ARTICLE IN PRESS

International Journal of Hygiene and Environmental Health xxx (xxxx) xxx-xxx



Review

Contents lists available at ScienceDirect

International Journal of Hygiene and Environmental Health



journal homepage: www.elsevier.com/locate/ijheh

Reactive indoor air chemistry and health—A workshop summary

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ARTICLE INFO

Keywords: Health effects of indoor air chemistry Indoor air quality Indoor air modeling Oxidants Reactive indoor chemistry

ABSTRACT

The chemical composition of indoor air changes due to the reactive nature of the indoor environment. Historically, only the stable parent compounds were investigated due to their ease of measurement by conventional methods. Today, however, scientists can better characterize oxidation products (gas and particulatephase) formed by indoor chemistry. An understanding of occupant exposure can be developed through the investigation of indoor oxidants, the use of derivatization techniques, atmospheric pressure detection, the development of real-time technologies, and improved complex modeling techniques. Moreover, the connection between exposure and health effects is now receiving more attention from the research community. Nevertheless, a need still exists for improved understanding of the possible link between indoor air chemistry and observed acute or chronic health effects and long-term effects such as work-related asthma.

1. Introduction

Indoor chemicals' oxidation processes can be driven in the gas phase by oxidants like ozone (O₃), hydroxyl radicals (OH) and nitrate radicals (NO₃) and can lead to the formation of oxygenated species (*e.g.* formaldehyde) and secondary organic aerosols (SOA). Detection and quantification of these oxidants in conjunction with oxidant precursors, reactants, *and* the reaction products (such as oxygenated organics, organic nitrates, SOA) are necessary to understand the oxidation processes indoors. This capability to measure oxidized species is important for characterizing the numerous contributions (emission, outdoor input, homogeneous and heterogeneous chemistry, ...) that can lead to their formation, as well as gas- and surface-phase chemistry that can lead to their removal and the formation of new oxidation products.

Oxidative chemistry occurring indoors leads to the formation of several traditionally observed organics such as aldehydes (*e.g.* formaldehyde), ketones (e.g. acetone), carboxylic acids, esters, epoxides and dicarbonyls (Atkinson and Arey, 2003; Finlayson-Pitts and Pitts, 2000); if their vapor pressure is sufficiently low, SOA are formed. However, numerous other oxidized species, such as primary/secondary ozonides, peroxides, organic nitrates, and multi-functional organics (*e.g.* hydroxy and nitroxy alkyl radicals, peroxy-hemiacetals, and carbonyl nitrates), and polymeric species are also generated indoors and require specialized detection methods (Atkinson and Arey, 2003; Docherty et al., 2005; Epstein et al., 2010; Li et al., 2002; Mutzel et al., 2013; Nørgaard et al., 2013; Tobias and Ziemann, 2000). Characterizing the formation (identification and yields) of these products and their respective phases (gas or particulate) in the indoor environment may help to resolve the gap between indoor occupant exposure and health effects. Understanding the physiological responses to these exposures is also a challenging endeavor. Potential avenues leading to health effects in the airways and the cardiovascular system include: sensory irritation, inflammatory reactions in the airways, sensitization, heart rate effects, delayed physiological response, and possibly dermal exposure routes (Nazaroff and Goldstein, 2015; Weschler and Nazaroff, 2012).

In an effort to highlight the recent developments toward understanding indoor air quality (IAQ), a session entitled "Reactive Indoor Air Chemistry and Health" was held at the 14th International Conference on Indoor Air Quality and Climate (Indoor Air 2016) in Ghent, Belgium, July 3–8, 2016. The workshop presentations included the following topics: The Role of Oxidants, Analytical Technologies, Modeling, and Health Effect Studies. Each of these topics will be discussed in a separate section below. While this summary is not all inclusive, it provides a current update of the topics highlighted above, recognizing that "reactive chemistry", *per se*, is a much broader field,

http://dx.doi.org/10.1016/j.ijheh.2017.09.009

Please cite this article as: Wells, J.R., International Journal of Hygiene and Environmental Health (2017), http://dx.doi.org/10.1016/j.ijheh.2017.09.009

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Received 18 May 2017; Received in revised form 18 September 2017; Accepted 22 September 2017 1438-4639/ Published by Elsevier GmbH.

see e.g. (Uhde and Salthammer, 2007).

2. Role of oxidants: sources and impact on the indoor air quality

The limited number of oxidant (O_3 , OH, and NO_3) measurements is due to both concentration and analytical challenges. There are commercially available instruments to measure the concentration of indoor O_3 which is about 10^{11} molecules cm⁻³ (a few dozen ppb). However, indoor O_3 concentration is strongly dependent on the air exchange rate (AER) and the outdoor concentration. OH and NO_3 are even more challenging to measure because their concentrations have been estimated to be 10^5 and 10^7 molecules cm⁻³ (4×10^{-3} and 0.4 ppt), respectively, and being highly reactive, they are difficult to collect and analyze (Sarwar et al., 2003). Only one article reports a measurement of the sum of N_2O_5 and NO_3 concentrations indoors in the range of 10^7 – 10^8 molecules cm⁻³ (Nøjgaard, 2010). Existing instruments, developed for atmospheric measurements could be deployed in the future (Fuchs et al., 2008) to characterize NO_3 and other species such as N_2O_5 (Goulette et al., 2016; Schuster et al., 2009; Womack et al., 2017).

For OH radicals indoors and more generally HO_x (OH and hydroperoxyl (HO₂) radicals), advances in optical spectroscopy and detection technologies have contributed to improved characterization of these elusive species. Instruments based on spectroscopic techniques (Fluorescence Assay by Gas Expansion, FAGE) capable of measuring real-time OH and HO₂ radicals have already been deployed by two groups: Lille (France) (Blocquet et al., 2016; Gómez Alvarez et al., 2013) and Leeds (United Kingdom) (Carslaw et al., 2017) to quantify HO_x in different buildings under different conditions.

Two major sources of HO_x radicals have been identified: the photolysis of nitrous acid (HONO) and the reaction of O₃ with alkenes (Blocquet et al., 2016; Carslaw et al., 2017; Gómez Alvarez et al., 2013; Mendez et al., 2017a; Weschler and Shields, 1996). Measurements indicate multiple sources of HO_x and the relative importance of each source will depend strongly on the ambient conditions, an association that has been implicated in recent models (Carslaw, 2016; Mendez et al., 2017a). Compared to predicted and previously measured indoors (Sarwar et al., 2003; Weschler and Shields, 1997; White et al., 2010), high concentrations (up to 10^7 molecules cm⁻³ for OH) have been measured during the use of an air cleaning device and cleaning products (Carslaw et al., 2017).

The investigation of indoor radical concentrations has highlighted the need for numerous ancilliary measurement techniques such as: the sunlight transmission through windows to quantify the photolysis processes and the light distribution in the room (Gandolfo et al., 2016; Kowal et al., 2017), the radicals' precursors (HONO, O₃, alkenes, ...), species involved in the recycling of the radicals (like NO), and a better understanding of the linkage of heterogeneous processes (especially HONO production) on indoor surfaces (Gómez Alvarez et al., 2014; Mendez et al., 2017b). These investigations have also challenged previous assumptions about indoor oxidation pathways such as photolysis indoors. Additionally, recent research using high temporal resolution instruments such as proton-transfer reaction mass spectrometry (PTR-MS) investigated the potential of occupants to contribute to indoor chemistry (Tang et al., 2016; Wisthaler and Weschler, 2010) and showed that occupants can react with ozone and emit oxidized organic compounds (Liu et al., 2016, 2017; Tang et al., 2015; Zhou et al., 2016a,b). There is continous development of new methods for the detection of transient oxidant species, related intermediate species (like peroxyl radicals RO₂) for atmospheric applications (Tan et al., 2016; Whalley et al., 2013), and parameters such as the OH reactivity (representing the sum of OH removal reactions) (Blocquet et al., 2016; Fuchs et al., 2017). There is also interest in developing the use of these instruments in indoor environments to better characterize the gasphase chemistry. Complementary research concerning kinetic studies of interest for indoor chemistry (Borduas et al., 2016) and measurement in real condtions are needed to evaluate its impact.

3. Analytical technologies: methods/instrumentation for indoor air contaminants

3.1. Gas-phase and particulate-phase measurements

As discussed above, understanding volatile organic compounds' (VOC) oxidation indoors is important for assessing gas-phase and particulate-phase occupant exposure. Thus, collection and transport of compounds without degradation for off-line laboratory analysis becomes relevant. Several methods have been used for field measurements that maintain compound stability until analysis, such as: active and passive desorption sampling tubes, canisters, annular denuders, impingers, and solid-phase microextraction (SPME) and chemical derivatization (Forester and Wells, 2009; Ham et al., 2016, 2015; Harrison and Wells, 2013; Jackson et al., 2017; Plog, 2012; Wells and Ham, 2014).

Gas-phase oxidation products have been measured using Fourier transform infrared spectroscopy (FTIR), gas and high-performance liquid chromatography/mass spectrometry (GC/MS and HPLC/MS), but recent techniques such as PTR-MS and atmospheric-pressure ionization mass spectrometry (API-MS) provide the advantage of real-time or near real-time data of target compounds coupled with high sensitivity (Cochran et al., 2016; Nozière et al., 2015).

The need to characterize gas- and particle-phase species in real-time continues to grow. Current real-time instrumentation typically collects information on total organics for the gas-phase species and particle number, size, surface area and distribution over time for the particulate-phase (Stefaniak, 2016). Manufacturers and academia have worked to address this need through the development of miniature GC/MS devices and gas-specific sensors; however, chromatography limits, power requirements, sensor "fouling", and the sheer number of potential oxidized compounds continue to plague their integration into the field (Brüggemann et al., 2015; Laborie et al., 2016; Nölscher et al., 2012; Wang et al., 2015; Wolf et al., 2016; Wolf et al., 2015a,b; Zhou et al., 2015).

3.2. Reactive oxygen species (ROS)

Reactive oxygen species can occur indoors (Fan et al., 2005) and include chemical species such as peroxides (ROOR'), OH, superoxide (O_2^-), hydrogen peroxide (H_2O_2), HO_2 , hypochlorite ions (OCl⁻) and O₃. Exposure to these species could induce oxidative stress in the respiratory tract and other areas such as skin (Brem et al., 2017; Kehrer, 1993; Klaunig and Kamendulis, 2004; Schuch et al., 2017). Indoor ROS concentrations have been measured by derivatizing them with 2',7'dichlorofluorescein diacetate to form the fluorescent compound, dichlorofluorescein (Hung and Wang, 2001; Venkatachari et al., 2005). Indoor air ROS is determined as a concentration, yet actual chemical structural information remains elusive (Hopke et al., 2011; Khurshid et al., 2014, 2016; Liu and Hopke, 2014; Pavlovic and Hopke, 2011).

4. Modeling to characterize personal exposure to reactive chemistry

Exposure is defined as the time integral of concentration between relevant time durations of interest. Utilizing the U.S. National Research Council 1983 Risk Assessment Paradigm, once *hazard identification* occurs, *exposure* and *dose-response assessments* occur in parallel and are combined to provide information on the *risk characterization*, for which a *risk management* program may be developed if warranted. One problem with using modeling within this approach is that modeling typically predicts indoor, not personal concentrations, which is the most relevant parameter for exposure assessments. However, combining computational fluid dynamics (CFD) with reactive chemistry modeling could be used to develop personal factors, *PF*, defined as *PF* = (personal exposure concentration/room concentration), in the future for a variety Download English Version:

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