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Oxygenated VOCs, aqueous chemistry, and potential impacts on residential indoor air composition

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Abstract

Dampness affects a substantial percentage of homes and is associated with increased risk of respiratory ailments; yet the effects of dampness on indoor chemistry are largely unknown. We hypothesize that the presence of water-soluble gases and their aqueous processing alters the chemical composition of indoor air and thereby affects inhalation and dermal exposures in damp homes.

Herein, we use the existing literature and new measurements to examine the plausibility of this hypothesis, summarize existing evidence, and identify key knowledge gaps. While measurements of indoor volatile organic compounds (VOCs) are abundant, measurements of water-soluble organic gases (WSOGs) are not. We found that concentrations of total WSOGs were, on average, 15 times higher inside homes than immediately outside (N=13). We provide insights into WSOG compounds likely to be present indoors using peer-reviewed literature and insights from atmospheric chemistry. Finally, we discuss types of aqueous chemistry that may occur on indoor surfaces and speculate how this chemistry could affect indoor exposures. Liquid water quantities, identities of water-soluble compounds, the dominant chemistry, and fate of aqueous products are poorly understood. These limitations hamper our ability to determine the effects of aqueous indoor chemistry on dermal and inhalation exposures in damp homes.

Keywords

Damp homes; Interfacial chemistry; Aqueous chemistry; Water-soluble organic compounds; OVOC; Human exposure

Introduction

Dampness in buildings is common in the United States, with estimates ranging from 18 to 50% percent of buildings affected, where buildings were defined as “damp” based on observation of standing water, water damaged materials, presence of mold and/or high measured relative humidity (RH)^{1,2}. Dampness and high RH indoors can be caused by high humidity outdoors, water leakage into buildings, leakage of pipes, flooding, water

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infiltration into building materials, and moisture resulting from human activities such as cooking and bathing³. Dampness can also occur as a result of water vapor condensation in the building structure or on surfaces indoors. Building dampness is associated with increased risk of respiratory symptoms such as cough, wheeze, asthma, and respiratory infections⁴. Dampness is also associated with mold exposures. Two scientific reviews report that dampness or mold exposures result in increased respiratory symptoms with a range of odds ratios from 1.30–1.75 and 1.4–2.2, respectively^{3,4}. Mold and mildew indoors and its health effects have been studied extensively. However, to date, mold and mildew have only been able to partially explain the associations between damp buildings and adverse health outcomes, and causal linkages remain weak⁴. Certainly, there could be additional chemical, physical, or biological factors that contribute to adverse health in damp spaces.

We hypothesize that aqueous uptake and processing of water-soluble gases in damp homes alters the chemical composition of indoor air and can affect dermal and inhalation exposure. If true, aqueous chemistry indoors could plausibly affect health in damp homes. Aqueous chemistry outdoors (in clouds, fogs, and wet particles) has been demonstrated to alter the concentrations and composition of *outdoor* gases and particles;^{5–8} aqueous chemistry may also occur indoors in damp buildings where the surface area for water condensation is large. In fact, the observation of HONO production in residences^{9,10} provides definitive evidence that indoor liquid water concentrations are sufficient, at least in certain homes/times, to drive aqueous chemistry because HONO is produced on surfaces (from NO₂ hydrolysis) only in the presence of liquid water¹¹.

For aqueous processing indoors to be an important factor for indoor air chemical composition, liquid water and water-soluble gases must be present indoors. When dampness occurs in homes, liquid water can be present in and on indoor surfaces as well as in hygroscopic airborne particles. Knowledge about water-soluble gases indoors is limited, although nonpolar volatile organic compounds (VOCs) have been extensively measured. VOCs are frequently measured at much higher concentrations indoors than outdoors. A systematic review of indoor and outdoor VOC measurements found indoor/outdoor residential VOC ratios range from 1 to 150¹². One study that measured 30 VOCs using GC-FID inside and outside homes in Helsinki, Finland, found indoor/outdoor VOC ratios of measured compounds ranged from 0.25 to 55 with an average ratio of 6¹³ (compounds with higher concentrations outside homes tended to be associated with vehicle emissions). We speculate that, similar to non-polar VOCs, polar and water-soluble VOCs are present at higher concentrations indoors than outdoors. While measurements of formaldehyde are relatively common and other targeted polar VOCs are sometimes measured indoors^{14–16}, measurement of polar and water-soluble VOCs (also called *oxygenated* VOCs, OVOCs; or water-soluble organic gases, WSOGs) is, in general, more challenging.

WSOGs are emitted from indoor sources and formed through oxidation. They will be taken up into liquid water, when present, and subsequently react. Thus, aqueous chemistry could affect exposure by acting as a sink for certain water-soluble gases in indoor air and a source of other volatile products (altering inhalation exposure) and condensed phase products (altering particle inhalation and dermal exposure).

This paper provides insights into WSOGs likely to be in residential indoor air and examines the potential for aqueous chemistry indoors to alter the chemical composition of this air, with the motivation of further understanding dermal and inhalation exposure to gaseous and particulate species in homes. We demonstrate that water-soluble organic gases are elevated in residential indoor environments. We make use of the literature to document water-soluble compounds measured or likely to be present in homes. Finally, we discuss knowledge and knowledge gaps concerning indoor aqueous chemistry and its implications, and we make recommendations for future research.

Approach

First, we address the potential importance of indoor water-soluble organic gases (WSOGs). Because, to our knowledge, measurements of *total* WSOGs have not previously been made indoors, we conducted these measurements inside 13 homes and directly outside for comparison. We then provide a list of water-soluble gas-phase compounds that have been measured in residential indoor air. Next, we use knowledge about emissions, non-polar VOC concentrations, and oxidation chemistry to identify additional water-soluble organic compounds that are likely to be present indoors. This is followed by presentation of the case for indoor aqueous chemistry. We familiarize the reader with outdoor atmospheric aqueous chemistry and speculate about wet chemistry on indoor surfaces. Finally, we discuss the potential for aqueous chemistry to alter indoor exposures and summarize major knowledge gaps.

Indoor and outdoor concentrations of *total* WSOG were determined by collecting the ambient mix of water-soluble gases into water using Cofer scrubbers¹⁷, also called mist chambers, and measuring the total carbon collected. While it is possible to target specific water-soluble compounds by optimizing analytical methods and through the use of authentic standards, such an approach would likely only provide a small fraction of total. The goal of these measurements is to determine whether or not total WSOG is enriched indoors compared to outdoors.

For this purpose, field samples were collected from a convenience sample of 3 homes in central New Jersey and 10 homes in the Triangle region of North Carolina between June and October 2015; each home was sampled once. WSOGs were collected from particle-filtered air (particles filtered with pre-baked quartz fiber filters (Pall, 47 mm)) using 2 mist chambers¹⁷ in parallel sampling from a common area of the home and 2 simultaneously sampling directly outside the home. Each mist chamber scrubbed the air with an air flow rate of 25 L/min and water collection volume of 25 mL (ultra-pure deionized water, conductivity = 17.8 ± 0.5 M Ω) for a duration of two hours, twice consecutively. Indoor and outdoor samples were composited separately for each home, analyzed for total organic carbon using a Shimadzu 5000a TOC analyzer as described previously^(18,19), and the remaining volume was frozen for future research.

In order to gain additional insights into WSOGs in indoor air, a literature search was performed on September 14–15, 2015 using the Scopus database. The search terms using the Boolean operators “and” and “or” were:

One of the following: Volatile, semi-volatile, gas phase, gas-phase, organic, organic compounds, organic gases, aldehydes, aromatic hydrocarbons, terpenes, alcohols, alkanes, alkenes, epoxides, phenols, ethers, carboxylic acids, amines, pesticides, termiticides, insecticides, flame retardants, cooking emissions, cleaning, repellent, deodorant, deodorizer, PBDE, or PAH;

And: indoor or indoors

And: air, gas phase or gas-phase;

And: concentration or concentrations;

And: measured or calculated.

The search yielded 517 results. From among these, 37 articles provided concentration data on gas-phase species measured in homes in the United States. Compounds with Henry's law constants greater than 1 M/atm were considered water-soluble, as recommended by Sander (2015)²⁰. Potential aqueous phase concentrations were calculated from typical indoor air concentrations reported in the literature and Henry's law constants. This provides a "first assessment" of *compounds* that could be important to indoor aqueous chemistry. (Note that the magnitude of the aqueous reservoir depends also on the liquid water volume.)

Although the literature search yielded an extensive list of WSOGs in indoor air, we expect that there are many more water-soluble gases present in indoor air that have not been measured, because advances in measurement methods for oxidized compounds are a recent phenomenon. Thus, we made use of chemical insights from the *outdoor* (atmospheric) chemistry literature (e.g., smog chamber studies) to propose additional types of oxidized compounds that we expect to be present indoors. Knowledge of indoor source emissions, including cooking, microbial activity, and occupant skin off-gassing provided a few additional WSOGs.

Finally, we used knowledge about outdoor aqueous chemistry and indoor spaces to speculate on how indoor aqueous chemistry may affect indoor air composition and therefore affect exposure.

Total water-soluble organic gases measured during this study

Figure 1 compares the concentrations of organic carbon collected concurrently in the mist chamber samples inside and immediately outside the 13 homes. Indoor samples contained 540–1,400 $\mu\text{M-C}$, with mean indoor WSOG concentration of 940 $\mu\text{M-C}$, standard deviation of 300 $\mu\text{M-C}$ and the 95% confidence interval of 770 – 1,100 $\mu\text{M-C}$. In contrast, concurrently-collected samples from residential outdoor air contained only 28 – 110 $\mu\text{M-C}$, with a mean of 69 $\mu\text{M-C}$, standard deviation of 25 $\mu\text{M-C}$, and 95% confidence interval of 56 – 83 $\mu\text{M-C}$.

Indoor concentrations of WSOG were, on average, 15 times higher than outdoor concentrations (Figure 1), suggesting that the vast majority of indoor WSOG is emitted or formed indoors. A lower-bound estimate of the percentage of indoor WSOG that is of indoor origin can be calculated by assuming that all water-soluble organic gases that originated

outdoors penetrate and persist indoors with 100% efficiency (100% penetration, no indoor losses). Using this lower-bound assumption, the mean, median and range of indoor-origin WSOG are 86%, 92%, and 83% to 98%. Thus, the indoor residential environment is substantially enriched in water-soluble organic gases and this WSOG is predominantly emitted or formed indoors.

Comparison of indoor and outdoor organic carbon concurrently collected under identical conditions enables a quantitative comparison of WSOG indoors relative to outdoors. However, to calculate gas phase WSOG concentrations ($\mu\text{g-C}/\text{m}^3$ air) rather than the concentration of WSOG in the collected aqueous samples ($\mu\text{g-C}/\text{L}$ water) would require the average Henry's Law constant for the mixture. In this work, sample collection times were long to increase organic compound concentrations in aqueous samples for more detailed chemical characterization and laboratory experiments. At long collection times, mist chamber water approaches Henry's Law equilibrium with the sampled air. Thus, for long collection times, calculation of WSOG concentrations in $\mu\text{g-C}/\text{m}^3$ requires an estimate of the average Henry's Law constant of the collected WSOG mixture. To provide an example, if the average Henry's Law concentration of the mixture were 3000 M/atm (the same as formaldehyde), the average indoor WSOG concentration would be 350 ppb_v (470 $\mu\text{g}/\text{m}^3$). However, Henry's Law constants vary tremendously. For example, the Henry's Law constant is 4×10^6 M/atm for glyoxal and 20 M/atm for 2-butanone²⁰, both water-soluble gases found in air. Thus since the composition of the WSOG mixture is unknown, the error in such a calculation could potentially be large. Despite this limitation, the knowledge that WSOG concentrations indoors are substantially greater than outdoors, and thus indoor WSOG sources/production is substantial, constitutes a major step forward in understanding indoor WSOG.

Below we use knowledge from the existing literature to infer what water-soluble organic gases are likely to be available to participate in indoor aqueous chemistry.

Water-soluble organic gases in indoor air

Compounds previously measured

Table 1 shows *water-soluble* organic compounds identified in the literature search that have a potential aqueous concentration equal to or greater than 10^{-3} μM , organized by potential aqueous-phase concentrations. Concentrations are the product of the Henry's law constant²⁰ and typical gas-phase concentration.

Compounds predicted to be present

In addition to the water-soluble gases *measured* in air, many more are expected to be present. Some of these compounds may be directly emitted while others will be formed through gas phase reactions. Table 2 provides examples of water-soluble gases measured in emissions from sources typical of indoor environments, specifically from building occupants, cooking, wood burning, and microbial metabolism. We also included volatile disinfection byproducts (DBPs) from drinking water because, if present in the household water supply, they can partition to indoor air as a result of activities such as showering and water boiling. Microbial

VOCs (MVOCs) are released from metabolism of bacteria and fungi; the majority of MVOCs listed are emitted from other indoor sources as well³¹. Although not included here, off gassing from building materials, furniture, tobacco smoke, and consumer products can also produce a wide variety of VOCs including WSOGs^{32–35}. In most cases listed compounds were measured in controlled chamber studies using analytical techniques optimized to particular compound classes (e.g. phenols and syringols using GC-FID³⁶). We expect that as sophisticated methods for measurement of polar gases are applied to indoor air measurement, these and other WSOGs will be increasingly found in indoor spaces.

In addition to WSOG directly released into indoor air, WSOG will also be formed indoors from gas-phase VOC oxidation and oxidation of organic surfaces. Gas-phase oxidation has been observed to form WSOGs on time scales that are competitive with the air exchange rate, for example from d-limonene⁴². Oxidation of surface materials is considered the major formation pathway for WSOGs because surface oxidation can produce indoor WSOGs even when reaction times are long relative to the time-scale for air exchange. Three oxidants are of particular importance to the formation of WSOG indoors: ozone, hydroxyl radical (OH·) and nitrate radical (NO₃·). Ozone infiltrates from outdoors while OH· and NO₃· are produced indoors^{43–45}. Nitrogen pentoxide (N₂O₅) and chlorine radical may also play an important role occasionally, such as in homes with natural gas combustion and ozone (NO₂ + O₃) or cleaning and photolysis, respectively.

Ozone oxidation of unsaturated VOCs has long been recognized as an indoor source of polar organics (e.g., gas phase aldehydes);⁴⁶ the importance of ozonolysis of surface-bound organics to WSOG formation has more recently been recognized^{47,48}. The predominant source of indoor residential ozone is infiltration from outdoors, yielding indoor concentrations are typically 20–70% of those outside⁴⁵. It is not uncommon for indoor ozone concentrations to range from negligible to 40 ppb, depending on outdoor levels, air exchange rate and indoor sinks⁴³. Higher concentrations usually occur midday, during summertime, although high concentrations can also occur in wintertime under certain circumstances⁴⁹. In addition to formation of oxidized organic gases, ozone – VOC reactions can produce indoor OH·.

Indoor hydroxyl and nitrate radical concentrations are not well constrained, but are expected to be substantial at times. Ozone - NO₂ reactions provide a source of nitrate radicals⁴³, particularly when natural gas combustion provides an indoor source of NO₂^{50,51}. Because there is less photolysis indoors, NO₃· can persist longer in indoor spaces than outdoors. NO₃· concentrations on the order of 10⁻³ ppb^{52,54} can occur indoors. When ozone concentrations are elevated (e.g., through outdoor-to-indoor transport), or NO₂ is elevated indoors, OH can be produced through VOC oxidation reactions⁴⁴ or with adequate infiltrated sunlight, through HONO photolysis¹⁰. New evidence suggests that indoor OH can reach concentrations comparable to daytime outdoor concentrations through HONO photolysis¹⁰ (10⁻⁵ to 10⁻⁶ ppb). However, indoor OH concentrations are probably frequently lower than peak daytime outdoor levels. Given that indoor oxidants co-exist with elevated indoor VOCs and organic-rich surface materials, formation of water-soluble organic compounds in indoors spaces is inevitable.

To provide insights into secondary oxidized compounds expected from gas phase or surface oxidation chemistry in indoor air, Table 3 shows compounds identified indoors and their oxidation products gleaned from the atmospheric and indoor chemistry literature. Smog chamber studies designed to better understand atmospheric chemistry and indoor literature examining oxidation products of cleaning agents and human skin lipids provide the bulk of the insights provided in Table 3. Several examples of non-polar VOC precursors and soluble oxidation products are given. However, this list is not exhaustive.

As shown in Table 3, oxidation of organics leads to the formation of water-soluble gases, such as ketones, acids, alcohols, organic nitrates, peroxides, and epoxides. Some of these have been measured indoors and more are likely to be present. These water-soluble organic gases, have the potential to play a key role in aqueous chemistry indoors when liquid water is present.

Aqueous chemistry in outdoor air

Since water-soluble organic gases are plentiful indoors and liquid water is expected to be present in a substantial fraction of homes, it is plausible that aqueous chemistry could alter the concentrations of gas and condensed phase compounds indoors, like it does outdoors. In outdoor air, aqueous chemistry occurs because WSOGs partition into clouds, fog and wet aerosols and react through radical and non-radical reactions⁶⁵. This reactive uptake is a substantial sink for some atmospheric WSOGs⁷. Low-volatility products of atmospheric chemistry in clouds, fog and wet aerosol remain in the particle phase even after water evaporation; thus aqueous organic chemistry is a source of secondary atmospheric organic particulate matter (secondary organic aerosol; SOA; ^{5,6668}). Aqueous chemistry also alters the gas phase composition via volatile products⁶⁵.

There is substantial atmospheric evidence for the importance of aqueous atmospheric chemistry. Perhaps the most well-established and best-known example is aqueous oxidation of SO₂ by H₂O₂. Dramatic co-depletion of SO₂ and H₂O₂ has been documented with cloud on-set, for example at Whiteface Mountain, NY⁶⁹. Aqueous SO₂ oxidation is much more rapid than gas phase oxidation, and forms the majority of particulate sulfate and acid rain⁶⁹⁷¹. Additionally, aqueous processing (heterogeneous hydrolysis of NO₂) is a major source of HONO⁷². Global modeling suggests that SOA formation via OH oxidation of aldehydes and acetic acid in clouds is comparable in magnitude to SOA formation via gas phase chemistry and vapor pressure driven partitioning⁷³, and this chemistry largely explains the global atmospheric loading of oxalate, the most abundant dicarboxylic acid in the atmosphere⁷⁴. Isoprene-derived epoxides and peroxides are known to react in wet acidic sulfate aerosol; the products of these reactions are substantial contributors to organic aerosol in the southeastern U.S.⁷⁵. Likewise, a variety of compelling atmospheric evidence suggests that aqueous uptake can be a major sink for WSOGs, such as formic and acetic acids⁷⁶, formaldehyde and acetaldehyde⁷⁷, and glyoxal^{7,78}.

In the atmosphere, liquid water is found in the form of clouds, fog, and wet aerosols. Interestingly, liquid water is the largest component of fine particulate matter in the atmosphere (PM_{2.5}), globally⁷⁹ and the largest summertime component of PM_{2.5} in the

eastern United States⁸⁰. Additionally, water-soluble gases are ubiquitous and abundant outdoors,^{80,81} mostly due to atmospheric oxidation of VOCs. For example, glyoxal and other aldehydes are formed from the hydroxyl radical oxidation of isoprene and 2-methyl-3-buten-2-ol (MBO) emitted from trees⁸² and from the oxidation of aromatics emitted from fossil fuel combustion⁸³. There are also primary sources of water-soluble gases such as acetone, formaldehyde, acetic acid, and phenols from biomass burning^{84,85}.

Once in the atmosphere, water-soluble gases can partition into liquid water⁵ and hydrate, dissociate, undergo hydrolysis, nucleophilic or other acid-base and catalytic reactions, or be oxidized by ozone, H₂O₂, OH or NO₃ radicals^{66,67,86} dissolved from the gas phase or formed in the aqueous phase (e.g. aqueous OH formation through Fenton reaction, or HONO, nitrate, or peroxide photolysis). Lifetimes with respect to OH are frequently shorter in the aqueous phase than in the gas phase. For example, for OH in equilibrium between the gas phase and aqueous phase (i.e., for [OH] = 10⁶ molecules/cm³ in the gas phase and [OH] = 10⁻¹² M in the aqueous phase), the lifetimes of glyoxal, formaldehyde and phenol are 1.2 days, 1.4 days and 10 hours, respectively, in the gas phase⁸⁷ and 15 min, 40 s, and 3 min., respectively, in the aqueous phase⁸⁸. OH radicals are so reactive, that their uptake or aqueous formation rate controls product formation rates⁸⁹, and concentrations will be lower than equilibrium values unless aqueous OH formation is substantial. In clouds and fogs, where solute concentrations are low, OH radicals dominate daytime aqueous organic chemistry due to their abundance, high reactivity and water solubility^{90,91}. NO₃ radical and Fenton reactions are more important contributors at night^{92,93}. Formaldehyde, for example, partitions into the aqueous-phase (H = 3000 M/atm⁹⁴), hydrates and reacts with OH radical to make formic acid, HO₂ radicals and water^{95,96}. Nucleophilic addition to formaldehyde is also possible; for example, addition of bisulfite leads to the formation of hydroxymethanesulfonate⁹⁷.

Aqueous-phase reaction can form different products than gas-phase reaction. For example, gas-phase glyoxal oxidation produces formaldehyde^{98,99}, whereas aqueous-phase glyoxal oxidation makes glyoxylic and oxalic acids^{100,101}; subsequent neutralization (e.g., by ammonium) can produce low volatility salts (e.g., ammonium oxalate) that remain in the condensed phase (in particles or on surfaces) even after water evaporation^{102,103}. This difference in chemistry occurs because glyoxal is doubly hydrated in the aqueous phase (exists as a tetrol)¹⁰⁰.

Chemistry in wet aerosols differs from that in clouds and fogs because aerosol liquid water is highly concentrated with solutes. Non-radical reactions can compete with radical reactions in wet aerosols because high solute concentrations make up for their lower rate constants¹⁰⁴. For example, acid-catalyzed ring opening and nucleophilic ammonium reaction with isoprene epoxydiols lead to the formation of oligomers and organosulfates in wet atmospheric particles^{105,106}. The process of water evaporation (i.e. in evaporating fogs and clouds) speeds otherwise slow reactions, e.g., between aldehydes and amines,¹⁰⁷ facilitating chemical transformation. Further, photosensitized radical reactions involving humic-like substances that are found in wet aerosols and at air-sea interfaces have been shown to produce oxygenated organic gases¹⁰⁸. Thus, aqueous chemistry can be both a sink

for water-soluble gases and a source of both gases and condensed phase species in airborne particles and on surfaces^{5,67,73}.

The now substantial body of research being conducted to understand *atmospheric* aqueous chemistry provides insights into the potential for aqueous chemistry to alter concentrations and exposures indoors.

The case for aqueous chemistry in indoor air

Surfaces play an important role in indoor chemistry. Indoor surface area-to-volume (S/V) ratios are greater than $3 \text{ m}^2/\text{m}^3$, and much greater when internal surface areas are considered for materials such as carpets and upholstery^{109,110}. This is orders of magnitude greater than the $<0.01 \text{ m}^2/\text{m}^3$ typical of ambient outdoor air. Even a 1 nm water film on indoor surfaces, a film consistent with simple water adsorption, will provide more than 1000 times the volume of liquid water as is found in aerosols in outdoor air (assuming $3 \mu\text{g}/\text{m}^3$ of aerosol water). Combined with the knowledge that WSOGs are present at much higher concentrations indoors than outdoors, the potential for aqueous chemistry to play an important role indoors, as it does outdoors, is substantial.

The measurement of HONO indoors provides clear evidence that there is sufficient liquid water to drive aqueous chemistry in residences, and that aqueous chemistry can alter the composition of indoor air. HONO is produced from the heterogeneous hydrolysis of NO_2 . HONO has been measured in many homes over the last few decades, including the early study of 10 homes by Spengler and Brauer, who found HONO concentrations ranging from 2 to 8 ppb.⁹ Recently, measured indoor OH radical concentrations of $1.8 \times 10^6 \text{ molecules}/\text{m}^3$ were linked to HONO photolysis on windows¹⁰. Interestingly, this suggests that OH radicals can be produced through photolysis in indoor liquid water films on windows. Those OH radicals can then go on to oxidize aldehydes (e.g., glyoxal), organic acids (e.g., acetic, lactic) and phenols (e.g., guaiacol, phenol) in the aqueous phase to produce oxalic acid, oligomers and other products^{111–113}. Aqueous OH oxidation of formaldehyde will be a source of gas phase formic acid⁶⁵. We also expect that ammonia/amines, which have varying water-solubilities and could also be elevated indoors¹¹⁴, will react with aldehydes as water evaporates from indoor surfaces. These reactions will form brown surface films¹¹⁵. Other likely chemistry is described below.

One important knowledge gap pertains to indoor liquid water. The hygroscopicity of indoor surfaces, indoor liquid water quantities and chemical characteristics of aqueous solutions are not well understood. Evidence for the presence of water has been documented in a substantial percentage of buildings^{1,2}, and water can be present indoors for a variety of reasons. Some indoor locations are constantly wet, like toilet bowl and sink traps, but have limited interfacial surface areas for chemical mass transport. Other locations are seasonally wet, such as air conditioning coils (summer) and windows/exterior walls (winter) that are colder than the indoor air and thus condense moisture from that air. Still other locations are subject to regular condensation/ evaporation cycles such as sinks, showers and bathroom mirrors. Liquid water can also be found indoors due to snow melt, rain water, leaks and faulty plumbing³. Building occupants themselves have liquid water in their respiratory tract

and hygroscopic salts on human skin also take up water when the RH exceeds the deliquescence point¹¹⁶. Finally, a wide variety of hygroscopic materials (e.g., clothing, furniture, building materials, aerosols and surface grime) absorb water vapor,^{117–122} creating highly concentrated aqueous solutions where reactions can take place. For example, a room with walls consisting of lime–gypsum interior plaster can reduce the RH by 10%¹²¹. This accounts for a water uptake of 3 mL per square meter of plaster (equivalent to a water layer 3 μm thick, were it all to exist as a surface film). Airborne particles are also hygroscopic, increasing their water content with increasing RH. While the hygroscopicity of indoor-generated particles is not well known, roughly half of indoor fine particle mass is of outdoor origin. Globally, ambient fine particles are 70% water, on average⁷⁹. At particularly high RH, ambient particle mass can increase by a factor of five due to water uptake¹²³. Indoor surfaces are known to accumulate surface films 1–14 nm thick^{124–128}. Although polar compounds have been measured in surface films¹²⁴, the hygroscopicity of these films is not known. Based on aerosol hygroscopicity, it is not unreasonable to think that the film thickness could double or triple at high RH, due to hygroscopic growth, leading to highly concentrated aqueous mixtures.

The extent of WSOG uptake to wet surfaces depends on the volume of liquid water indoors, which is poorly understood, the Henry's law constant, and on subsequent reactions in the aqueous phase. For example, assuming aqueous uptake to reach Henry's Law equilibrium but no subsequent aqueous reaction, 20 ppb_v of formaldehyde (a typical indoor concentration) would decrease by 1% in a 40 m³ room that has a liquid water film 1 nm thick (surface water adsorption only) and 15% if the liquid water film were 30 nm thick. Once taken up, formaldehyde will hydrate and reactions in liquid water will enhance uptake to reestablish Henry's law equilibrium.

Unfortunately, little work has been done to characterize the quantity and the physical and chemical properties of liquid water on indoor surfaces. Many factors may affect the aqueous chemistry that could occur indoors including, but not limited to:

- The hygroscopicity of surface materials. If the hygroscopicity is high (e.g. fabric furniture, unpainted walls, salty skin, infiltrated outdoor particles), then water vapor can be taken up into the material, sometimes even at modest humidities¹¹⁸, forming highly concentrated aqueous solutions¹²⁹
- The surface area and volume of liquid water films. The capacity to remove water-soluble gases depends on the water volume but also on compound removal through aqueous reaction; the (reactive) uptake rate can depend on the water surface area or on the rate of reaction after uptake⁸⁹.
- The features of the aqueous solution such as pH, water activity, presence of salts (which can change Henry's law constants and affect hygroscopicity)¹³⁰, production of oxidants¹³¹, photosensitizers¹³², nucleophiles¹⁰⁵, transition metals¹³³, and other reactants.
- The chemical composition of the surface. Indoor surfaces can become soiled with semi-volatile organic compounds that partition and continuously re-partition between the gas and adsorbed phases, particles released by occupant activities,

and occupant skin flakes¹³⁴. Soiled surfaces can also become oxidized, e.g., by ozone transported from outdoors^{47,48}. The properties of the soiled surfaces will affect hygroscopicity¹³⁵ and aqueous reactivity.

Chemical composition affects surface water uptake. While some atmospheric aerosol constituents (i.e., sulfuric acid) take up water even at 10% RH, others do not deliquesce until high RH (e.g., 85% RH, inorganic salts). Non-polar hydrocarbons are not hygroscopic, but some aerosol organics do retain water even as low as 10% RH^{136,137} (e.g., malonic, citric, tartaric acids). We expect that indoor surface films will be comprised of complex mixtures. Aerosol particles comprised of complex mixtures take up water at lower RH values than single component particles¹³⁸. For example, a particle mixture of ammonium sulfate, ammonium nitrate, levoglucosan, fulvic acid and succinic acid experiences deliquescence behavior at relative humidities from 55 to 70%. Independently, the inorganic salts would not be in solution until an RH of 85%^{86,138}. Also, because of the presence of ammonia and amines indoors and atmospheric aerosol studies of their effect on hygroscopicity, we anticipate that reaction of indoor organic surface films with ammonia and amines will increase water uptake by indoor surfaces^{114,139}. Additionally, as surfaces age, they become oxidized leading to carbonyl, alcohol, and carboxylic acid functional groups.⁴⁷ Methods to predict atmospheric aerosol water uptake as a function of RH and surface composition (oxygen-to-carbon ratio or functional group contributions)¹³⁵ have the potential to predict the hygroscopicity of indoor surfaces, if they are chemically-characterized.

Once partitioned in the aqueous phase, water-soluble compounds could undergo several types of reactions including radical reactions, acid-base, nucleophilic and hydrolysis reactions. We expect that aqueous surface films will contain high concentrations of solutes, and thus the chemistry will be more similar to the chemistry in wet aerosols rather than clouds. Compounds taken up from the gas phase may react with carbonyl, alcohol, and carboxylic acid functional groups present in surface films⁴⁷ or with compounds taken up from the gas phase. Since ammonia and amines are water soluble, they are likely to be taken up from the gas phase and to participate in aqueous chemistry. TiO₂, present in most paints, is a photosensitizer¹⁴⁰ and thus can initiate radical reactions in aqueous solutions in the presence of light, as can humic-like organics^{108,141}. Because solar radiation penetrates through windows, photosensitized reactions could yield volatile products, as has been observed previously for photosensitizers at air-ocean interfaces¹⁰⁸ and could be a major pathway for OH formation in water films. This is especially true when condensation forms on windows. When window condensate evaporates, it will release volatile products to the gas phase^{107,115}. Aqueous photolysis⁹⁰ of HONO¹⁰, HOCl¹⁴², organic peroxides¹³¹ and nitrate are also likely sources of OH in indoor water films. While OH radicals can also be taken up from the gas phase, they are so reactive that diffusion would be the rate limiting step to product formation in the absence of aqueous phase sources. Even though reaction rate constants are much smaller, non-radical reactions are competitive with OH radical reactions in wet aerosols because solute concentrations are so high (e.g., molar). We expect the same to be true in indoor water films. Note also that, unlike gas phase reactions, surface reactions do not have to be faster than air exchange in order to be important, because molecules sorbed to surfaces have much longer residence times indoors.

pH is likely to affect the types of chemical reactions that occur in liquid water. Wet airborne particles are frequently acidic, while thin aqueous films on indoor surfaces may be basic or acidic depending on the composition of the surface and adsorbed gases. For example, indoor carbon dioxide concentrations are quite high due to the presence of occupants; dissolved carbon dioxide will lead to the formation of carbonic acid making the aqueous film acidic. Or ammonia may dominate pH since it can be present in high quantities indoors¹⁴³ leading to more basic conditions. The presence of salts, for example on skin, can also affect uptake and reactions. Compounds that may undergo hydrolysis in liquid water partitioned onto surfaces include phthalates from plasticizers and organophosphates from pesticides¹⁴⁴ as well other compounds that can be easily protonated or deprotonated, such as alcohols and amines. OH radicals ($H=30$ M/atm), ozone ($H=10^{-2}$ M/atm) and peroxides ($H=\text{varies, high}$) will also be taken up into liquid water, and can react with WSOG in the aqueous phase to form volatile and low volatility products.

Some reactions will produce products that remain in the condensed phase (e.g. on surfaces) even after water evaporation. For example, OH radical oxidation of acetic acid and glyoxal may occur in indoor water films. In dilute solution these reactions produce oxalic acid, which remains in the condensed phase as an ammonium salt^{112,145}. In concentrated aqueous solution low volatility oligomers form^{112,145}. Evaporation of water films containing aldehydes and amines or amino acids, also found indoors, enables the formation of low-volatility imidazoles and other nitrogen containing oligomers^{107,115}. Oligomers are also expected from nucleophilic reaction of organic epoxides, e.g. in the presence of ammonium¹⁰⁵. Organic hydroperoxides are very water soluble and very reactive in the condensed phase. If present, we expect that they will react with aldehydes or carbonyl functionalities in indoor surface films to form peroxyhemiacetals¹¹¹.

Chemistry in liquid water indoors may also be a source of volatile compounds that will be released into the gas phase upon formation or undergo additional chemistry before they are released. For example, aqueous OH radical oxidation of phenols emitted from wood combustion forms formic and acetic acids that will evaporate when the surface dries (and oxidized aromatic oligomers that remain in the condensed phase)¹⁴⁶. Formaldehyde oxidation by OH radicals also produces formic acid⁶⁵. The fate of oxalic acid, an aqueous oxidation product of phenol, glyoxal, and acetic acid^{100,112,146}, depends on whether it remains a volatile acid or is neutralized and present as a low volatility salt¹⁰².

Human skin contains hygroscopic salts that take up water as relative humidity increases, therefore building occupants themselves are expected to be a medium for aqueous chemistry. In addition to the types of reactions discussed above, indoors, these salts may participate in displacement reactions such as those observed in wet marine aerosols. Chlorine present in marine aerosol, is displaced by organic acids and is released into the atmosphere as HCl¹⁴⁷. This could happen on wet skin with organic acids such as acetic acid (which has been measured indoors)¹⁴⁷. Damp skin may be a source of reactive halogen gases such as Cl₂ and Br₂ as well as nitrate especially if there is sufficient sunlight indoors¹⁴⁸. These halogens can then drive further gas phase chemistry. Also, there is evidence that NO₂ and HCl forms ClNO and ClNO₂ in the presence of water molecules at air-surface interfaces. These products are highly volatile and reactive and can further drive indoor chemistry¹⁴⁹.

Lung fluid, a saline aqueous solution, is also a probable location for aqueous chemistry of indoor water-soluble gases. Formaldehyde, is known to dissolve in lung fluid and produce reactive oxygenated species¹⁵⁰. Water-soluble organic peroxides and quinones found in atmospheric particles have been shown to produce strong oxidants in synthetic lung fluid^{151,152}. Chemically similar WSOGs may also be expected to do so. However, little is known about the potential health effects of inhalation exposures to most WSOG and how their aqueous products that are released from lung fluid may affect indoor chemistry.

Conclusions and future work

Aqueous chemistry has been found to be an important factor in the atmospheric processing of organic compounds. Because the indoor surface area to volume ratios are so much higher than the outdoor values, even a thin water surface coating would provide more water per unit volume than found outdoors. Since the concentrations of WSOGs and several other potential reactants are also much higher indoors than out, we argue that aqueous chemistry is likely to affect indoor air composition and affect inhalation and dermal exposures in damp homes.

Oxidized, polar, water-soluble organic compounds are poorly characterized because methods for their gas-phase measurement are less developed than measurement methods for non-polar VOCs. Thus, herein we report measurements of total water-soluble organic gases in 13 real homes, and find for the first time that WSOG concentrations are substantially higher indoors than outdoors where photochemistry is known to make WSOG ubiquitous and abundant⁸⁰. Literature-based evidence suggests that indoor WSOG includes carbonyl compounds, carboxylic acids, epoxides, organic peroxides, organic nitrates, amines, and phenols. When liquid water is present as indoor surface films, on skin and in wet particles (i.e., at elevated RH), we expect that WSOG will partition into that water and react further. This chemistry is likely to remove some compounds from the gas phase, while introducing others to the gas phase, thereby altering indoor air composition and inhalation exposure. When reactions occur on damp skin, dermal exposures could also be altered.

While we argue that aqueous chemistry will alter exposures in damp indoor spaces, the following are critically needed to assess of the magnitude of this effect and are poorly understood:

- The composition of indoor WSOG
- The hygroscopicity of indoor surfaces
- The chemical and physical properties of indoor aqueous surface films
- The relative importance of radical and no-radical chemistry and thermodynamic properties of the products

Ultimately we wish to know the degree to which “dampness” alters indoor air composition, inhalation and dermal exposures through indoor aqueous chemistry, and whether this chemistry helps to explain adverse health effects in damp homes.

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Practical Implications

Associations between “dampness” in homes and adverse health effects are only partially explained by mold and mildew. We provide evidence that aqueous chemistry could plausibly alter inhalation and dermal exposures in damp homes. However, key knowledge gaps hamper efforts to understand the magnitude of these effects and whether changes in exposure due to aqueous chemistry could help to explain associations observed between “dampness” and adverse health effects.

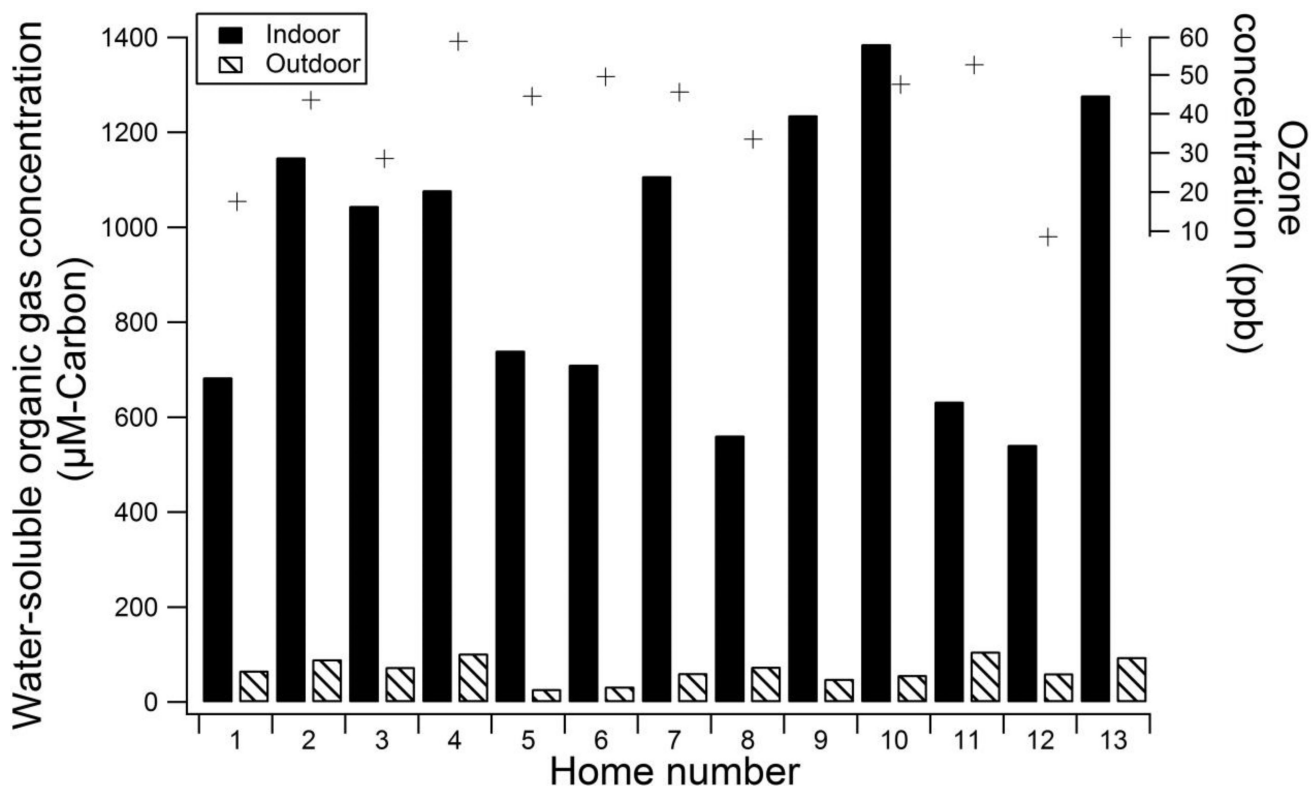




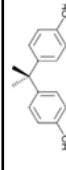
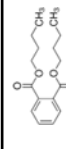










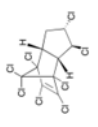







Figure 1.

Total organic carbon ($\mu\text{M-C}$) in aqueous mist chamber samples collected concurrently from particle-filtered ambient air inside (black bars) and directly outside (striped bars) 13 New Jersey and North Carolina homes. Indoor concentrations were significantly higher (paired t-test, $\alpha=0.05$, $p=0.00000018$) than outdoor concentrations. (Source blanks were below the detection limit and field blanks = $7 \pm 2.5 \mu\text{M-C}$. Neither were not subtracted from the sample totals) Nearby outdoor ozone concentrations for each home are plotted directly above each pair of bars. For homes in New Jersey and North Carolina, outdoor ozone concentrations were reported from the New Jersey Department of Environmental Protection Rider University Campus and the North Carolina Department of Environmental Quality Durham Armory ambient air quality sites respectively (ozone was not available for home 9). Indoor relative humidity ranged from 53 to 67%, indoor temperatures ranged from 20 to 26 °C, and the absolute difference between indoor and outdoor temperatures ranged from 0.5 to 8 °C.

Table 1

Water-soluble organic gases measured inside homes in the United States

Compound	Structures	Henry's law constant (M/atm)	Mean (and range) gas phase concentrations (ppb)	Mean (and range) potential aqueous concentration (μ M)	References
Glyoxal		4,000,000	1 (5 – 1.8)	4,300 (1,900 – 7,500)	21
Formaldehyde		3,000	22 (4 – 100)	66 (10 – 300)	14,15,22-25
Methylglyoxal		32,000	0.9 (0.4 – 1.6)	30 (10 – 50)	14
Trichloroacetic acid (TCA)		74,000	0.07 (NA)	5.2	26
Bisphenol A (BPA)		25,000,000	0.00015 (BDL – 0.02)	3.8 (500)	27,28
Dibutyl phthalate (DBP)		1,000	(0.018 – 0.11)	(1.3 – 8.1)	27
Acetone		31	16 (0.4 – 280)	0.5(0.01 – 9)	14-16,22-24,29
Acetaldehyde		15	7.4 (0.7 – 23)	0.11 (0.01 – 0.3)	14-16,21,23,24
2-Butanone (methyl-ethyl-ketone)		18	2 (BDL – 8.4)	0.035 (0.15)	15,23,26,30
Propionaldehyde (propanal)		13	1 (BDL – 7.3)	0.012 (0.1)	14-16,23

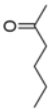
Compound	Structures	Henry's law constant (M/atm)	Mean (and range) gas phase concentrations (ppb)	Mean (and range) potential aqueous concentration (μM)	References
Crotonaldehyde		60	0.15 (BDL – 1.1)	0.012 (0.07)	14
Benzaldehyde		20	0.5 (BDL – 1.4)	0.010 (0.03)	14–16,23,24
Valeraldehyde (pentanal, pentanaldehyde)		6.1	1 (BDL – 23)	0.007 (0.14)	15,16,23,24
Hexaldehyde (hexanal)		5.1	1.2 (BDL – 8.5)	0.006 (0.0078)	14–16,23,24
Chlordane		18	0.26 (BDL – 5.7)	0.0047 (0.1)	25,27
Tetrahydrofuran		10	0.44 (NA – 83)	0.0044 (0.84)	26
Butyraldehyde (butanal)		6.5	0.66 (BDL – 4.7)	0.0044 (0.31)	15,16,23,24
Nonylaldehyde (nonanal)		2	2 (NA – 4.9)	0.0044 (0.01)	23
Acrolein (propenal)		10	0.25 (BDL – 2.4)	0.0026 (0.024)	14
Methyl isobutyl ketone, 3-methyl 2-pentanone (MIBK)		7.4	0.3 (BDL – 81)	0.0024 (0.6)	26,30
Octylaldehyde (octanal)		2	0.9 (NA – 2.4)	0.0018 (0.0044)	23
Heptaldehyde (heptanal)		4	0.4 (NA – 1.1)	0.0017 (0.01)	23

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Compound	Structures	Henry's law constant (M/atm)	Mean (and range) gas phase concentrations (ppb)	Mean (and range) potential aqueous concentration (μM)	References
2-Hexanone		11	0.13 (NA – 0.8)	0.0014 (0.009)	23

NA = not available, values not reported. BDL = below detection limit

Table 2

Water-soluble organic compounds measured in emissions from indoor sources types. These are expected to be released into indoor air and may then partition into any liquid water present.

Emission source	WSOG emitted	Source
Building occupants	<p>Acids such as:</p> <ul style="list-style-type: none"> • Oxopentanoic acid (H=4,000 M/atm) • Lactic acid (H=12,000 M/atm) • Pyruvic acid (H=310,000 M/atm) <p>Ketones, such as:</p> <ul style="list-style-type: none"> • Acetone (H=31 M/atm) <p>Alcohols, such as:</p> <ul style="list-style-type: none"> • Ethanol (H=200 M/atm) • Propanol (H=150 M/atm) • Phenol (H=1,500 M/atm) <p>Other carbonyls such as:</p> <ul style="list-style-type: none"> • 4- Oxopentanal (H=430 M/atm) 	37,38
Cooking	<p>Aldehydes, such as:</p> <ul style="list-style-type: none"> • Acetaldehyde (H=15 M/atm) • Propanal (H=10 M/atm) • Acrolein (H=10 M/atm) <p>Ketones, such as:</p> <ul style="list-style-type: none"> • 2-hexanone (H=11 M/atm) • 2-heptanone (H=6 M/atm) <p>Alcohols, such as:</p> <ul style="list-style-type: none"> • Pentanol (H=80 M/atm) • Hexanol (H=60 M/atm) • 1-octen-3-ol (H=13 M/atm) 	39
Fireplace	<p>Phenols and substituted phenols, such as:</p> <ul style="list-style-type: none"> • Phenol (H=1,500 M/atm) • <i>o</i>-benzenediol (H=4,600 M/atm) • <i>m</i>- and <i>p</i>-cresol (H=1,200 M/atm) <p>Guaiacol and substituted guaiacols, such as:</p> <ul style="list-style-type: none"> • Guaiacol (H=1,000 M/atm) • Vanillin (H=470,000 M/atm) • Eugenol (H=500 M/atm) <p>Syringols and substituted syringols, such as:</p> <ul style="list-style-type: none"> • Syringol (H=NA, highly oxidized) • 4-methylsyringol (H=NA, highly oxidized) <p>Aliphatic aldehydes, such as:</p> <ul style="list-style-type: none"> • Formaldehyde (H=3,000 M/atm) • Acetaldehyde (H=15 M/atm) 	36

Emission source	WSOG emitted	Source
	<ul style="list-style-type: none"> • Hexanal (H=5.1 M/atm) Aliphatic ketones, such as: <ul style="list-style-type: none"> • Acetone (H=31 M/atm) • Butanone (H=18 M/atm) Olefinic aldehydes, such as: <ul style="list-style-type: none"> • Acrolein (H=10 M/atm) • Crotonaldehyde (H=50 M/atm) Aromatic carbonyls, such as: <ul style="list-style-type: none"> • Benzaldehyde (H=20 M/atm) • <i>m</i>- and <i>p</i>-toluadehyde (H=40 M/atm) Dicarboxyls, such as: <ul style="list-style-type: none"> • Glyoxal (H=4,000,000 M/atm) • Methylglyoxal (H=32,000 M/atm) • Biacetyl (H=50 M/atm) 	
Disinfection byproducts in drinking water	Volatile trihalomethanes such as: <ul style="list-style-type: none"> • Chloroform • Bromodichloromethane • Chrolodibromomethane • Bromoform 	40
	Volatile DBPs: <ul style="list-style-type: none"> • 1,1,1,2-Tetrabromo-2-chloroethane • 1,1,2,2-Tetrabromo-2-chloroethane benzyl chloride 	41
Microbial VOCs	Aldehydes, such as: <ul style="list-style-type: none"> • Formaldehyde (H=3,000 M/atm) • Propanol (H=150 M/atm) • Benzaldehyde (20 M/atm) Acids, such as: <ul style="list-style-type: none"> • Acetic acid (H=4,000 M/atm) • Octanoic acid (H=15 M/atm) Ethers, such as: <ul style="list-style-type: none"> • Anisole (H=3 M/atm) Esters, such as: <ul style="list-style-type: none"> • Methyl acetate (H=10 M/atm) • Ethyl acetate (H=6 M/atm) Ketones, such as: <ul style="list-style-type: none"> • Cyclopentanone (H=80 M/atm) • 3-hydroxy-2-butanone (H= 60M/atm)Acetone (H=25 M/atm) 	31

NA = not available

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Table 3

VOCs identified indoors and water-soluble gas-phase oxidation products expected to be found indoors based on knowledge from atmospheric (smog chamber) and indoor chemistry studies.

VOC	Oxidant	Water-soluble product	Source
C ₅ -C ₈ <i>n</i> -alkanes (from attached garages)	OH·	Hydroxycarbonyls, such as: <ul style="list-style-type: none"> 5-hydroxy-2-pentanone (H=NA, highly oxidized) 4-hydroxypentanal (H=NA, highly oxidized) 	55
Monoterpenes (from cleaning products, wood floors, e.g. α-pinene, β-pinene, limonene)	Ozone	Aldehydes and acids, such as: <ul style="list-style-type: none"> Pinonaldehyde (H=9,000 M/atm) Succinic semialdehyde (H=NA, highly oxidized) Glyoxal (H=4,000,000 M/atm) Pinic acid (H=NA, highly oxidized) 	56,57
	OH·	Aldehydes and ketones, such as: <ul style="list-style-type: none"> Pinonaldehyde (H=9,000 M/atm) Acetone (H=30 M/atm) Formaldehyde (H=3,000 M/atm) Organic nitrates, such as: <ul style="list-style-type: none"> Peroxyacyl nitrates (H=NA, highly oxidized) 	57-59
Alkylbenzenes (building materials, furniture, attached garages)	OH·	Quinones, such as: <ul style="list-style-type: none"> Benzoquinone (H=2 M/atm) Hydroxyl dicarbonyls	60
Squalene (human skin lipid)	Ozone	<ul style="list-style-type: none"> Acetone (H=30 M/atm) Hydroxyacetone (H=7,700 M/atm) 1,4-butanediol (H=800,000 M/atm) Levulinic acid (H=3,900 M/atm) 4-oxobutanoic acid (H=4,900 M/atm) 	48
isoprene (indoor plants, people)	OH·	Carbonyls, such as: <ul style="list-style-type: none"> Acetaldehyde (H=13 M/atm) Acetone (H=30 M/atm) Butanone (H=18 M/atm) Hydroperoxides, such as: <ul style="list-style-type: none"> Isoprene hydroxyl hydroperoxide ISOPOOH (H=NA, highly oxidized) Methylhydroperoxide (H=NA, highly oxidized) Hydroxymethyl hydroperoxide (H=NA, highly oxidized) Epoxides, such as: <ul style="list-style-type: none"> Isoprene epoxide (H=30,000,000 M/atm) 	61-63
	NO ₃ ·	Alkyl nitrates (H is dependent on exact compound, expected to be high) Hydroxycarbonyls (H is dependent on exact compound, expected to be high)	64

NA = not available

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