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1 **Indoor Air Quality Implications of Germicidal 222 nm Light**

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52 Germicidal ultraviolet light at 222 nm (GUV222) can inactivate airborne pathogens, but has an

- 53 unknown effect on indoor air quality. This study shows that GUV222 forms ozone and hydroxyl
- radicals, forming oxidized byproducts and fine particulate matter.

TOC Graphic

Introduction

- The COVID-19 pandemic has highlighted the critical need to develop and implement strategies
- to decrease the transmission of airborne pathogens. Approaches include both source control
- (isolation, masking), and remediation (ventilation, air cleaning). One approach that has received
- substantial attention is the use of germicidal ultraviolet (GUV) light, which inactivates airborne
- 63 pathogens. This approach goes back decades,¹ traditionally using 254 nm light from mercury
- lamps. Since light of this wavelength can cause damage to skin and eyes, care must be taken to
- 65 minimize occupants' direct exposure to the GUV light. $2,3$
- A promising new approach to GUV-based air cleaning is the use of KrCl excimer lamps, which
- 67 emit at 222 nm (GUV $_{222}$).⁴ In contrast to 254 nm GUV, GUV $_{222}$ does not penetrate deeply into
- 68 biological materials. Therefore, while GUV_{222} is effective at inactivating airborne viruses and
- 69 bacteria, it is unable to penetrate the outer layer of dead skin cells or the ocular tear layer.⁵ 222
- nm light is hence less likely to reach and damage living human tissues, offering the potential for
- air disinfection throughout an entire, occupied indoor space.

A risk with GUV222-based air cleaning, as with all types of air cleaning that rely on chemical

- 73 and/or photolytic processes, is the potential formation of unwanted secondary byproducts.^{6,7} A
- 74 particular concern with GUV_{222} is the formation of ozone (O_3) , a harmful air pollutant that acts
- 75 as a strong oxidant and can lead to respiratory distress when inhaled.⁸ O₃ is formed by the UV
- photodissociation of oxygen (R1-2)

$$
O_2 + h v_{\lambda < 242 \text{ nm}} \rightarrow O + O \tag{R1}
$$

$$
78 \t\t\t O + O2 + M \rightarrow O3 + M \t\t (R2)
$$

79 Since absorption of UV by O_2 , and hence O_3 production, is strongest at short wavelengths,⁹

- manufacturers of KrCl lamps have added filters to block wavelengths shorter than 222 nm. But
- 81 since O₂ absorbs weakly even at 222 nm (σ = 4.09x10⁻²⁴ cm²⁹), all KrCl lamps have the potential
- 82 to generate ozone, possibly in concentrations higher than is typically found indoors.¹⁰
- Ozone generated indoors, in addition to posing a direct health hazard, can set off a cascade of
- chemical reactions that can also affect indoor air quality. Ozone reacts directly with alkenes,
- present both in the air and on indoor surfaces, forming a range of oxidized volatile organic
- 86 compounds $(OVOCs)^{11,12}$ and secondary organic aerosol (SOA) ,¹³ which may negatively impact
- 87 human health.^{14–17} O₃ chemistry can also lead to the formation of the hydroxyl radical (OH), an
- even stronger oxidant. This occurs both through reactions with alkenes, which are known to form

89 OH (R3),^{11,18} and through O₃ photolysis
$$
(R4-5)^{19}
$$
:

90 Alkene + O₃
$$
\rightarrow
$$
 OH + other products (R3)

91
$$
O_3 + h v_{\lambda \leq 370 \text{ nm}} \to O_2 + O(^1D)
$$
 (R4)

$$
92 \t\t\t O(^{1}D) + H_{2}O \rightarrow 2OH \t\t\t (R5)
$$

- 93 Any increased levels of indoor O_3 from GUV_{222} would likely enhance the importance of these
- reactions, leading to higher levels of indoor OH. This includes O3 photolysis (R4-5), which is the
- main source of OH in the troposphere, but under normal conditions is negligible in indoor
- environments, due to the lack of low-wavelength UV. Any OH radicals formed from R3-5 may
- then oxidize a wide range of organic species and contribute further to the formation of OVOCs
- and SOA.
- GUV222 therefore has the potential to dramatically affect the chemical composition of indoor air,
- and may lead to the formation of chemical species that are hazardous to human health. However,
- 101 the extent and nature of this impact remains quite uncertain, even as GUV_{222} is being deployed in
- 102 indoor spaces.²⁰ Two very recent experimental studies^{21,22} demonstrate O₃ production from
- GUV222, but these do not examine the overall effects on indoor air quality (including the
- production of OH, OVOCs, and SOA) by GUV222. To our knowledge the only published work
- 105 that has is a box-modeling study by Peng et al.²³. That work predicted that 222 nm irradiation
- could lead to elevated levels of O3 and other secondary species relative to non-illuminated
- conditions, especially under low-ventilation conditions.
- Here we describe a series of laboratory experiments aimed at better understanding the effects of
- 222 nm irradiation on indoor air quality. The goal of this work is to gain process-based insight
- into how such irradiation affects the chemical composition of the air; we do not examine the
- 111 effects of GUV₂₂₂ light on pathogens, indoor surfaces, or human health. These experiments,
- which use a flow-through Teflon chamber coupled to a range of real-time analytical instruments,
- explore the effects of several parameters relevant to indoor air processes (VOC level, ventilation,
- 222 nm light intensity, and humidity) on the generation of oxidants and secondary products.
- 115 Results are then used to validate a simple chemical model of GUV222 irradiation of indoor air,
- which in turn is used to examine the interplay between GUV222 and ventilation in controlling the
- levels of ozone and other chemical species in the indoor environment.

Materials and Methods

Experimental Methods

- Experiments are carried out in a 150 L Teflon chamber, outfitted with inlet ports (for
- introduction of clean air and trace species) and outlet ports (for sampling by analytical
- instrumentation). Clean dry air from a zero-air generator (Aadco Model 737) is introduced into
- the chamber either directly, or after passing through a bubbler filled with Milli-Q water. Mass
- flow controllers are used to adjust these two flows to control chamber relative humidity. Dilution
- 125 rates are measured using acetonitrile, an inert dilution tracer $(8.0 \times 10^{-4} 9.7 \times 10^{-4} \text{ s}^{-1}, 2.9 3.5)$
- 126 ACH). Most experiments are conducted at 22 °C and ~25% RH, while "higher RH" experiments
- 127 are carried out at ~45% RH.
- GUV222 light is provided by a single filtered KrCl excimer lamp (Ushio, Care222 B1 Illuminator,
- peak emission at 222 nm), centered directly above the Teflon chamber. Average fluence rate
- 130 within the chamber is \sim 45 μ W/cm², estimated geometrically from the lamp intensity profile
- 131 provided by the manufacturer²⁴ (see Section S1.1). The indirect estimation of the lamp intensity
- is a limitation of this work, but the agreement between modeled and measured ozone production
- (see Results and Discussion, below) indicates that it is reasonably accurate. More quantitative
- 134 estimates of UV fluence rate and its relationship to O₃ generation are available in recent work by
- 135 Peng et al.²² and Link et al.²¹ Most experiments are carried out at the full light intensity. For "low"
- 136 light" experiments, the lamp emission is attenuated by several layerPs of plastic, achieving a
- 137 factor of \sim 5 reduction in intensity (determined by the reduction in the steady-state O₃
- 138 concentration which is assumed to scale linearly with average UV fluence).^{21,22} For the "O₃-
- 139 only" experiments, the light is left off, and O_3 is introduced via a Pen-Ray ozone generator, with
- 140 a steady-state O₃ concentration matching that of the GUV₂₂₂ experiments (\sim 100 ppb). Reaction
- 141 conditions for each experiment are described in detail in Table S1.
- 142 For all VOC oxidation experiments, the chamber is first allowed to reach a steady-state
- 143 concentration of O3, either via 222 nm irradiation or direct addition. This is followed by the
- 144 addition of 5.3 ppb of acetonitrile (the dilution tracer) (C₂H₃N, 99.8%, Sigma Aldrich), 1.2 ppb
- 145 of 1-butan-d9-ol (intended as an OH tracer, but not used here due to the relatively low OH
- levels) (C₄D₉OH, 98%, Cambridge Isotope Laboratories, Inc.), and $120 \pm 11 \,\mu g \text{ m}^3$ of
- 147 ammonium sulfate particles (to act as seed particles for any SOA production) ((NH₄)₂SO₄,
- 148 ≥99%, Sigma Aldrich). Finally, the relevant VOC (10 or 100 ppb) is added to chamber. VOCs
- 149 used in this study include hexanal (C6H12O, 98%, Sigma Aldrich), cyclohexene (C6H10, 99%,
- 150 Sigma Aldrich), and (R)-(+)-limonene (C10H16, 97%, Sigma Aldrich). Reactant addition
- 151 procedures are described in greater detail in Section S1.2. Because the oxidants are already
- 152 present in the chamber, oxidation begins immediately, so VOC injection is taken as $t = 0$.
- 153 Real-time measurements of gas- and particle-phase composition in the chamber are conducted using a
- 154 suite of analytical instruments. Ozone is measured by a UV absorption monitor (2BTech). NO_x is
- 155 monitored using a chemiluminescence $NO-NO₂-NO_X$ analyzer (Thermo Fisher Scientific) and was below
- 156 the instrument detection limit in all experiments. Reactant VOC and OVOC products are monitored
- 157 using a Vocus proton transfer-reaction mass spectrometer (PTR-MS, Tofwerk, Aerodyne Research,
- 158 Inc.²⁵), and an ammonium chemical ionization mass spectrometer (NH₄⁺ CIMS, modified PTR3, see
- 159 Zaytsev et al.²⁶). Particle concentration and composition are measured using a scanning mobility particle
- 160 sizer (SMPS, TSI) and an aerosol mass spectrometer (AMS, Aerodyne Research, Inc.²⁷). Analytical 161 instruments are summarized in Table S2. Gas-phase mass spectrometric data is background-subtracted
- 162 and corrected for dilution. The analysis does not account for variations in detection efficiencies, which
- 163 may be substantial,²⁶ and we therefore report relative signals, which are unaffected by such calibration
- 164 uncertainties, rather than absolute concentrations. Particle-phase data is corrected for dilution and wall
- 165 losses by normalizing to the ammonium sulfate seed particle concentration. Data analysis and
- 166 guantification approaches are described in more detail in Section S1.3.
- 167 **Results and Discussion**

168 *Ozone production*

- 169 The production of ozone by 222 nm light is examined via the irradiation of clean chamber air.
- 170 Figure 1 shows results from four representative irradiation experiments, run at different
- 171 ventilation rates (1.3 to 3.1 air changes per hour (ACH)) and relative humidities (25%-45%). O₃
- 172 production is observed to occur immediately when the lights are turned on. O3 levels increase
- 173 quickly at first, eventually leveling off to a steady-state value, in which photolytic production is
- 174 balanced by removal by outflow. The O₃ production rate is measured at 324 ± 18 ppb hr⁻¹, in
- 175 reasonably good agreement with previous measurements²² when differences in average GUV $_{222}$
- 176 fluence rate are considered (see Section S3.1). The steady-state $O₃$ concentration is independent
- of relative humidity, and inversely proportional to ventilation rate (Figure S3).
- Dashed lines in Figure 1 denote O3 concentrations predicted from a simple box model. This
- 179 model includes O_2 photolysis (R1-2), O_x -H O_x chemistry, and dilution (See Table S3 for rate
- 180 constants and photochemical parameters^{9–11,28–32}). Model parameters (e.g., light intensity, air-
- exchange rate, and RH) are matched to each experiment. O3 deposition, which is likely small on
- 182 Teflon surfaces, is not included. The model accurately predicts measured O3 levels, indicating
- that the processes describing ozone levels (formation from O3 photolysis at 222 nm, loss by
- outflow) are well-captured by the simple model.

agree well with the predictions from the simple box model (dashed lines) across a range of

ventilation rates and relative humidities. Measurements shown in red are taken at 25% RH.

-
- *Decay of VOCs upon 222 nm irradiation.*

In a second set of experiments (Table S1), VOCs are added to the irradiated chamber after O3

levels reach steady state. Experiments center on two VOCs: hexanal, a C6 compound that reacts

only with OH, and cyclohexene, a C6 compound that reacts with both OH and O3. VOC decays

are shown in Figure 2. Negligible change in O3 concentration is observed upon introduction of

195 10 ppb of VOC; when 100 ppb of cyclohexene is introduced, a small O₃ depletion $(\sim 4.3 \text{ pb})$ is observed.

 Figure 2: Normalized decays of two VOCs (hexanal and cyclohexene) after introduction to the GUV222-irradiated chamber (see also Figure S4). Time = 0 refers to when the VOC was injected into the chamber. Traces are background- and dilution-corrected, so observed decays are from oxidative loss only. Details of each experimental condition (base, O3 only, low light) are given in the text and Table S1. Solid black lines denote single-exponential fits to the observed decays; 204 dashed black lines show the expected decay of cyclohexene from reaction with O3 only.²⁸

Under "base conditions" (10 ppb VOC precursor, 222 nm light, ~25% RH) (Figure 2AB), the

concentrations of both hexanal and cyclohexene decrease after being introduced to the irradiated

chamber. Concentrations are corrected for dilution; losses by direct photolysis and uptake to

surfaces are expected to be minimal (see Section S3.2). Therefore, decays indicate oxidative loss

210 only. This oxidation cannot be explained by O_3 alone. Hexanal does not react with O_3 – a very

- small decay of hexanal is attributed to minor, non-oxidative loss pathways (see SI). While
- 212 cyclohexene does react with O_3 , its decay is far faster than what can be attributed to the O_3
- 213 reaction (dashed line). Indeed, for experiments in which the GUV_{222} light is off and VOCs are
- 214 exposed to the same levels of O_3 as in the irradiated case (Figure 2CD), the hexanal does not
- decrease at all, and cyclohexene decays far less than in the irradiation case, at a rate consistent 216 with reaction with O_3 (plus a small contribution from OH generated by the ozonolysis reaction,
- reaction R3). This observed "excess reactivity" (the difference in observed decays and decays
-
- 218 expected from O_3 reaction alone) indicates that GUV_{222} irradiation generates not only O_3 but
- other oxidants as well.
- Additional experiments carried out under a range of reaction conditions provide evidence that
- these additional oxidants are OH radicals, formed from reactions 3-5. For example, experiments
- 222 with the 222 nm fluence rate attenuated substantially $({\sim} 9 \mu W \text{ cm}^{-2}$, Figure 2EF) exhibit VOC
- decay rates that are much slower compared to those under base conditions. This attenuation is
- 224 assumed to decrease steady-state O₃ concentrations proportionally. However, the observed
- excess reactivity disproportionately decreases, by approximately an order of magnitude. This is
- 226 consistent with OH formation, which depends on the photolysis of both O_2 and O_3 , as well as (in the case of cyclohexene) the ozonolysis reactions. The dependence of decays on other
- experimental parameters, such as VOC concentration and relative humidity, are also consistent
- 229 with OH production from GUV_{222} lights; this is discussed in detail in Section S3.3.
- We estimate average OH levels in all experiments using an exponential fit to the VOC timeseries
- 231 and known OH rate constants.^{29,30} For cyclohexene experiments, average measured [O₃] and the
- 232 O_3 + cyclohexene rate constant are included in the fit to account for excess reactivity. The range
- of [OH] measured in each experiment is calculated by applying the same exponential fits to a
- rolling 15-minute window (see Section S3.4 for more details). We also calculate OH levels using
- our simple box model by including a highly simplified oxidation scheme (Table S3) for each
- 236 injected VOC. Reaction rates of the VOC with OH and O₃ are taken from the literature, and
- oxidation products are assumed to have the same OH reactivities as their precursors. Measured
- 238 and modeled average [OH] agree well (Figure 3), providing strong evidence that GUV_{222}
- 239 produces not only O_3 (R1-2) but also OH (R3-5), and that oxidation by both O_3 and OH can take
- place upon irradiation with 222 nm light.

- *Figure 3*: Experimentally-derived average OH concentration vs. average OH concentration
- predicted by the box model, for all cyclohexene and hexanal experiments (see Section S3.4).

244 Note the break in the x-axis. Error bars represent the range of values observed throughout the 245 experiment.

- 246 *Formation of gas-phase oxidation products*
- 247 The formation of oxidized gas-phase products is observed in all experiments in which VOC
- 248 oxidation occurs. Product distributions for three cyclohexene experiments (base conditions, O_3)
- 249 only, and low light) are shown in Figure 4. Additional product distributions and time-series
- 250 results (including for the hexanal experiments) are provided in Figures S5 and S6.

251
252

Figure 4: Gas-phase products from cyclohexene experiments. Panel A: Normalized mass

- 253 spectrometric signal of products formed for the GUV222 irradiation (base conditions), O3-only,
- 254 and low-light experiments (see Section S3.5 for calculations and Figure S5 for other
- 255 experimental conditions). Signals are integrated from $t = 250$ s to 2500 s, normalized to total
- 256 integrated ion signal and grouped by carbon number (nC). In all cases products are dominated by
- $257 \text{ C}_6\text{H}_{10}\text{O}_2$ (the major cyclohexene + OH reaction product) and $\text{C}_6\text{H}_{10}\text{O}_3$, (the major cyclohexene +
- 258 O₃ product). Panel B: The ratio of the $C_6H_{10}O_2$ -to- $C_6H_{10}O_3$ signals vs. the ratio of the rates of 259 OH and O_3 oxidation, for all cyclohexene experiments. Concentrations of OH are determined
- 260 from the fits in Figure 2, while concentrations of O3 are measured directly. The dashed line is a
- 261 linear fit to the data; since the two products have differing sensitivities in the instrument, this
- 262 differs from the 1:1 line. Error bars represent the range of values observed throughout the
- 263 experiment.
- 264 Measured products are dominated by C6 and C5 compounds, as expected given that cyclohexene
- 265 is a C6 species. The two products with the largest mass spectrometric signals, $C_6H_{10}O_2$ and
- 266 C₆H₁₀O₃, are the major products of the OH and O₃ initiated oxidation of cyclohexene,
- 267 respectively^{33,34} (see Scheme S1) (Products are detected as the analyte-NH₄⁺ adduct, and reported
- as the analyte formula.) The ratios of the signals from the two products vary among experiments,
- indicating differences in the relative concentrations of OH and O3. In Figure 4b, the ratio of the
- 270 mass spectrometric signals of these two products is shown vs. the relative OH-to-O₃ oxidation
- 271 rate ratios (calculated from the experimentally-determined values of [OH] and [O₃]) for each
- 272 cyclohexene experiment. A strong correlation ($R^2 = 0.98$) is found between the two ratios,
- providing further support for OH-initiated oxidation, and more generally for OH radical
- production from irradiation by 222 nm light. The products formed in the 222 nm irradiation of
- 275 hexanal are also broadly consistent with OH-initiated oxidation (see Scheme S2).³⁵
- *Secondary organic aerosol formation*
- In all experiments, dry ammonium sulfate seed particles are added to the chamber, providing
- surface area onto which low-volatility species may condense, and enabling the assessment of
- potential SOA formation. SOA formation is observed in a number of experiments (Table S1 and
- Figure S7). SOA formation is generally modest for most hexanal and cyclohexene experiments,
- 281 likely due to the relatively small size (C6) and low concentrations (10 ppb) of those species.
- Higher concentrations of SOA are observed for experiments with high initial concentrations (100
- 283 ppb) of hexanal or cyclohexane, and for those using limonene $(C_{10}H_{16}$, a monoterpene commonly
- 284 found in fragrances and cleaning products). In fact, the GUV_{222} irradiation of 100 ppb limonene
- 285 (a level that can be found in indoor environments immediately after cleaning events $36,37$) results
- 286 in exceedingly high SOA loadings, on the order of $400 \pm 80 \,\mu g \, \text{m}^{-3}$. Additionally, the formation
- of new particles is observed upon 222 nm irradiation under some conditions (Section S3.6 and
- 288 Figure S8). This effect is not observed when O_3 is added without 222 nm irradiation. GUV $_{222}$ -
- induced nucleation occurs even when no VOCs are added, and so may result from
- photochemistry of organic species on the chamber surfaces, or even of the surface materials
- themselves. Whether this is a general feature of the irradiation of organics on indoor surfaces is
- unclear from the present experiments, but it does suggest that 222 nm irradiation may induce
- new particle formation in some environments.

Extrapolation to indoor environments

- 296 The above laboratory experiments demonstrate that GUV_{222} irradiation forms ozone, OH, and a
- range of oxidation products; measured ozone and inferred OH agree broadly with predictions by
- a photochemical box model. However, real-world indoor environments are substantially different
- than our simple laboratory system: they involve a large number of organic compounds,
- depositional loss of ozone and other species, infiltration of outdoor pollutants, a wide range of
- possible ventilation rates, and typically much lower average UV fluence rates. Here we extend
- our photochemical model to a more realistic indoor air scenario, with the goal of understanding
- how GUV222 may impact indoor air quality under a range of ventilation and irradiation
- conditions.
- For simulations of chemistry in a more realistic indoor environment, two "lumped" VOCs are
- 306 included in the model: one (VOC1) that reacts with OH but not with O_3 , and another (VOC2)
- that reacts with both OH and O3. Rate constants for VOC1 are chosen based on typical values for
- indoor VOCs (Section S2.1 and Tables S3 and S4); rate constants for VOC2 are assumed to be
- 309 equal to those of limonene. OH yields from $O_3 + \text{VOC2}$ are assumed to by 0.86, equal to that of
- 310 limonene.¹¹ All oxidation reactions form lumped organic products that can also react with OH.
- 311 VOC emission rates (84 ppb hr^{-1} and 4.2 ppb hr^{-1} for VOC1 and VOC2, respectively) are
- 312 determined from previous measurements of OH and $O₃$ reactivities in indoor environments;^{38,39}
- details of these calculations are given in Section S2.1. The model is run at 298 K, 1 atm, and
- 30% RH. We also include a background concentration of O3 in the ventilation air (40 ppb,
- 315 consistent with typical outdoor O_3 concentrations), a 25% loss of O_3 to the ventilation system,
- 316 and an O_3 deposition constant of 3 hr^{-1 10,31}.
- The range of light fluence rates chosen covers US and international guidelines on 222 nm
- 318 exposure limits (ranging from 0.8 to 16 μ W/cm² assuming a continuous 8-hour exposure^{40,41}) as
- well as the values in previous studies used for pathogen deactivation (average irradiance of up to
- 320 2.73 μ W/cm² at 1.7 m above the ground from Eadie et al.⁴² and 3.5 μ W/cm² from Peng et al.²³).
- The range used in our model extends higher to take into account proposals for the use of
- 322 significantly higher light fluence rates, and include the fluence rates in our experiments (\sim 45
- $323 \mu W/cm^2$). Ventilation rates span a range of typical indoor values, and include the minimum
- American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE)
- recommendations for homes (0.35 ACH), offices (~2-3 ACH), and health care settings (10
- 326 ACH).⁴⁴

 Figure 5: Effects of ventilation and GUV $_{222}$ fluence rate on modeled GUV efficacy and indoor air quality (see also Figures S9-S12). Panel A: effective air changes per hour (eACH) for indoor 330 pathogens, based on the previously reported inactivation rate of SARS-CoV-2 at 222 nm⁴⁵ (Section S4.2). Panels B-D: steady-state concentrations of (B) O3,(C) OH, and (D) organic oxidation products, respectively, as predicted by the photochemical box model. Panel D calculations assume unit yields, and do not account for VOC production from surfaces (see 334 Figure S11) or recycling by NO_x -HO_x interactions (Figure S12), so likely represent lower limits. Lighter colors represent larger values; note that the logarithmic color scaling is different for each panel. Additional model results are given in Figures S9 and S10.

- Key model results are provided in Figure 5. Figure 5A shows the effective air change rate
- (eACH) across a wide range of GUV $_{222}$ fluence and ventilation rates; even modest irradiation
- levels lead to substantial increases in eACH (see also Figure S9A). Figures 5B, C, and D show
- the steady-state indoor concentrations of O3, OH, and total oxidation products (assuming unit
- yield), respectively.
- Steady-state ozone levels (Figure 5B) are higher with 222 nm irradiation than without. Sources
- of O₃ include photochemistry (R1-2) and infiltration of outdoor air, while sinks include
- deposition, ventilation, and chemical reaction (rates and contributions of individual processes are
- given in Figures S9B-E). With low irradiation, O3 levels are governed mainly by infiltration of
- 347 outdoor air, and Ω_3 increases are modest. Under the highest fluence rates ($>25 \mu W/cm^2$), and
- 348 especially under low ventilation rates (\leq 1 ACH), indoor O₃ can reach levels exceeding that of the outdoors, and can even exceed the OSHA indoor limit of 100 ppb. However, even a small
- 350 change in indoor O_3 levels can have a dramatic effect on people's total ozone exposure, 46 given
- the large fraction of time people spend indoors. In most cases, deposition represents the
-
- dominant sink of ozone (Figure S9D).
- Figure 5C shows steady-state levels of OH as a function of ventilation and 222 nm light
- 354 intensity. Sources of OH include O₃-alkene reactions $(R3)$ and photochemistry $(R4-5)$, while
- 355 sinks are dominated by reactive losses (see also Figures S9F-G). In the absence of GUV222
- 356 irradiation, modeled OH is from alkene ozonolysis only, with predicted levels $(\sim 10^5 \text{ molec cm}^{-3})$
- overlapping but falling on the low end of measured and modeled OH in indoor spaces (which
- 358 range from $6x10^4$ -1.6x10⁶ molec cm⁻³);^{47–56} this underestimate may arise from the omission of
- photolysis of trace species such as nitrous acid (HONO) or aldehydes, which may be important
- 360 in some environments.⁵⁷ As with O_3 , GUV₂₂₂ irradiation leads to increases in indoor levels of
- OH. At low to moderate irradiation levels, this increase in OH is mostly due to the alkene ozonolysis reaction, while at higher levels, ozone photolysis plays a larger role (Figure S9G).
- 363 OH increases with increasing photochemistry (higher GUV₂₂₂ fluence rates and ozone
- concentrations), but is substantially modulated by losses from reaction with VOCs. VOC
- concentrations are higher at low ventilation rates (see Figure S9H), due to the buildup of emitted
- VOCs, which suppresses OH concentrations. At high light intensities, steady-state OH levels can
- approach outdoor levels, matching or exceeding indoor OH measurements during transient
- 368 events such as cleaning or cooking activities.^{58,59}
- 369 The production of O_3 and OH by GUV $_{222}$ -driven chemistry and their subsequent reactions with
- VOCs leads to an increase in organic oxidation products (OVOCs and SOA). Steady-state levels
- and production rates of such products (assuming unit yields) are shown in Figures 5D and S9I.
- Concentrations increase with increased light intensity, and are especially high at low ventilation
- rates. Since more than one product molecule may be formed per oxidation reaction, and OVOCs
- may also be formed by surface reactions of O3 or OH, these numbers likely represent lower
- limits. Of particular concern is the production of hazardous air pollutants (HAPs, such as CH2O)
- and SOA, both of which may represent health hazards in the indoor environment. Concentrations
- of SOA are challenging to predict, as SOA production depends on the amounts and identity of
- the indoor VOCs, as well as on a host of reaction conditions. However, SOA levels on the order
- 379 of a few μ g/m³ might occur (Figure S10); the production of SOA from 222 nm irradiation in
- realistic indoor settings is an important area of future research.
- The simplicity of the model neglects some additional secondary effects, which are highly
- uncertain. For example, volatile secondary organic products stemming from reactive surface
- 383 losses of O₃ (e.g., to paint, textiles, skin)^{12,60} could represent an additional secondary effect of
- GUV222 on indoor air quality. Preliminary modeling suggests that this may increase OVOC
- concentrations by as much as a factor of 100 (Figure S11). Similarly, indoor environments 386 contain NO_X, which can affect the levels and fates of oxidants. While NO_X chemistry is not
- modeled explicitly here, due in part to uncertainties in NO_X photolysis processes, we have
- carried out additional simulations to estimate the role of HOx-NOX cycling. As shown in Section
- S4.1 and Figure S12, such cycling increases OH concentrations and OVOC product formation
- substantially. We do not examine the role of HONO, which can be present in high (ppb) levels
- 391 indoors⁵⁵ and absorbs strongly at 222 nm (σ = 1.35 x 10⁻¹⁸ cm² ⁹); HONO photolysis may lead to
- even higher OH levels than predicted here. All of these effects have the potential to increase
- OVOC formation, suggesting that the OVOC concentrations presented in Figure 5 are best
- understood as a lower limit and that the indoor air quality impacts of 222 nm irradiation could be
- more severe than predicted here.

Implications

- 397 Our laboratory studies demonstrate that GUV_{222} light leads to the production of (1) ozone, (2)
- OH radicals, and (3) secondary organic species (OVOCs and SOA); these are in broad agreement
- 399 with prior model predictions.²³ The resulting concentrations of such secondary species can be
- substantially higher than are normally found in indoor environments; in extreme cases, these
- increases can be dramatic, leading to oxidation conditions similar to those found in outdoor
- environments. The negative health impacts associated with the unavoidable generation of these 403 secondary species – most importantly O_3 , fine particular matter, and $HAPs$ – thus need to be
- taken into account (and ideally mitigated) when considering the use of 222 nm disinfection in
- indoor spaces.
- 406 While a detailed analysis of the health impacts of GUV_{222} use (both the benefits from
- inactivation of airborne pathogens and the drawbacks from secondary pollutant formation) is
- beyond the scope of this work, our results offer some broad guidance as to the optimal use of
- GUV₂₂₂ in indoor environments. Most importantly, GUV₂₂₂ disinfection alone is not a safe
- substitute for ventilation as a means to control levels of indoor airborne pathogens, as it can lead
- to the buildup of indoor ozone and other pollutants to dangerous levels (Figure 5). However,
- GUV222 may be effectively used in conjunction with ventilation: relatively modest irradiation
- levels combined with carefully chosen ventilation conditions can greatly enhance the effective
- air change rate (Figure 5A), while limiting the levels of secondary pollutants (Figures 5B-D).
- 415 Moreover, due to the unavoidable formation of secondary pollutants, GUV222 lights should be
- 416 run at the lowest effective levels whenever possible. Further, the combination of GUV_{222}
- irradiation with air-cleaning technologies (e.g., sorbents for ozone and OVOCs, filters for
- particulate matter) may serve to minimize indoor secondary pollutant levels, potentially enabling
- safer use of GUV222 under poorly-ventilated environments. Quantifying the benefits and
- 420 tradeoffs of these combined approaches (ventilation, GUV222 irradiation, and/or air cleaning) in
- terms of pathogen transmission, air pollutant levels, human health, and cost-effectiveness, is a
- critical next step toward ensuring healthier indoor environments.

Supporting Information

- Experimental conditions and methods, additional modeling methods, additional results and
- discussion (incl. ozone steady-state calculations, discussion of VOC loss pathways, discussion of
- elevated VOC and RH experiments, more complete product distribution figures, reaction
- schemes, and SMPS data), model results (incl. additional model output figures and calculation of
- effective air changes per hour) (PDF)

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