Hydrogen peroxide emission and fate indoors during non-bleach cleaning: a chamber and modeling study

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# ABSTRACT

Activities such as household cleaning can greatly alter the composition of air in indoor environments. We continuously monitored hydrogen peroxide (H2O2) from household non-bleach surface cleaning in a chamber designed to simulate a residential room. Mixing ratios of up to 610 ppbv gaseous H2O2 were observed following cleaning, orders of magnitude higher than background levels (sub-ppbv). Gaseous H2O2 levels decreased rapid and apparently irreversibly, with removal rate constants (*k*H2O2) 17 – 73 times larger than air change rates (ACR). Increasing the surface area-to-volume ratio within the room caused peak H2O2 mixing ratios to decrease and *k*H2O2 to increase, suggesting that surface uptake dominated H2O2 loss. Volatile organic compound (VOC) levels increased rapidly after cleaning, then decreased with removal rate constants 1.2 – 7.2 times larger than ACR, indicating loss due to surface partitioning or chemical reactions. We predicted photochemical radical production rates and steady-state concentrations in the simulated room using a detailed chemical model for indoor air (the INDCM). Model results suggest that, following cleaning, H2O2 photolysis increased OH concentrations by 10 – 40% to 9.7 × 105 molec cm-3 and hydroperoxy radical (HO2) concentrations by 50 – 70% to 2.3 × 107 molec cm-3 depending on the cleaning method and lighting conditions.

**TOC Art**

A close up of a map

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1. **INTRODUCTION**

Indoor air quality is of increasing concern, especially because most human exposure to air pollution occurs indoors where people typically spend 90% of their time.1 The cleaning of indoor environments is a common activity that aims to improve indoor cleanliness and hygiene, but it introduces a wide variety of chemicals that can impact indoor air quality and adversely affect human health.2-5 Field studies and emission tests have measured greatly increased levels of volatile organic compounds (VOCs) in indoor air following the use of cleaning agents.6-9 Some, such as carbon tetrachloride (CCl4), chloroform (CHCl3), dichloromethane (CH₂Cl₂), and trichloroethylene (C2HCl3) are toxic and regulated by the US federal government as hazardous air pollutants.8-10 Cleaning product constituents can react with oxidants to generate secondary products, many of which are known or anticipated to adversely affect health.11 For example, exposure to acetaldehyde and acrolein poses long-term hazards to the respiratory system,6, 12 and some secondary products from ozone-initiated oxidation of terpenes are suspected carcinogens (e.g, formaldehyde, certain organic nitrates, secondary organic aerosol).6, 13 There is also the possibility that an inhaled mixture of oxidant and the unsaturated reactive constituent of the cleaning product may produce free radicals as the mixture travels down the respiratory tract towards the lungs.14

Despite the large overall research effort devoted to cleaning activities, studies of the emission and chemistry of oxidants and their precursors from cleaning are limited. Recently, high concentrations of hydroxyl radical (OH) have been observed during desk cleaning with limonene-containing cleaner.15 Release of reactive chlorine species (e.g., hypochlorous acid (HOCl), molecular chlorine (Cl2), and nitryl chloride (ClNO2)) were observed during surface cleaning with bleach (solutions containing sodium hypochlorite as the primary cleaning ingredient).16-18 Many of them are photolabile and could generate free chlorine atoms (Cl) indoors that serve as an indoor oxidant.19 Dawe et al made the first real-time measurements of hydrogen chloride (HCl),20 the dominant product of Cl and organic compounds, in a residence and observed similar trends as other reactive chlorine species following floor mopping with bleach.

Hydrogen peroxide (H2O2) based cleaners are common bleach alternatives. Aqueous H2O2 is the active ingredient in a number of household non-bleach cleaning products.21, 22 Although it is an important oxidant directly related to the hydroxyl (OH) and hydroperoxyl (HO2) radical budget, and therefore the oxidative capacity of indoor air, the emissions and chemistry from H2O2-based cleaning indoors have not been investigated. Two studies observed 1– 1.9 ppbv of hydroperoxides (H2O2\*, which includes H2O2 and organic peroxides) in simulated or controlled indoor settings in the absence of cleaning.23, 24 Results from these studies suggest that reactions between ozone (O3) and terpenes can be an important source of indoor H2O2 at high indoor O3 levels of 40 – 175 ppbv. Much lower O3 levels (<5 ppb) have been reported in residences,18, 25 however, making this chemistry an unlikely source of H2O2 in residences.

We carried out an intensive cleaning campaign, using multiple cleaning products, in a simulated and controlled residential room in the Indoor Environmental Quality (IEQ) chamber at Syracuse University, referred to as Radicals and Oxidants from Cleaning in a Chamber (ROCC), to investigate the sources and fates of oxidant and oxidant precursors arising from household surface cleaning. In this study, we focus on characterizing the emissions and chemistry of household non-bleach H2O2 cleaning applications. By combining real-time chamber observations and model simulations, we (1) quantify the mixing ratios of H2O2 that arise when non-bleach cleaner is used indoors; (2) investigate the influence of indoor surfaces on H2O2 and VOCs; and (3) study the impact of different indoor light sources on radical formation and indoor chemistry.

1. **MATERIALS AND METHODS**

**2.1 Simulated Residential Room**

Experiments during ROCC were performed in a residential room (29.1 m3) built with typical wood-framed residential wall and ceiling materials inside the IEQ chamber of the Building Energy and Environmental Systems Laboratory (BEESL) at Syracuse University. Room walls and ceiling were finished with gypsum wallboard coated with low-VOC paint (Behr Premium Plus Ultra-Pure White Interior Paint) and the floor was finished with sheet vinyl flooring. No furniture was present in the room; the estimated surface-to-volume (S/V) ratio was 0.62 m-1. The built-room does not have the complexity of a real occupied residential room – it was not occupied and there was generally not much accumulation of human-related chemical compounds, such as skin oil/flakes or cooking emissions. All cleaning experiments were performed under controlled air change rate (ACR) and temperature conditions. Ventilation was mechanically provided. Supply air drawn from outdoors entered the room through a slot diffuser in the center of the ceiling and the exhaust port was located at the bottom of one side of a wall. Two axial fans were operated during experiments to promote mixing of room air. The ACR was kept constant at 0.510 ± 0.004 h-1. Temperature in the room was controlled at 25.7 ± 0.9 ˚C. Relative humidity (RH) in the room was not controlled and averaged 25.8 ± 9.5% during the experimental periods. This is within the RH ranges measured in various indoor environments (13 – 80%) and especially consistent with lower RH indoors in the winter.26-28

Lighting during some experiments was provided by four uncovered fluorescent lights (GE 48’’ workspace bright white F32 T8 tube) approximately 8 ft above the floor to mimic office lighting conditions. A solar illuminator (XE-LUM Large Area Luminaire, Sciencetech Inc., Canada) placed outside of a window of the residential room was used during some of the experiments to simulate indoor sunlight. The illumination area was 0.5 m × 0.5 m at the source and 1 m × 1 m at a distance of 1 m from the illuminator. Wavelength-resolved photon fluxes near the instrumentation inlet for both light sources can be found in Fig. S1 in the supplement. Solar photon flux at the inlet was on the lower end of values measured by windows in real residences but within the range of predicted fluxes considering different window glass transmittance.29 Additional sheets of cardboard (4 ft × 8 ft) painted with the same low-VOC paint used on the interior walls were placed in the room during some experiments to increase the indoor surface area, and hence the S/V ratio.

**2.2 Cleaning Experiments**

For non-bleach cleaning applications, a bottle of commercially available non-bleach H2O2 multi-purpose cleaner spray was used, which contained 0.88% H2O2 by volume. The cleaning solution was applied to a 0.75 m2 section of the vinyl floor as per the manufacturer’s instructions for general purpose use: we sprayed the cleaner 12 times (approximately ~15 mL of cleaner solution in total) to thoroughly wet the floor section and then wiped it dry with clean paper towels twice over a span of 1.5 – 2 minutes. Another form of deep cleaning – “cleaning greasy soil” – was also carried out as per the manufacturer’s instructions: after applying the cleaner to the floor section (12 sprays), the product was left on the floor for more than 1 hour before being wiped clean. Cleaning applications rotated between 10 non-overlapping regions of the floor, and researchers walked on the floor for short durations between applications. The level of soil on the simulated room floor was less than that expected in an occupied room. Soiling may play a role in the observed emission and loss behavior of H2O2 indoors, but this was not investigated in the current work.

**2.3 Instrumentation**

Real-time measurements of gaseous H2O2 were made using cavity ring-down spectroscopy (CRDS; PI2114 Hydrogen Peroxide Analyzer; Picarro, Inc., USA). The CRDS detects trace gaseous H2O2 using a high precision distributed feedback laser. The limit of detection (LOD), determined as 3× the standard deviation of the signal for zero air, was 0.7 ppbv for 30 s averages. Calibration was performed during the campaign. VOCs were detected using a quadrupole proton-transfer-reaction mass spectrometer (PTR-MS; Ionicon Analytik Ges.m.b.H., Austria).30 Isoprene and formaldehyde mixing ratios were calibrated at an RH of 41% and 43%, respectively, while other VOCs were not calibrated. The humidity-dependent instrument response may lead to an underestimation of formaldehyde levels (on the order of ~45%) in the room,31 but this uncertainty does not affect the study results, which rely on relative changes in VOC mixing ratios. Identification of VOC compounds followed the library provided by Pagonis et al.32 An ultraviolet (UV) photometric analyzer (Serinus 10, Ecotech, USA) measured ozone levels. Two chemiluminescence analyzers (Serinus 40, Ecotech, USA) were deployed and provided quantitative measurements of nitrogen oxides (NOx: NO and NO2), and nitrous acid (HONO) following the difference method described in Zhou et al.33 All instruments were situated outside the chamber, except for the meter that monitors room temperature, RH, and CO2 mixing ratios (IAQ-CALC Indoor Air Quality Meter 7545, TSI Incorporated, USA), which was situated inside the room.

Air was continuously drawn through a 5.8-meter-long 3/8’’ Teflon tube at a constant flow rate of ~4.45 L/min from the inlet at the center of the residential room (~1.5 m above the floor). For headspace analysis of the cleaning solution, a set volume of 20 mL of the cleaner was poured into a dry glass beaker that was cleaned by washing with detergent followed by tap water and deionized water. The headspace of the beaker was sampled by placing the inlet in the beaker above the solution. Spectral irradiance was measured inside the room using an Ocean Optics USB4000 spectrometer coupled to a 1 m fiber optic cable and a cosine corrector, and these values were converted to photon fluxes as described previously.34 The composition of the cleaning solution was analyzed using two dimensional 1H-13C heteronuclear multiple bond correlation nuclear magnetic resonance (NMR) spectroscopy.

**2.4 Indoor chemistry model**

The model used in this work is the INDCM (INdoor Detailed Chemical Model), a near explicit zero dimensional photochemical box model constructed based on a comprehensive chemical mechanism (the Master Chemical Mechanism, MCM v3.2, <http://mcm.leeds.ac.uk/MCM/>).35, 36 This mechanism includes 20,000 reactions and 5,000 species and represents the gas-phase degradation of ~143 VOCs.37-40 The degradation of each VOC is initiated by reaction with species such as NO3, OH, Cl, and O3, and photolysis where relevant. The process continues until H2O and CO2 are formed as the final oxidation products. The INDCM also includes terms that consider change with outdoors, internal emissions, photolysis, and deposition to surfaces.

Total photolysis rate is represented in the INDCM as the sum of the contribution from attenuated outdoor sunlight and indoor artificial lighting.16, 35 According to the method developed by Nazaroff and Cass,41 indoor light sources in the model are assumed to have constant transmission between 300 ‒ 400 nm (UV) and 400 ‒ 700 nm (visible). This method was used for all 37 species in the model that undergo photolysis, except for the species likely to have the greatest impact on photochemistry; photolysis coefficients of H2O2, NO2, HONO, NO3, O3 and HCHO were determined using the spectrally resolved irradiance measured at the inlet during ROCC (Table S1).

**3. RESULTS AND DISCUSSIONS**

**3.1 H2O2 direct emissions**

Background H2O2 levels in the simulated residential room during unperturbed periods prior to cleaning applications were on average (± SD) 0.64 (± 0.43) ppbv, below the detection limit (64% of data were below LOD). This is comparable to H2O2 mixing ratios (<1 ppbv) observed in urban ambient outdoor atmospheres,42, 43 but higher than those in an unperturbed office and a simulated indoor environment (<0.26 ppbv) using offline sampling and various analytical methods.23, 24 To provide context, average H2O2 mixing ratios measured in a student office and an engineering laboratory during ROCC using the same method were 1.6 (± 1.1) and 2.1 (± 0.49) ppbv, respectively. These higher levels may be due to H2O2 formation from ozone-alkene reactions, which lead to H2O2 formation via reactions of stabilized Criegee intermediates with water molecules.44 Increased H2O2\* levels of 1 – 1.9 ppbv were reported under conditions with simultaneously elevated ozone (40 – 175 ppbv) and terpene (124 – 360 ppbv) levels.23, 24 We did not quantify alkene levels in the office and laboratory during ROCC, but O3 levels in the office and lab were higher than those in the simulated residential room (~20 vs. 2.5 ppbv) due to higher ACR (~3.8 vs. 0.5 h-1), supporting this hypothesis.



**Figure 1.** H2O2 mixing ratios measured during a) four sequential regular cleaning applications and b) one deep cleaning application. Shaded regions indicate application periods.

Greatly elevated H2O2 mixing ratios were observed in the simulated room during each of the consecutive cleaning applications (Fig. 1). Within 2 minutes following regular cleaning, H2O2 levels increased rapidly, and peaked within 42 (± 16) seconds (Fig. 1a). This increase is likely due to emission of H2O2 from the floor of the room. All regular cleaning applications (N = 43) showed similar temporal changes in H2O2 mixing ratios significantly above the background. Peak H2O2 mixing ratios ranged from 80 to 480 ppbv, which are at least 2 – 3 orders of magnitude higher than background levels. The deep cleaning applications (N = 3), in which the cleaning solution was left on the surface for more than an hour, showed a much slower growth profile after application (Fig. 1b) and reached peak H2O2 levels higher than those of the regular cleaning applications (570 – 608 ppbv). This could be due to the prolonged presence of liquid water on the chamber floor following deep cleaning events. While we are not aware of published air-vinyl partitioning coefficients of H2O2, the number of H2O2 molecules that can be accommodated within a 3-dimensional volume of liquid water is likely much larger than the number that can be accommodated at a 2-dimensional surface. Our model sensitivity tests for both regular and deep cleaning suggest that a direct emission of ~5% of the H2O2 applied to the floor successfully reproduced the H2O2 profiles observed (Fig. S2).

For all cleaning events, there was a clear exponential decay of H2O2 mixing ratios after the peak level was reached (Fig. 1). The estimated first-order decay/removal rate constant of H2O2 (kH2O2) following exponential fitting of the decay data spanned a large range between 8.5 and 37.1 h-1 for regular cleaning applications, which is 17 – 73 times the ACR in the room. This indicates important removal pathways other than ventilation. The removal rate constants of H2O2 were similar to those of chlorinated compounds (e.g., HOCl, Cl2, ClNO2, Cl2O, NHCl2) observed in a residence with the same ACH of 0.5 h-1 after bleach cleaning events (~13 – 150 h-1),17 and significantly larger than those of NO, NO2, and HONO observed in a residence after cooking (0.55 – 2.23 h-1 with ACH of ~0.6 h-1).33 The deep cleaning application yielded a much lower decay rate constant of ~1.8 h-1, possibly due to the prolonged residence of the liquid coating on the floor resulting in H2O2 off-gassing to the gas phase for an extended period after cleaning. Overall, elevated H2O2 levels during deep cleaning persisted for a longer time; the integrated H2O2 mixing ratio over 2 hours after deep cleaning were 20 – 130 times that after regular cleaning.

**3.2 Surface uptake of H2O2**

Deposition of gaseous H2O2 onto indoor surfaces may be an important process that impacts its airborne levels. In ambient outdoor air, dry deposition is one of the major atmospheric fates of H2O2, accounting for 24 – 73% of the total H2O2 loss.45 Indoors, the surface-to-volume ratio can be 3 ‒ 4 orders of magnitude greater than that outdoors, as interior surface area is dominated by constructed materials such as walls and furniture, which can comprise ~105 more surface area than indoor particulate matter.17, 46 To investigate surface deposition of H2O2 in the room, we varied the amount of painted surface available under three conditions (low, medium, and high) that correspond to calculated S/V ratios of 2.0, 6.9, and 11.8 m-1, respectively, andperformed regular cleaning experiments. The S/V ratio under the low surface area condition (i.e., 2.0 m-1 for the empty room) is consistent with literature values when the surface area of the contents in a room are not accounted for.46 In previous studies of rooms in actual residences accounting for furnishings at cm level resolutions, the S/V ratio ranged from 2.0 to 6.8 m-1.46-48 These values are represented by the low and medium surface area conditions in this study. As most existing studies focused on visible surfaces, contents inside drawers, closets, and cabinets including the vast surface areas of indoor textiles, unseen surfaces in building interstitial spaces (e.g., wall cavities), buffer zones (e.g., attic), and HVAC systems were not accounted for.19 The “real” surface area in residences are likely often much greater, with S/V ratios larger than 6.8 m-1, and are represented by the high surface area condition in this study.



**Figure 2.** (a) Peak H2O2 mixing ratios and (b) H2O2 exponential decay rate constants observed during regular cleaning events under three surface area-to-volume (S/V) conditions. Data for individual cleaning events are in light markers. Error bars of light markers in (b) were coefficient errors from the exponential regression. The box and whisker plots show the median (line), mean (dark marker), upper and lower quartiles (box), and 10th and 90th percentiles (whiskers). Data fitting between mean peak levels / *k*H2O2 and S/V ratio was performed using the weighted orthogonal distance.

Figure 2a shows the summary data for peak H2O2 mixing ratios during all regular cleaning events. A strong decreasing trend was observed between peak H2O2 mixing ratios and S/V ratio. The mean peak H2O2 level decreased by 40% from 343 (± 75) ppbv under the low surface area condition to 210 (± 80) ppbv under the high surface area condition, demonstrating significant loss of H2O2 to surfaces. This is further supported by the observation of a strong increasing trend of H2O2 removal rate constant *k*H2O2 with S/V ratio (Fig. 2b). The removal rate of H2O2 increased by 2.8-fold from 10.2 (± 1.2) h-1 under the low surface condition to 28.9 (± 7.1) h-1 under the high surface area condition. These observations confirm that surface uptake is an important sink for indoor H2O2 and that indoor surfaces significantly impact both indoor H2O2 mixing ratios and residence time. The apparent linear fits to the data in both panels is consistent with surface uptake, as the number of available surface sites is expected to scale linearly with surface area (and also with S/V, since volume in the chamber is constant). The pseudo-first order uptake rate constant we report is given by *kobs* = *kadsNs*, where *kads* is the 2nd order adsorption rate constant and Ns is the number of available surface sites. With that said, we acknowledge that these lines are fit to only 3 data points; more thorough investigations of the effects of surface area on H2O2 loss kinetics are needed to draw strong conclusions from this apparent relationship.

Photolysis accounts for 13 – 39% of the total loss of H2O2 in the ambient outdoor atmosphere.45 In this study, we did not observe apparent differences in peak concentrations or removal rate constants of H2O2 under different lighting conditions (dark vs. sunlight vs. fluorescent light; Fig. S3). Our photochemical model results also showed similar H2O2 concentrations under different lighting conditions (Fig. S2). These results suggest that photolysis had a negligible impact on indoor H2O2 levels, which is expected given the slow photolysis rates expected indoors.34 Loss via reaction with OH is another important sink of atmospheric boundary layer H2O2 in outdoor air. Based on literature values of the rate constant for this mechanism (~10-6 s-1),45 we estimated that reaction with OH may account for up to 0.3% of the total loss of H2O2 in the room. One unique removal mechanism for indoor H2O2 relative to outdoors is ventilation, as the room is continuously exchanging air. We estimated that the 0.5 h-1 ventilation accounted for less than 6% of H2O2 loss in this room under all experimental conditions. Under all lighting and surface conditions, the dominant sink for H2O2 in the room is deposition to indoor surfaces, which accounts for at least ~94% of total loss.

To investigate whether surface uptake of H2O2 was dominated by physical or chemical processes, we conducted perturbation experiments by flushing the room with outdoor air at elevated ventilation (EV; ACR of ~1.3 h-1) following cleaning applications after H2O2 mixing ratios decreased to a few ppbv (Fig. S4). In similar perturbation experiments in a residence, many indoor gas-phase species (e.g., VOCs, NH3, and HONO) showed decreased levels during EV periods and recovered to steady-state concentrations after EV with a characteristic response time, illustrating that indoor surfaces provide an important transient reservoir of these indoor species.49 In this study, H2O2 mixing ratios decreased during EV but no temporary increase in H2O2 was observed after EV (Fig. S4), which would be expected if H2O2 was physically partitioned to the surfaces. This suggests that the rapid surface uptake of H2O2 is irreversible and that heterogenous reactions are occurring on indoor surfaces, which potentially include both surface films, indoor building materials, and furnishings. Hydrogen peroxide is a known active ingredient in many paint and coating removers and a bleaching agent for wood and paper; it may react with some components of the paint and wood in the simulated room. Studies have also shown that H2O2 gas can undergo physical and reactive uptake to titanium dioxide (TiO2),50-52 which is an important component of the white pigment in the paint used in this study.

**3.3 VOCs and surface partitioning**

Twenty-one mass to charge ratios (*m/z)* were monitored in real time for some of the regular cleaning events under the low S/V condition. They all showed elevated signals during cleaning events, suggesting that in addition to H2O2, surface cleaning introduced many VOCs into the indoor air. The largest increase due to cleaning was observed at *m/z* 33 (methanol) followed by *m/z* 59 (which likely reflects some combination of glyoxal, propanal, and acetone, which are isobaric species with the mass-resolution of the PTR-MS). Five organic compounds – methanol, acetone, acetic acid (*m/z* 61), butene (*m/z* 57), and pentene (*m/z* 71) – that were detected in the gas phase were also detected in the cleaning solution by NMR analysis (Figs. S5 and S6), suggesting that these VOCs were from direct emissions. Some VOCs that were detected in the headspace of the cleaning solution by PTR-MS were not detected in solution by NMR. Some of these species (e.g., formaldehyde (*m/z* 31), acetaldeyde *(m/z* 45), ethanol (*m/z* 47), methyl glyoxal and butanal (*m/z* 73), and pentanediol (*m/z* 105)) are not NMR-active; these likely are present in the cleaning solution. Others, including methyl chloride (*m/z* 51), furan and/or isoprene (*m/z* 69), pentanenitrile (*m/z* 84), and monoterpenes (*m/z* 137) are NMR active. It is possible that these species were formed in the gas phase above the solution or that they were present in solution, but at concentrations too low to be detected by NMR.



**Figure 3.** Time series of H2O2 and select VOCs (*m/z*) during a regular cleaning event under the low surface area condition on November 9, 2018. H2O2, isoprene, and formaldehyde are in units of ppbv; the remaining PTR-MS ions are uncalibrated and provided in arbitrary units (au). Door-open and cleaning application periods are indicated by grey and pink shaded regions, respectively.

Different VOCs showed different temporal trends after cleaning (Fig. 3). For example, methanol levels, as well as isoprene, methyl chloride, and *m/z* 47 (formic acid and/or ethanol), increased immediately after the cleaning application and peaked within a few minutes, while *m/z* 57 (acrolein and/or butene) and *m/z* 59 (glyoxal/propanal/acetone) showed a slower growth profile and peaked after ~20 minutes. Formaldehyde (*m/z* 31) and acetaldehyde (*m/z* 45) levels, on the other hand, decreased initially due to mixing of laboratory air via opening the door and then gradually recovered to pre-cleaning levels. Most observed VOCs deviated from the ideal temporal pattern that would be expected if the compound was volatilized instantaneously when the product was used. The different growth profiles observed between individual VOC were likely linked to their physical properties such as air-surface partitioning coefficients and Henry’s law constants.53, 54 Multiphase chemistry could also occur on surfaces and contribute to different VOC emission rates.

First-order decay rate constants of nine ions that showed clear decays during the sampling period were calculated (Table S2). The mean decay rate constant for an individual ion ranged from 0.58 to 3.6 h-1, all significantly lower than kH2O2 but higher than the ACR. Ventilation accounted for a varying fraction of total removal from indoor air, spanning 14 – 86%, necessitating other loss mechanisms to explain the observations. Modeled decay rates of five VOC species generally agree with the observations (Table S2), suggesting that in addition to direct emissions and removal by ventilation, chemical reactions (potentially gas-phase and surface / multiphase) occurred in the room, resulting in the loss of VOCs introduced by cleaning.

**3.4 Effect of indoor light and cleaning on predicted radical concentrations**

In the absence of cleaning events, the most important oxidants indoors are thought to be O3 and OH. Ozone is primarily introduced via infiltration of outdoor air, and is only expected to be an important oxidant when ACR are high, such as in residential buildings with open windows and in non-residential buildings.25, 55 Hydroxyl radicals are generated via O3-alkene reactions and via HONO photolysis. Ozone-alkene reactions are only important when O3 levels are high, as discussed above, and HONO photolysis is only important under illuminated conditions.19 Formaldehyde photolysis has also been predicted to form HO2, and organic peroxy radicals (RO2) can be formed from reactions of OH and RO2 with VOCs.34 Photolysis of H2O2 following the use of H2O2-based cleaners may provide an additional source of indoor radicals. Figure 4 shows the modeled concentrations of OH, HO2, and organic peroxy radicals (RO2) during the regular cleaning experiment on November 9, 2018, under different lighting conditions. Average ozone (2.5 (± 0.9) ppbv), NO (0.9 (± 0.6) ppbv), NO2 (5.7 (± 0.8) ppbv), and HONO (4.0 (± 2.1) ppbv) mixing ratios observed in the room during the cleaning events were used as input for model simulations. H2O2 mixing ratios reached 312.6 ppbv during this regular cleaning episode (Fig. 2).



**Figure 4.** Predicted radical concentrations under different lighting conditions during regular cleaning under the low surface condition on November 9, 2018.

Model results showed that under unperturbed dark background conditions the average OH concentration was 1.7 × 105 molec cm-3. This value is lower than the typical outdoor midday OH levels in urban environments (1.2 ‒ 20 × 106 molec cm-3), but similar to those oberved outdoors at night (1.8 ‒ 2.6 × 105 molec cm-3).56 Background HO2 concentration in the dark was 3.8 × 106 molec cm-3, much lower than outdoor levels previously reported (0.16 – 10 × 108 molec cm-3),56 and a similar RO2 concentration was predicted in the dark background period (4.0 × 106 molec cm-3). Dark reactions between O3 and alkenes coming from outdoors in the supply air and emitted from building materials in the room were the dominant HOx source. Under non-cleaning conditions with attenuated sunlight, OH was formed from HONO photolysis, resulting in an increase of background OH by 4-fold to 8.8 × 105 molec cm-3 compared to that in the dark, which is in general agreement with previous predictions and observations of indoor OH concentrations under sunlit conditions in the absence of indoor activities (1.7 – 24 × 105 molec cm-3).35, 57-59 Photolysis initiated by indoor sunlight also led to increases of background HO2 and RO2 concentrations by 3 and 2.4-fold, respectively. This is because of photolysis of HCHO to produce HO2 and additional OH present due to HONO photolysis reacting with VOCs to form RO2 radicals. Radical concentrations were about 1.3 – 2.1 times higher under illumination by sunlight compared to illumination by fluorescent tubes. Photolysis of H2O2 was negligible under non-cleaning conditions, due to small absorption cross sections at relevant wavelengths and low mixing ratios (< 0.7 ppb).45

Indoor OH concentrations have been predicted to increase to levels comparable to typical outdoor levels during cleaning events.5, 15-17, 20 Elevated levels of OH (~4 – 17 × 106 molec cm-3) andHO2 (6 – 60 × 107 molec cm-3) were reported in two mechanically ventilated offices following surface cleaning using limonene-containing non-bleach cleaners, although these measurements were performed under high indoor ozone levels (20 – 180 ppbv).15, 60 Mopping with bleach solution has been predicted to increase indoor OH concentrations up to 2.0 × 106 molec cm-3.16 Our model results suggest that in the absence of light, the use of commercial H2O2-based cleaners will not strongly affect the radical budget (Fig. 4); OH and RO2 concentrations remained similar after cleaning and HO2 levels increased by only 25%. We do predict higher radical levels following cleaning events under illuminated conditions due to H2O2 photolysis; the low absorption cross sections at wavelengths longer than 330 nm were offset by high concentrations following cleaning events. We predicted that regular cleaning increased OH and RO2 concentrations by 10% under sunlit conditions (to 9.7 × 105 molec cm-3 and 1.5 × 107 molec cm-3, respectively), and increased HO2 concentrations by 50% (to 2.3 × 107 molec cm-3). Under fluorescent lighting, we predicted increases of 41, 77, and 32% for OH, HO2, and RO2, respectively, after regular cleaning. While the relative increases in radical concentrations were larger under illumination from fluorescent tubes than from indoor sunlight, the peak radical concentrations were lower (by ~50%) due to slower HONO photolysis. The predicted radical levels are expected only in illuminated volumes in the room, so room-averaged radical levels will generally be lower. Radical levels within illuminated volumes may often be higher, as the photon fluxes in the simulated room and in the model are representative of low photon fluxes in early morning or late evening.

Predicted peak radical concentrations following deep cleaning were generally higher than those following regular cleaning (by up to 45%). In addition, elevated radical concentrations under illuminated conditions persisted for ~2 hours following deep cleaning, compared to 25 minutes following regular cleaning (Fig. S7).

The model predictions discussed above assumed that background VOC levels were maintained during cleaning. To determine whether the VOCs released during cleaning affected radical levels, sensitivity tests introducing a “pulse” of a few VOCs in the model during the regular cleaning event (Fig. S8) were performed. These runs generally showed similar radical profiles as those without the VOC pulses. Cleaning in the dark or under indoor sunlight with VOC emissions resulted in a slight decrease (by 7% and 10%, respectively) in the OH radical concentration (Fig. S8). Under fluorescent lighting, peak OH radical concentrations increased by 27% after cleaning when VOC emissions during cleaning were included, which is lower than the 41% increase predicted in the absence of a VOC pulse (Figs. 4 and S8). Peak HO2 and RO2 radical concentrations, on the other hand, increased by greater amounts following cleaning under all lighting conditions when VOC emissions were introduced (Figs. 4 and S8). We predicted increases ranging from 60 to 114% for HO2 and 31 to 74% (depending on illumination condition) for RO2 following cleaning when VOC pulses were introduced (Fig. S9). The lower OH levels and higher HO2 and RO2 levels predicted when VOC emissions during cleaning are included are consistent with OH reacting with VOCs and generating HO2 and RO2.

1. **ENVIRONMENTAL IMPLICATIONS**

Hydrogen peroxide is commonly used in residential and non-residential buildings as a cleaning agent and disinfectant. This combined chamber and modeling study provides information about expected H2O2 and oxidant levels following cleaning under conditions relevant to a closed room in a house. While the H2O2 levels we measured were always below the long-term (8 h) and short-term (15 min) occupational exposure limits of 1 and 2 ppm, respectively,61 we speculate that higher concentrations might be observed when larger surface areas are cleaned, or when more cleaner is applied. Cleaning solutions with higher H2O2 content could also led to higher indoor H2O2 levels. Additionally, we note that the measured mixing ratios reflect room average H2O2 levels, as the sampling inlet was placed at the center of the room. We speculate that H2O2 levels near the application surface (floor) might be much higher. This could imply that although H2O2-based cleaning products are likely safe during household cleaning, people who apply the cleaning agent could possibly be exposed to episodic exceedances of safe H2O2 levels during the application of the cleaner. Exposure may be lower when ACR is higher (for example when windows are opened). The potential health hazard of H2O2-based cleaner, especially for people such as janitorial or housekeeping staff who use these products regularly, warrants further investigation.

Indoor radical levels are expected to be dominated by HONO photolysis under sunlit conditions and by ozone-alkene reactions under non-sunlit conditions even during and following the use of H2O2-based cleaners, but we predict that H2O2 use will increase indoor radical levels; we predicted enhancements in indoor OH, HO2, and RO2 levels following cleaning events ranging from 10 – 77% depending on the radical, type of cleaning, and illumination source. Higher radical levels could be generated if H2O2 levels are higher (as discussed above), or if photon fluxes are larger; integrated UV photon fluxes in sunlit rooms previously reported were 1.04 – 5.67 times larger than those measured near the sample inlet and used in the INDCM calculations.29 This work demonstrates that the use of H2O2-based cleaners can temporarily alter indoor composition greatly. Further work is required to quantify the budget, chemistry, and influence of H2O2 on air composition in real indoor environments and to determine what consequences the use of H2O2-based cleaners may have to indoor air quality and human health.

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**Supporting Information**

Photolysis rate constants of key species in the model; VOC decay rates during the regular cleaning event; Spectral photon flux of two light sources in the room; Modeled and observed H2O2 time series; decay rate constants of H2O2; H2O2 time series during elevated ventilation periods; NMR results; predicted radical concentrations during the deep cleaning event and the regular cleaning including VOC emissions. This information is available free of charge via the Internet at http://pubs.acs.org.

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