formation of formaldehyde in household cleaners.

Advances of indoor environmental chemistry also pose challenges to indoor environmental risk management. Thousands of chemicals have been brought to the indoor environment by consumer products, only a fraction of those chemicals have been characterized. More potentially hazardous chemicals are yet to be identified in the indoor environment. For example, recent discovery of high indoor-to-outdoor ratios for perfluorinated carbonyl compounds[109, 110] show the importance of indoor exposure to these potentially hazardous chemicals. Hundreds of pesticides are current used for indoor applications but there is little data about their levels in homes.

Examination of potential risk management options requires evaluation of their effectiveness, feasibility, costs, benefits, unintended consequences, and culture and social impacts.[111] Many types of air cleaning devices are available on the market. With few exceptions, most chemically based air cleaners have some unwanted consequences such as generation of air pollutants (e.g., ozone and NOx) and incomplete oxidation of organic air pollutants (e.g., aldehydes and acids). Data are needed for quantitative evaluation of unwanted consequences. As an example, many papers have been published on photocatalytic oxidation, few paid attention to incomplete oxidation. The lack of data for potential negative effects has made it difficult to weigh the potential benefits and risks for certain air cleaners.

Formation of secondary pollutants due to indoor reactions demands more careful evaluation of risk management options. For instance, to control indoor formaldehyde levels, one need not only to consider primary sources such as furniture, but also contributions from other sources (e.g., ozone-terpene reactions, autoxidation of certain chemicals in paint and water-based cleaners, and even certain air cleaning devices). In addition, certain seemingly harmless chemicals (i.e., terpenes) may produce hazardous pollutants by reacting with other chemicals in air or on surfaces. Thus, identification and prioritization of primary and secondary sources is essential to pollution prevention and risk reduction strategies.

CONCLUSION

Indoor environmental chemistry utilizes the theory and methods of chemistry to study pollutant generation, transport, transformation, and fate indoors, and to improve indoor environmental quality. While gas-phase homogeneous reactions and heterogeneous reactions on interior surfaces have been rightfully under the spotlight in recent years, Significant advances have been achieved in other areas including identification of chemical pollutants and other chemicals in the indoor environment, chemicals reactions in indoor sources, chemically based air cleaning, chemically based decontamination, and indoor environmental analytical chemistry. As a decision-making process, indoor environmental risk management can benefit from the knowledge of indoor chemistry through better understanding of the behaviors of the pollutants and pollution control options. On the other hand, indoor chemistry poses challenges to risk management by adding new dimensions to the decision-making process. For instance, the risk reduction strategies must consider secondary pollutants; selection of methods for air cleaning and building decontamination must weigh the benefits and unwanted consequences.

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KEY WORDS

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