
Appendix – HOCl in indoor atmospheres

The focus is laid on the creation of radicals and downstream reaction cascades driven by chlorine [Cl•] and hydroxyl [HO•] radicals created by the decay of HOCl. The oxidative power of gaseous HOCl must be understood as the combined action of HOCl itself and its two initial 'descendants', the hydroxyl [HO•] and chlorine [Cl•] radicals.

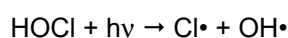
Background: Indoor air quality is influenced by the contents and activities within a building, along with the properties of the building itself and its environmental setting. Good air quality is typically characterized by the lack of pollutants like airborne particles and harmful trace gases. The concentration of any component of indoor air is set by a balance between the rates of production and removal. Primary sources of trace gases and particulate matter (PM) include cooking, cleaning, building materials, and commercial products.

Secondary chemistry can transform primary pollutants, especially when oxidant levels are elevated. Whereas much of outdoor atmospheric chemistry is driven by photo-oxidation processes, indoor environments may be subject to lower light (except near windows in direct sunlight) and a high surface area-to-volume ratio. Thus, the development of organic and aqueous films on indoor surfaces strongly influences indoor air as they mediate multiphase chemical reactions and equilibrium partitioning of semi-volatile components." [117]

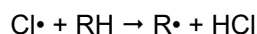
Indoor gas-phase chemistry occurs with ozone (O₃), hydroxyl (HO•) radicals, and other oxidants. O₃ is mainly introduced into the indoor environment from outdoor air or commercial products, while HO• radicals can be chemically produced indoors. Secondary oxidation chemistry removes some compounds (e.g., alkenes), but produces others (e.g., aldehydes, peroxides), and can lead to an increased mass concentration of particulate matter and increased number concentration of ultrafine aerosol. While human breath is a source of biological particulate matter (PM) of relevance for transmission of some diseases, human lungs are also efficient sinks of particles [117]

Vaporized HOCl is a strong oxidizing agent and reacts (deactivates) with many types of pathogens, e.g.: the reaction with coronavirus is happening at the outer layer of the virus, particularly the spike proteins, which are modified and become non-reactive.

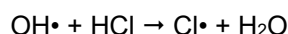
HOCl and its oxidation cascade: Reaction and decay of HOCl: Airborne HOCl decays with rates of 0.3–4%/min [17–91%/h] depending on a couple of factors like surface characteristics, sunlight, temperature, oxygen concentration, humidity, air exchange rates, and others. The main decay route runs along formation of Cl• radicals, which in turn react with volatile or surface laden organic material:



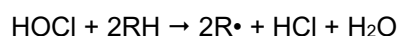
Chlorine radicals produced photolytically may react with and oxidize volatile organic compounds (VOC = RH) and leave a reduced chloride anion to form HCl:



The hydroxyl radical can react with HCl to resubstitute a Cl radical



Unlike OH•, chlorine radicals are not regenerated within the oxidation cycle. However, chlorine radicals can be regenerated by heterogeneous cycling from chloride containing aerosols. The net effect is:



This also reflects the high oxidative potential of airborne HOCl molecules towards volatile organic compounds. In essence HOCl oxidizes airborne or surface laden organic material (e.g.: bacteria, viruses, other pathogens) forming R• components, which in turn react towards higher and usually less reactive oxidized forms. The progressive breakdown of VOC and the oxidation by-products ultimately leads to a reduction in total VOC, thereby improving air quality.

The HOCl reach in indoor environments is very substantial, even at high decay rates of 3%/min (Figure 4).

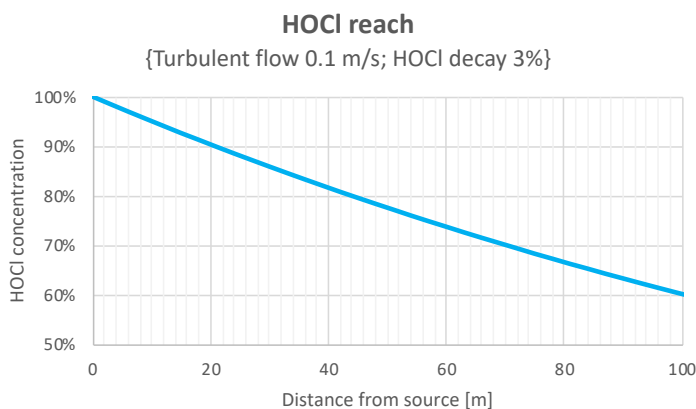


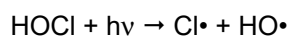
Figure 4: 1st order estimate of room concentration gradient in case of a turbulent flow pattern

Independent studies on the effect of the hydroxyl radical – which is part of the HOCl reaction chain – conducted by Columbia Laboratories demonstrated that the total VOC levels were reduced, and steady state concentrations of oxidation by-products remained within the low ppb range, at or near ambient levels, and did not accumulate[199].

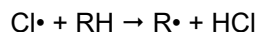
The deactivation of gaseous HOCl in environments which are free of organic components (VOCs and surface laden contaminants) is reduced to basically three processes:

- Photochemical transformation to Cl• radicals
- Catalytic degradation on metal carrying indoor surfaces
- Reaction with water vapor to dissociate into H⁺ and OCl⁻ ions

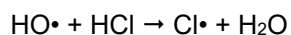
Creation of Hydroxyl (HO•) and Chlorine (Cl•) radicals: The hydroxyl cascade is like the HOCl induced reaction cascade [200], [201]. The difference is that the HOCl molecules are way more stable (they 'decay' [= form radicals]) with 2–3% /min) and therefore reach 'far' points in rooms themselves.



Chlorine radicals produced photolytically may react with and oxidize volatile organic compounds (VOC = RH) and leave a reduced Chlor anion to form HCl:



The hydroxyl radical can react with HCl to resubstitute a Cl radical



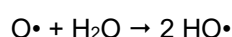
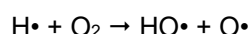
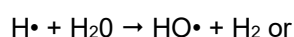
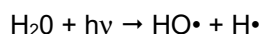
Hydroxyl and chlorine radicals are extremely reactive (high redox potential) and thus extremely short lived and don't accumulate. The indoor reactivity is only based on their ability to initiate a cascade of redox reactions where one radical transmits its redox power in a chain reaction where involved radicals have an ever-decreasing redox potential, but an increasing lifetime. In this way the redox power of the initial hydroxyl and chlorine radicals can 'travel' relatively large indoor distances.

It is important to note that the hydroxyl radical itself belongs to a group of reactive oxygen species (ROS), which in biological systems can cause primary molecular damage, particularly concerning proteins or membrane lipids – because of a lack of specific counter-acting system. When created in indoor atmospheres, it almost immediately transfers its oxidative potential onto air-borne organic components with retain a reduced redox potential and so allow to mediate and safeguard the hydroxyl's original strength [202].

The resulting 'cascade redox power' can disinfect airborne VOCs as well as surface bound pathogens. Unlike the situation with HOCl, the hydroxyl effect cannot be measured by indoor sensors. The necessary amount must be estimated based on lab experiments.

Hydroxyl radicals: There are, on average, 3 to 10 million hydroxyl radicals (HO•) in each cubic centimeter of ambient outdoor air during sunny daylight hours [203], [204], [205]. Even at these relatively low concentrations, hydroxyls are the primary environmental purifying agent [206]. Hydroxyls neutralize most natural and man-made pollutants including isoprene, methane, benzene, toluene, aromatics, hydrogen sulfide, ammonia, and ozone [206], [207]. Hydroxyls decompose volatile organic compounds (VOC) to produce carbon dioxide and water [208]. Indoor environments do not naturally contain hydroxyl radicals as hydroxyls generated outdoors react too rapidly with volatile chemicals to diffuse indoors.

Hydroxyl is created in the atmosphere along the following processes [209]:



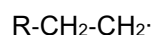
The decomposition of volatile organic and inorganic compounds by hydroxyl radicals involves a complex series of free radical oxidation steps that gradually result in the loss of individual carbon atoms to eventually form carbon dioxide (CO₂).

Hydroxyl ions are extremely short-lived molecules. They usually exist for no longer than a second or so before becoming involved in an air-cleansing chemical reaction. The process is very complex but continues to completion because the reactions involved are highly favored thermodynamically and kinetically. The rates of reactions of hydroxyls with VOCs are very fast. That is precisely the reason why it has been so difficult to accurately measure hydroxyl levels in the earth's atmosphere.

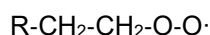
Hydroxyl molecules don't accumulate indoors. They rapidly react with microorganisms and nearly all volatile organic chemicals that exist in air.

Given the speed of reaction, most hydroxyls react inside or near the exhaust of a hydroxyl generating device (e.g.: an instrument from the company PYURE®), while the organic peroxy- and oxy-radicals are more stable than hydroxyls and have time to diffuse out of the device and circulate throughout a treated space.

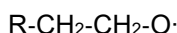
The hydroxyl radical is one of the most powerful oxidants found in nature, surpassed only by atomic fluorine in its oxidation potential. As an efficient oxidant, hydroxyls are driven to regain one electron to restore their very stable valence configuration. The most facile way to accomplish this in an environment rich in VOCs is to abstract a hydrogen atom with its single electron, leaving behind an organic free radical. The newly formed organic free radical has an unpaired electron that is initially localized on the carbon atom:



This moiety rapidly reacts with ambient oxygen to form a peroxy free radical in which the free electron is now localized on an oxygen atom:



This species is a powerful oxidant and will react with VOCs and inorganic gases. It is more stable than the hydroxyl radical and able to travel further away from the reaction chamber to react with VOCs and microorganisms in the treatment area. Peroxy radicals can also rapidly form oxy radicals in ambient air:



Oxy radicals are powerful oxidants and even more stable than their per-oxy counterparts, and therefore can travel even further throughout the treatment space to react with microorganisms and other VOCs.

These types of intermediate oxidants are how ODOROX® systems can penetrate and treat structures and contents at a distance from the device.

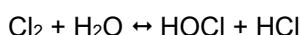
A kind of cascade effect takes hold: An ongoing free radical chain reaction sequence and the formation of a series of intermediate oxidants. The free radical transfer reactions essentially 'unzip' carbon chains rapidly. If hydroxyls are continuously formed, both thermodynamic and kinetic factors favor continued oxidation of VOCs. In the cascade effect, successive generations of oxidants continue to decompose and dissociate VOC carbon chains creating CO₂ along the 'deconstruction' way.

Studies of the Lovelace Respiratory Research Institute (LRRRI) and Columbia Labs, along with a very significant number of field experiences, have confirmed that the hydroxyl radicals and its 'descendant R• radicals' safely and effectively reduce ambient odor and VOC concentrations indoors [199]. In a study of Aerosol Laboratories (Olathe, KS, USA) it has been demonstrated that hydroxyl-initiated R• radical cascade disinfection effectively decontaminated surfaces by 99.99%–99.999% (log factors 4–5). The study included two vegetative bacteria, two viruses, a fungal spore, and a bacterial endospore as simulants for a broader range of pathogenic organisms [210].

Ozone aspects: The production of hydroxyl radicals also creates ozone. This ozone progressively decomposes in water bearing atmospheres partly into hydroxyl radicals, which themselves form complex intermediate functional agents with O₃ [211]:



Formation of HCl: Chlorine gas can disproportionate into hypochlorous acid and hydrogen chloride [212]:



In this reaction the chlorine is disproportionated into its –1 and +1 oxidation state. HOCl becomes a very active oxidation substance. The second chlorine loses all further oxidative (ala disinfecting) capability. The formed HCl is ultimately dissolved in air- or surface-humidity:



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