

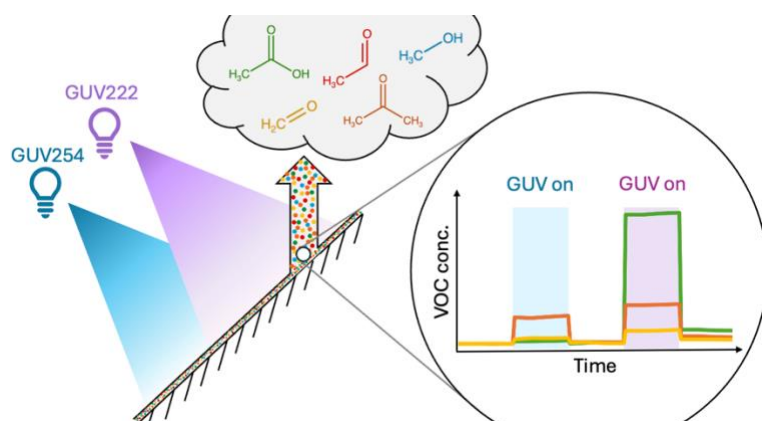
# Indoor Surface Emissions of Volatile Organic Compounds induced by Germicidal UV (222 & 254 nm) Illumination

Sara Bjerre Sørensen<sup>1</sup>, Kasper Kristensen<sup>1,\*</sup>

<sup>1</sup> Department of Biological and Chemical Engineering, Aarhus University, Aarhus C, 8000, Denmark

\*Corresponding email: [kasper.kristensen@bce.au.dk](mailto:kasper.kristensen@bce.au.dk)

## ABSTRACT



The application of germicidal ultraviolet (GUV) lamps has recently attracted increased attention as a measure to mitigate indoor disease transmission. Among the most commonly

employed are traditional mercury lamps and krypton-chloride excimer lamps, which emit UV-C light with peak wavelengths of 254 nm (GUV254) and 222 nm (GUV222) respectively. This study investigates volatile organic compound (VOC) surface emissions induced by GUV254 and GUV222 lamps across various surface materials. Near surface proton-transfer-reaction time-of-flight mass spectrometer (PTR-TOF-MS) measurements revealed significantly increased surface emissions during GUV illumination. In addition to compounds intrinsic to the illuminated surface, GUV light also increased surface off-gassing of externally applied compounds including limonene and 4-oxopentanal (4-OPA). The magnitude of the enhanced surface emissions was found to decrease with increasing GUV pathlength resembling the expected decrease of the irradiance. Overall, greater surface emissions were induced by the

GUV222 lamp compared to the GUV254 lamp. Conclusively, this study identifies UV-induced surface emissions as a potential source of indoor VOCs during GUV lamp application.

## KEYWORDS

*Indoor air quality (IAQ), GUV lamps, far UV-C, photochemistry, surface materials.*

## SYNOPSIS

The mechanisms driving changes in volatile organic compound (VOC) levels during germicidal ultraviolet radiation indoor remain underexplored. This study uncovers GUV-induced surface emissions as a source of indoor VOCs.

## 1 INTRODUCTION

Although light in the UV-C range (200-280 nm) has been known to constitute an effective way of inactivating pathogens for multiple decades [1, 2], interest in germicidal ultraviolet (GUV) lamps as a means to minimize disease transmission in indoor environments has increased after the recent COVID-19 pandemic [3]. In addition to the traditional filtered mercury lamps with a peak wavelength of 254 nm (hereafter referred to as GUV254), filtered krypton-chloride excimer (KrCl) lamps with a peak wavelength of 222 nm (referred to as GUV222) have also gained ground. However, although both GUV222 and GUV254 have proven effective in terms of inactivating pathogens [3-8], the possible implications of bringing non-tropospheric (>15 km) radiation into our indoor environments still remain underexplored.

In the outdoor atmosphere, solar radiation in the UV-C range photolyzes the constituents of the air (i.e. break their molecular bonds), thereby initiating formation of oxidants such as ozone ( $O_3$ ,  $\lambda < 242$  nm [9]) and hydroxyl radicals ( $\cdot OH$ , e.g., through subsequent photolysis of  $O_3$ ), along with a myriad of chemical reactions. Several studies have shown that

when utilized in indoor environments, GUV222 lamps form  $O_3$  [10-14] and  $\cdot OH$  [12]. In addition, both GUV222 and GUV254 lamps have been observed to cause increases in particle number concentrations [12-17] and in the levels of various volatile organic compounds (VOCs) [12-16]. While GUV-induced breakdown of larger VOCs has been shown to increase levels of VOCs such as acetic acid and propionic acid in the gas-phase [18], recent studies have hypothesized that elevated VOC levels in indoor air following GUV operation could also be caused by VOC emissions from indoor surfaces promoted by the high-energy light from the GUV222 and GUV254 lamps [13, 15].

In this paper, we study how GUV222 and GUV254 radiation enhance emissions of VOCs from distinct indoor surfaces containing different chemical constituents inherent to the illuminated materials and externally applied to the surface prior to illumination. In addition, we report on surface VOC emissions in relation to the UV pathlength and irradiance and address the potential impacts on indoor air composition from location of the GUV lamps.

## 2 MATERIALS AND METHODS

**Experimental design and measurements.** In this study, a filtered GUV222 lamp (Care222 w. B1 module, Ushio Inc.) and a GUV254 lamp (TUV PL-L 18W/4P 1CT/25, Philips) were used to illuminate various surfaces ( $\sim 1.44\text{ m}^2$ ,  $1.2 \times 1.2\text{ m}$ ) mounted on the wall in an unfurnished room ( $\sim 65\text{ m}^3$ ,  $6 \times 4 \times 2.7\text{ m}$ ). In each experiment, the lamp was placed 1 meter from the surface it was facing with its center 1.32 meter above the floor corresponding to the center of the surface. A sketch of the set-up can be seen in Figure S1a. The incident irradiance at the center of the surface was  $12.3\text{ }\mu\text{W cm}^{-2}$  during GUV222 illumination (see Figure S1b) and  $\sim 155\text{ }\mu\text{W cm}^{-2}$  during GUV254 illumination (see Supporting Information for elaboration).

In total, six different surface materials were exposed to the UV light: Teflon (white non-transparent, 1 mm PTFE sheet); a painted plaster wall (room wall, painted 3 years prior);

plywood (Radiata pine); painted plywood (Radiata pine painted with Flutex-Pro 10 acrylic paint from Flügger six weeks prior); Teflon coated with limonene ( $\sim 55 \text{ mg/m}^2$  corresponding to 2 wt% in 5 mL EtOH), a VOC found in cleaning agents [19, 20]; and Teflon coated with 4-oxopentanal (4-OPA,  $\sim 35 \text{ mg/m}^2$ ), a common constituent in occupied indoor environments from reaction of ozone with skin lipids [21, 22]. See Supplementary Information for elaboration on the preparation of the surfaces. Lastly, correlation between the GUV222 light pathlength and VOC surface emissions was assessed from experiments in which the lamp was placed at varying distances from the room wall (1-5 meters in steps of 1 m). An overview of the experiments can be seen in Table S1.

Measurements of VOCs were performed by proton-transfer-reaction time of flight mass spectrometer (PTR-TOF-MS 4000, IONICON Analytik GmbH, Austria) [23]. The PTR-TOF-MS drift tube was operated with a temperature of  $60^\circ\text{C}$ , a pressure of  $\sim 3.20 \text{ mbar}$  and voltage of  $900 \text{ V}$ , giving an  $E/N$  ratio of  $\sim 135 \text{ Td}$ . For all experiments, mass-to-charge ratios ( $m/z$ ) from 15 to 250 were measured with an inlet flow of  $100 \text{ sccm}$  and a time resolution of 20 seconds. Teflon tubing from the PTR-TOF-MS was connected to a switching valve (VICI), which for a subset of the experiments allowed alternating sampling 1 and 61 cm from the surface respectively. The data was subsequently analyzed with PTRwid [24] to quantify and assign chemical formulas. Tentative compound assignments of all chemical formulas presented throughout the paper can be seen in Table S2. PTR-TOF-MS transmission was obtained using a liquid calibration unit (LCU, Ionicon) generating known gas-phase concentrations of methanol, acetone, butanone, p-cresol, indole and phenyl-cyclohexanone. For conversion to mixing ratios, a rate constant of  $2 \cdot 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  was used yielding mixing ratios typically accurate to  $\pm 50\%$ . Formaldehyde concentrations from PTR-TOF-MS measurements were calibrated against high performance liquid chromatography (HPLC) in a 4-point calibration curve ranging from  $\sim 10$ -560 ppb. Indoor  $\text{O}_3$  levels were continuously monitored with an  $\text{O}_3$  monitor (model

202, 2B Technologies), while temperatures and relative humidities were measured using sensor nodes (AirNodes, AirLabs). Average temperatures and relative humidities in the room were  $21 \pm 1$  °C and  $32 \pm 3$  % respectively. Surface temperatures, measured with an InfraRed Thermometer (Art. 15-254, Biltema), were comparable to room temperatures prior to illumination. Post-illumination measurements conducted for a subset of experiments showed no temperature increase as a result of GUV illumination. Except for the PTR-TOF-MS Teflon tubing, no tubing or instrumentation was exposed to the GUV light.

**Enhancement Factors.** Near-surface (1 cm) concentration enhancement factors (EF) were calculated using the median concentrations (C) measured for each GUV on/off period (see Table S3 and S4). To account for non-related changes in the room concentrations (e.g. steady decreases due to natural loss or steady increases from room emissions) an average of the concentrations measured prior (30 min) and post (30 min) GUV illumination was used:

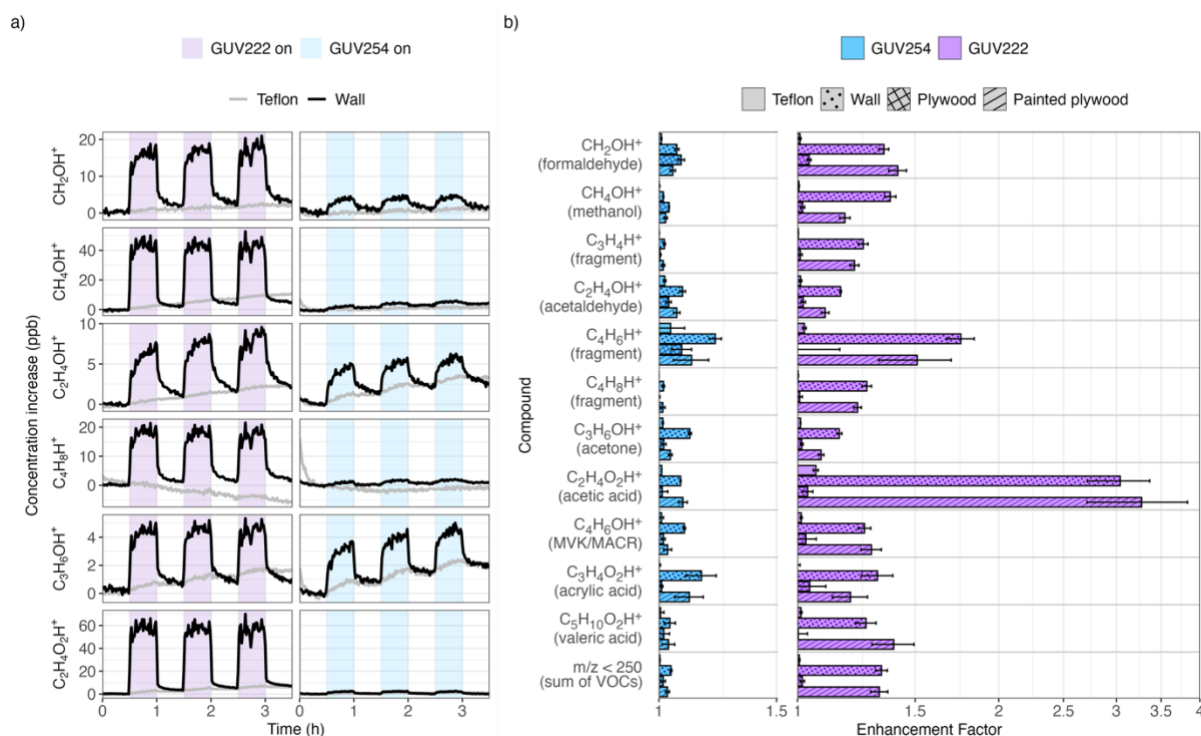
$$EF = \frac{C_{GUV\ on}}{(C_{GUV\ off,\ prior} + C_{GUV\ off,\ post})/2} \quad \text{Eq. (1)}$$

### 3 RESULTS AND DISCUSSION

**Surface Emissions of Volatile Organic Compounds from GUV Radiation.** Figure 1a shows the levels of selected VOCs measured 1 cm away from the room wall (painted plaster wall) and a Teflon surface respectively during three consecutive periods with on/off illumination with GUV lamps positioned 1 m from the surface. Illumination of the wall with 222 nm light showed instantaneous increases in the levels of numerous mass-to-charge ratios including, but not limited to,  $m/z$  31, 33, 45, 57, 59 and 61, assigned as  $\text{CH}_2\text{OH}^+$ ,  $\text{CH}_4\text{OH}^+$ ,  $\text{C}_2\text{H}_4\text{OH}^+$ ,  $\text{C}_4\text{H}_8\text{H}^+$ ,  $\text{C}_3\text{H}_6\text{OH}^+$  and  $\text{C}_2\text{H}_4\text{O}_2\text{H}^+$  respectively. It is suggested that these compounds represent the protonated ions of formaldehyde, methanol, acetaldehyde, an alcohol fragment [25], acetone and acetic acid respectively. The levels remained elevated during illumination but were

followed by immediate decreases as the lamp was turned off. While gas-phase production would typically lead to continuous changes in the VOC room levels, the abrupt changes observed suggest that a GUV222-triggered on/off mechanism at the surface, e.g. surface off-gassing, is the dominant cause of the altered VOC levels. This is supported by alternating measurements sampled 1 and 61 cm away from the wall respectively, which show little or no changes in VOC levels at the 61 cm distance (see Figure S2). Similar trends were also observed during illumination with the GUV254 lamp. However, despite the more than tenfold greater irradiance induced by the GUV254 lamp compared to the GUV222 lamp ( $\sim 155$  vs.  $12.3 \mu\text{W cm}^{-2}$  at the center), the effects of GUV254 illumination of the wall were much less pronounced.

When the wall was covered by Teflon, the increase in VOC levels upon illumination was greatly diminished resulting in close to stable measurements across the majority of compounds presented independent on GUV operation. For  $\text{C}_2\text{H}_4\text{OH}^+$  and  $\text{C}_3\text{H}_6\text{OH}^+$  however, gradual increases were observed during GUV illumination of the Teflon surface followed by stabilization during GUV off-periods. This is hypothesized to arise from either potential light spill onto the surrounding wall, reflection of UV light by the reflective Teflon surface [26] onto adjacent walls not covered by the Teflon sheet or contributions from gas phase reactions (i.e. OH or  $\text{O}_3$  oxidation chemistry) initiated by the GUV lamps [12, 14, 16, 27]. While  $\text{O}_3$  production was evident with GUV222,  $\text{O}_3$  levels remained at  $\sim 0$  ppb during GUV254 operation (see Figure S6), leaving OH oxidation as the only plausible oxidation pathway during GUV254 illumination. Overall however, PTR-TOF-MS measurements did not reveal any consistent decreases in compound levels that would suggest a pronounced depletion of larger VOCs from either gas-phase photolysis or oxidation during GUV illumination.



**Figure 1:** **a)** Measured concentration increase (ppb) of various VOCs 1 cm away from a Teflon surface (grey line) and the room wall (black line) during illumination with 222 nm light (purple background) and 254 nm light (blue background). All concentration increases are normalized to the last measurement before the first GUv on-period. **b)** Near-surface (1 cm) concentration enhancement factors (EF, see Eq. 1) across selected compounds during illumination of Teflon (no pattern), room wall (dots), plywood (crosshatches) and painted plywood (stripes) with 222 nm light (purple, 12.3  $\mu\text{W cm}^{-2}$  at the surface center) and 254 nm light (blue, 155  $\mu\text{W cm}^{-2}$  at the surface center). Tentatively assigned compound names are given in parenthesis below the compound formula. MVK and MACR refers to methyl vinyl ketone and methacrolein respectively. Please note that the x-axis has logarithmic spacing.

In addition to the wavelength of the applied GUv light (254 vs 222 nm), the surface illuminated was also found to play a key role in terms of potential off-gassing. Figure 1b shows the Enhancement Factors (EFs) across 11 compounds and for the sum of VOCs during illumination of the Teflon sheet, the room wall (painted plaster wall), plywood and painted plywood. EFs from painted plywood were found to be significantly greater than for unpainted plywood during GUv222 illumination. Time resolved data from illumination of plywood and painted plywood across selected compounds can be seen in Figure S3. In fact, EFs from the painted plywood strongly resembled those from the painted plaster wall suggesting that during GUv222 illumination of the wall, the majority of the off-gassed compounds originate from the paint coating. In support of this, PTR-TOF-MS headspace analysis of the wet paint confirmed the presence of all VOCs presented in Figure 1 (see Figure S4). Across the four surfaces

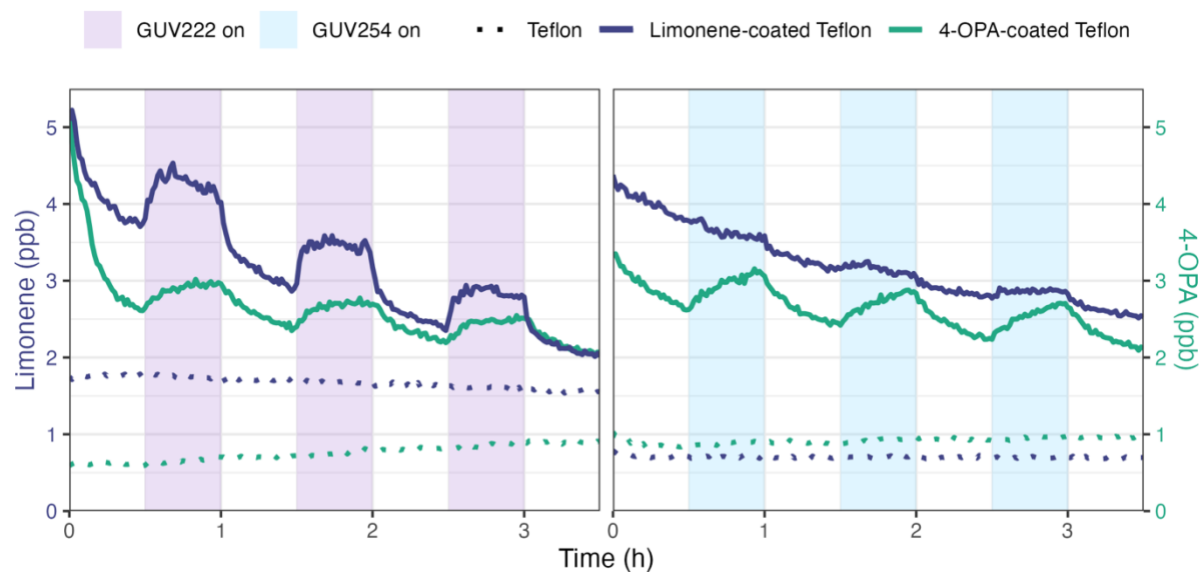
presented in Figure 1b, GUV222-induced surface off-gassing from the wall and painted plywood was unequivocally the most significant. The largest EFs observed overall was related to  $\text{C}_2\text{H}_4\text{O}_2\text{H}^+$  (acetic acid) off-gassing during GUV222 illumination of the painted plywood and the wall and was found to be  $3.3 \pm 0.5$  and  $3 \pm 0.3$  respectively. This is in line with a previous study showing increases in acetic acid concentrations in office air during GUV222 operation [13]. In general, surface off-gassing was found to be more pronounced during GUV222 illumination compared to GUV254. While the EFs of the sum of VOCs were  $1.34 \pm 0.03$  and  $1.33 \pm 0.04$  when the GUV222 lamp was used to illuminate the wall and the painted plywood respectively, this was  $1.043 \pm 0.003$  and  $1.030 \pm 0.007$  for the GUV254 lamp. Considering the more than tenfold lower irradiance induced by the GUV222 lamp compared to the GUV254 lamp, these results suggest that the surface off-gassing is wavelength-dependent, likely due to the absorption cross-sections of the irradiated surface molecules. Several studies report on UV-induced degradation of larger organic compounds and polymers resulting in formation and emissions of VOCs following exposure to UV-light [18, 28]. Photo-induced degradation of the illuminated surface materials, e.g. breakdown of paint polymers [29], could thus potentially explain the observed emissions.

Alternatively or in addition, the observed emissions could also be induced by photodesorption [30] or an internal energy increase in surface molecules following UV absorbance increasing the vibrational energy of the molecules allowing them to evaporate from the surface as hypothesized in previous studies [13, 15]. In relation, GUV application was found to promote off-gassing of chemicals externally applied to the surface prior to illumination. Figure 2 shows the limonene (sum of  $\text{C}_{10}\text{H}_{16}\text{H}^+$  and  $\text{C}_6\text{H}_8\text{H}^+$ ) and 4-OPA ( $\text{C}_5\text{H}_8\text{O}_2\text{H}^+$ ) levels during GUV222 and GUV254 illumination of Teflon sheets with and without coatings of limonene and 4-OPA solutions respectively. As expected, the limonene and 4-OPA levels remained stable during on/off illumination of the uncoated Teflon sheets. Only a small increase



in 4-OPA was observed during GUV222 illumination, likely due to the oxidation of naturally occurring squalene in the room by O<sub>3</sub> produced from the GUV222 lamp (see Figure S6). In contrast, experiments with coated Teflon sheets showed an overall decline in near-surface concentrations caused by the continuous depletion of limonene and 4-OPA on the surfaces. However, upon illumination, the GUV222 lamp caused instantaneous increases in the limonene levels from the limonene-coated Teflon sheet, confirming GUV222-induced evaporation of surface molecules. This effect was less pronounced for GUV254 in line with expected lower UV-absorption by limonene at this higher wavelength [31]. For the 4-OPA-coated surface, increased off-gassing of 4-OPA was observed from illumination by both GUV222 and GUV254. Comparison of the off-gassing magnitudes induced by the GUV lamps should be done with caution as these are expected to be highly sensitive to the amount of coating left on the Teflon sheets.

The findings presented in Figure 2 demonstrate that GUV induced surface off-gassing is not limited to compounds intrinsic to the illuminated surface but also apply to those externally applied e.g. during cleaning or formed from e.g. ozone-initiated reactions of skin lipids on indoor surfaces. In extension hereof, increases in the levels of compounds known to originate from oxidation of the applied compounds were also observed during both GUV222 and GUV254 illumination, including C<sub>9</sub>H<sub>14</sub>OH<sup>+</sup> (assigned as limona ketone). It remains unclear whether this compound was formed in the gas phase after off-gassing of limonene or if it was formed on the surface and subsequently off-gassed during illumination.

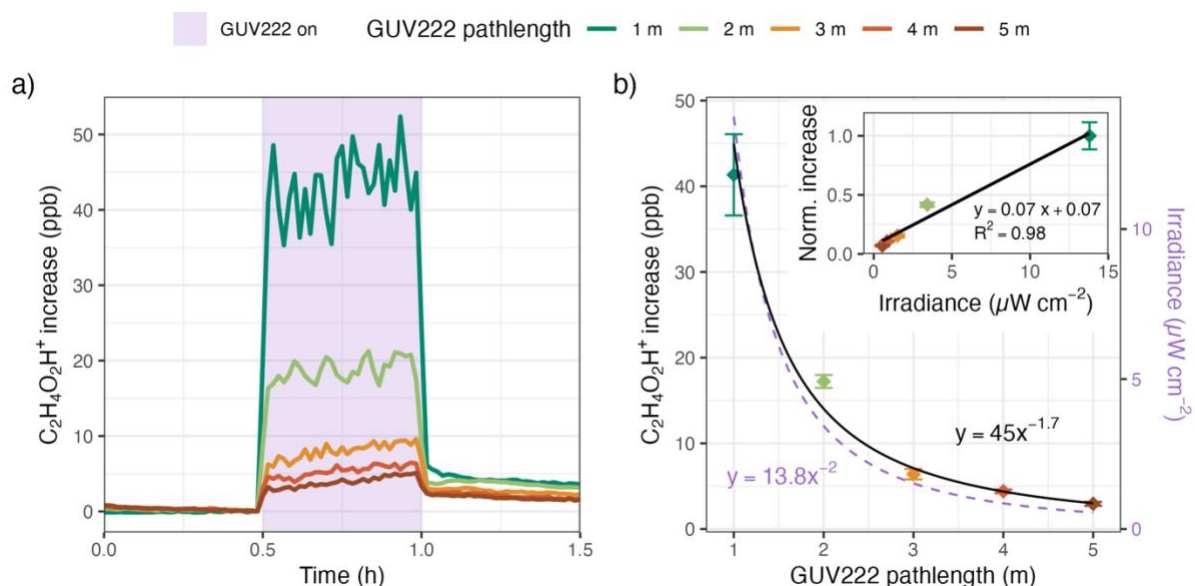


**Figure 2:** Concentrations (ppb) of limonene (sum of  $C_{10}H_{16}H^+$  and  $C_6H_8H^+$ , dark purple) and 4-OPA ( $C_5H_8O_2H^+$ , green) measured during GUV222 (purple background) and GUV254 (blue background) illumination of a Teflon surface (dotted lines), a Teflon surface coated with limonene (solid line) and a Teflon surface coated with 4-OPA (solid line).

**Impact of GUV Pathlength & Irradiance on Surface Emissions.** Figure 3 shows the correlation between the GUV222 pathlength and the magnitude of the surface off-gassing of  $C_2H_4O_2H^+$  from the wall. As illustrated by Figure 3a, GUV222-induced enhancements in near-surface concentrations of  $C_2H_4O_2H^+$  are highly dependent on the distance between the GUV lamp and the wall (i.e. the GUV pathlength) and hence the intensity of the light reaching the surface. For  $C_2H_4O_2H^+$ , the surface off-gassing was found to decrease with a power of  $-1.7 \pm 0.1$  with pathlength, thereby closely resembling that of irradiance which is expected to decrease with a power of -2 with GUV222 pathlength (Fig. 3b). This also implies a close to linear correlation between the enhancement of the surface off-gassing and the incident irradiance at the wall perpendicular to the GUV222 lamp as can be seen in the inset of Figure 3b showing a  $\sim 7\%$  change in surface off-gassing with each  $\mu W\ cm^{-2}$  of irradiance. Although larger uncertainties are related to the fits of other compounds observed to off-gas from the room wall, e.g.  $CH_2OH^+$ ,  $CH_4OH^+$ ,  $C_2H_4OH^+$ ,  $C_4H_8H^+$  and  $C_3H_6OH^+$  as presented in Figure S5, decreases with increasing GUV pathlength were observed in all cases. However, variations between the

potencies with which they decrease (i.e. -0.9 to -1.7) and the deviation from -2 (i.e. the expected irradiance decrease) suggest the presence of other influencing factors. For example, it is speculated that the surface reservoirs of the compounds might be a limiting factor in terms of surface off-gassing, especially at lower GUV pathlengths. Although not tested, GUV254-induced surface off-gassing is hypothesized to display a similar pathlength and irradiance dependency.

It is important to note that the examined GUV pathlengths in the current study were chosen with no regards to GUV222 exposure threshold limit values (TLV) as reported by e.g. American Conference of Governmental Industrial Hygienists (ACGIH). Presently, the ACGIH TLV for 222 nm is 161 mJ/cm<sup>2</sup> corresponding to an 8-hour exposure to 5.6  $\mu\text{W cm}^{-2}$  GUV222 irradiance. The 8-hour TLV is thus lower than the maximum irradiance of 12.3  $\mu\text{W cm}^{-2}$  obtained at 1 meter GUV222 pathlength in the current study, but equals the maximum irradiance obtained at a pathlength of ~1.6 m (see Figure 3b) or a ~3.5-hour exposure to 12.3  $\mu\text{W cm}^{-2}$ .



**Figure 3:** **a)**  $\text{C}_2\text{H}_4\text{O}_2\text{H}^+$  (ppb) measured 1 cm away from the wall during illumination with a GUV222 lamp (purple background) placed at various distances from the wall. All concentrations are normalized to the to the last measurement before the GUV was turned on. **b)** Correlation between the near-surface  $\text{C}_2\text{H}_4\text{O}_2\text{H}^+$  increase (ppb) upon GUV222 illumination and the GUV222 pathlength. The dashed line shows the expected irradiance at the surface perpendicular to the GUV222 lamp. The inset displays the normalized  $\text{C}_2\text{H}_4\text{O}_2\text{H}^+$  increase as a function of the expected irradiance.

## 4 IMPLICATIONS

Changes in VOC concentrations following application of GUV222 in laboratory chambers and real indoor environments have in-large been ascribed to GUV222-induced production and subsequent gas-phase reactions of  $O_3$  and OH-radicals with resulting VOC oxidation by-products [12, 14, 16]. In this work we report that in addition to formation from UV-induced gas-phase chemistry, several VOC species observed following indoor GUV application can be attributed to enhanced emissions from abundant indoor surface reservoirs. Along with VOCs intrinsic to the surface materials, increase in emissions from externally applied compounds are also reported upon GUV illumination, demonstrating that the observed effects are also of relevance for species typically applied to indoor surfaces, including limonene from e.g. cleaning agents [19, 20] and 4-OPA from e.g. reactions of  $O_3$  with skin oils on occupant and nonoccupant surfaces (e.g. dust, and furniture) [21, 22]. Considering the large surface-to-volume ratio of many indoor environments, the reported UV-induced surface off-gassing should be considered as a potentially significant contributor to changes in indoor VOCs associated with GUV application. As the GUV-induced surface emissions are found to be highly correlated with surface irradiance, this emphasizes the importance of GUV lamp irradiance and positioning (e.g. distance to illuminated surfaces) when assessing potential indoor air quality impacts from indoor GUV application. It should be noted that although increasing GUV pathlength will reduce the GUV-induced surface emissions from the illuminated area, the resulting room concentrations will likely be counteracted by an accompanying increase in the total area illuminated. In relation, it is important to note that the enhancements in VOC emissions from GUV illumination reported in this study is from near-surface measurements, thus any resulting increase in room air concentration will depend on factors beyond surface materials, GUV irradiance, pathlength, and irradiated surface area. These include room size (i.e. surface-to-

volume ratio) and ventilation which could significantly limit any indoor air quality impacts associated with the observed GUV-induced surface emissions. Also, the impact of the reported UV-induced emissions on indoor VOCs and air quality is expected to vary significantly considering the large diversity in indoor surfaces and residing chemical reservoirs. In this work, we report on the VOC emissions from a small subset of indoor surface materials and coatings thus calling for further studies of GUV-induced emissions encompassing diverse indoor settings and surfaces as well as a wider range of chemical species including semi-volatile organic compounds (SVOCs).

## **SUPPORTING INFORMATION**

Schematic of the experimental set-up and the inflicted irradiance by the GUV222 lamp (Figure S1), overview of performed experiments (Tables S1), description of the preparation of the surfaces, compound assignment (Table S2), additional volatile organic compound measurements (Table S3-S4 and Figure S2-S5) and ozone measurements (Figure S6).

## **ACKNOWLEDGEMENTS**

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