

Mitigation of Indoor Ozone and Secondary Products from 222 nm Germicidal UV Using Commercial Air Cleaners

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Abstract

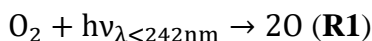
Far-UVC light, especially germicidal UV light at 222 nm (GUV₂₂₂), has received considerable attention for its potential to deactivate airborne pathogens indoors and prevent the spread of infectious disease. However, GUV₂₂₂ also generates ozone (O₃), posing human health risks and initiating additional photochemistry that may degrade indoor air quality. Air cleaners pose an opportunity to counteract the drawbacks of GUV₂₂₂ by removing harmful byproducts; however, this has never been demonstrated. Here, we conduct laboratory experiments in a 7.5 m³ Teflon

chamber using two commercially available air cleaners—a manganese-oxide-catalyst “ozone cleaner” and an activated-carbon-HEPA “volatile organic compound (VOC) + particulate matter (PM) cleaner”—in tandem with a GUV₂₂₂ lamp. We show that both cleaner types remove a wide range of chemical species, including many not specifically targeted by the cleaners. Application of chamber results to a photochemical model environment suggests that a single cleaner can achieve modest reductions in O₃ levels and substantial reductions in secondary pollutant levels within typical indoor environments. These results indicate that indoor air pollutants from GUV₂₂₂ can be mitigated through the use of air cleaning technology, thereby improving indoor air quality while maximizing the potential benefit of germicidal UV for human health.

Introduction

The COVID-19 pandemic brought substantial attention to air cleaning technologies that can be deployed for mitigating transmission of airborne pathogens in indoor environments. One notable example is germicidal UV (GUV), in which ultraviolet irradiation is used to inactivate pathogens¹; targets that can be inactivated by GUV include the microbes that cause COVID-19, measles, influenza, and tuberculosis.^{2–4} A major recent focus has been the use of far-UVC sources, particularly 222 nm KrCl excimer lamps (GUV₂₂₂).^{4,5} Light at this wavelength is safer for human exposure^{5,6} than older GUV technologies such as 254 nm light,^{4,7} enabling whole room irradiation even when the room is occupied.

One concern with GUV₂₂₂ is the potential for the formation of ozone (O₃), a major respiratory irritant. At short wavelengths (<242 nm), molecular oxygen photodissociates (**R1**), which can lead to O₃ formation (**R2**).⁸



Recent laboratory studies have demonstrated non-negligible O₃ production from GUV₂₂₂.^{9,10} In addition to O₃ itself being a health risk,¹¹ it can initiate further photochemistry, including formation of the hydroxyl radical (OH). O₃ and OH can react with alkenes in the indoor air and on indoor surfaces, leading to the formation of oxidized volatile organic compounds (OVOCs).^{12,13} Such chemistry can also produce secondary organic aerosol (SOA), including ultrafine particles, which poses a major human health hazard.^{14,15} Previous studies have demonstrated the formation of these secondary byproducts when using GUV₂₂₂.^{16–20} A simple approach for mitigating exposure to such secondary pollutants is increased ventilation;^{16,21} however, this is not always possible in all situations.

An alternative approach is the use of commercially available air cleaners to actively remove pollutants from the indoor air. Key technologies include manganese oxide catalysts to decompose O₃ to O₂^{22–24} (as well as oxidize formaldehyde (CH₂O)^{25–27}), activated carbon to adsorb gas-phase organic species and O₃,^{28,29} and HEPA filters to remove particulate matter (including SOA).³⁰ These air cleaning technologies present a promising opportunity to mitigate the negative effects of

GUV₂₂₂ by removing the various byproducts, both direct and indirect, produced by the light. However, to the best of our knowledge, this has never been investigated in a systematic way.

Here, we assess the efficacy of air cleaners in mitigating byproducts from a GUV₂₂₂ lamp within a Teflon chamber. In addition to studying the removal of O₃ generated from the lamp, we also examined the removal of secondary organic products (OVOCs, SOA) generated from the reaction of O₃ with added alkenes: limonene, a major component of cleaning products, and 6-methylhept-5-en-2-one (6-MHO), formed from the oxidation of squalene in skin oil. Because oxidative and photolytic air cleaning technologies may themselves risk the formation of byproducts,^{31,32} we focused solely on technologies based on physical removal (sorption, filtration) only. We observe substantial removal of a number of unwanted byproducts and other indoor pollutants, including many that are not directly targeted by the cleaners. Experimental results are then used to inform a photochemical model of air cleaner operation alongside GUV₂₂₂ irradiation in a typical indoor environment, to investigate the efficacy of air cleaners in mitigating harmful byproducts in more realistic indoor settings.

Materials and Methods

Experiments were carried out in a 7.5 m³ Teflon chamber, fitted with inlet ports for the introduction of chemical species and clean air flow, and outlet ports for instrument sampling.³³ Clean air was supplied via a zero-air generator (Aadco Model 737) at a total flow of 20 L min⁻¹ [equivalent to 0.16 air changes per hour (ACH)]; some of this flow first was sent through a bubbler filled with Milli-Q water to increase the humidity of the chamber. Experiments were conducted at 20 °C and 35% relative humidity in order to replicate typical indoor air conditions. A summary of experiments used in this study is provided in **Table S1**.

GUV₂₂₂ irradiation was supplied via a single KrCl excimer lamp (OSLUV Open Luminaire fitted with an Ushio Care 222 B1 emitter, peak emission at 222 nm), placed directly outside the Teflon chamber at the center of one side (the same setup as used in previous GUV₂₂₂ studies using this chamber²⁰) (**Figure S1**). The average fluence rate in the chamber was estimated to be 3.0 ± 0.2 μW cm⁻² based on chemical actinometry measurements; this is roughly consistent to (but somewhat lower than) estimates based on a geometric projection of the irradiance field (**Section S2.1**). Two air cleaners were tested: a manganese oxide catalyst-based cleaner (CDA-250, Oxidation Technologies) hereafter referred to as “ozone cleaner”, and an activated carbon- and HEPA-based cleaner (GC VOC, IQAir) hereafter referred to as “VOC + PM cleaner”. In both cases, the cleaner was placed along the bottom of the chamber in one of the corners, positioned ~0.5 m away from the walls (**Figure S1**). To insert the cleaner into the chamber, a large port on the chamber side was opened to place the cleaner in, and the port was then closed and the chamber flushed overnight. The VOC + PM cleaner was operated at a medium flow setting, corresponding to a flow rate of 61 L s⁻¹, in order to be most comparable with the measured flow rate of the ozone cleaner (59 L s⁻¹; see **Table S2** for air cleaner parameters), though there may be uncertainty in the measurements.

For all experiments, 10 ppb of a dilution tracer (hexafluorobenzene (C_6F_6), 99%, Sigma-Aldrich) was added to the chamber via injection in the clean air make-up flow line (**Figure S1**), followed by a 15-minute waiting period to allow mixing and obtain a stable signal. The subsequent steps (turning on the lamp, turning on the air cleaner, and/or adding chemical species) were then performed, in an order that varied with each type of experiment. For experiments with added VOC, either 50 ppb of (*R*)-(+)-limonene ($C_{10}H_{16}$, 97%, Sigma-Aldrich) or 50 ppb of 6-methylhept-5-en-2-one (6-MHO) ($C_8H_{14}O$, 99%, Sigma-Aldrich) was injected. For chemical actinometry experiments, 50 ppb of tetrachloroethylene (C_2Cl_4 , 99.9%, Sigma-Aldrich) was injected. For the NO_2 removal experiment, 80 ppb of NO_2 was introduced to the chamber by metering the flow from a standard cylinder (1.93 ppm, balance air, Linde). No aerosol seed or any other species were added.

Real-time measurements of gas- and particle-phase species in the chamber were conducted with a suite of analytical instrumentation, summarized in **Table S3**. Ozone was measured with a UV absorption O_3 monitor (2BTech). NO_x measurements were made with a chemiluminescence $NO-NO_2-NO_x$ analyzer (Thermo Fisher Scientific) and a cavity-attenuated phase shift NO_2 monitor (CAPS, Aerodyne Research, Inc.). A gas chromatograph with a flame ionization detector (GC-FID, SRI) was used to monitor limonene, hexafluorobenzene, and tetrachloroethylene. A modified PTR3³⁴ was run as an ammonium chemical ionization mass spectrometer (NH_4^+ -CIMS) in most experiments to provide measurements of OVOCs, including 6-MHO; in limonene removal experiments, the instrument was run as a proton-transfer-reaction mass spectrometer (PTR-MS) to measure limonene (see **Table S1** for experiment summary). Fiber laser-induced fluorescence (FILIF³⁵) was used for the measurement of CH_2O . An LI-7000 CO_2/H_2O analyzer (LI-COR) provided CO_2 measurements. Gas-phase mass spectrometric data were background-subtracted and corrected for dilution. Particle measurements included an aerosol mass spectrometer (AMS, Aerodyne Research, Inc.³⁷) for chemical composition and a scanning mobility particle sizer (SMPS, TSI) for size distributions. Particle-phase data were corrected for dilution and wall losses. Further details on data analysis and quantification approaches are described in **Section S1.1**.

Results and Discussion

Direct Emissions by the Cleaners

First, we examined direct emissions of air pollutants from the cleaners, which may themselves be sources of undesired gases: in our earlier work, we demonstrated that many commercially available air cleaners emit species such as CH_2O .³² Both cleaners studied were found to emit CO_2 and water vapor when turned on, as shown in **Figures S7 and S8**. The VOC + PM cleaner had no other apparent emissions. The ozone cleaner also emitted NO (**Figure S9**) and acetone (**Figure S10**) when turned on. The NO emissions were fitted to a rate of 0.005 ppb s^{-1} (**Figure S9**), though slight discrepancies between modeled and measured data suggest that the ozone cleaner may remove NO in addition to emitting it.

We note that other species that we cannot measure might also be emitted; for instance, alkanes are not detected by our instrument suite. Further, changes in emission rates may occur over longer

timescales than explored here. Despite these uncertainties, these measurements of direct emissions highlight the need to fully characterize air cleaners in order to better understand their implications for indoor air quality. The emissions can both initiate chemistry, as demonstrated by the NO_x chemistry from the ozone cleaner's NO emissions (Section S4.1 and Figure S11), and also may be harmful to human health. Such trade-offs must be considered to fully assess the net health benefit of the air cleaners.

Removal of Ozone Produced by 222 nm Light

The effectiveness of the air cleaners for removal of O_3 from GUV_{222} was investigated by operating the cleaners in the chamber alongside a 222 nm KrCl excimer lamp. Figure 1 shows O_3 levels when turning on first the GUV_{222} lamp and then the cleaner. Results are shown for the ozone cleaner (Figure 1a) and the VOC + PM cleaner (Figure 1b). The O_3 production rate during the lamp-on periods was measured as $25 \pm 0.2 \text{ ppb hr}^{-1}$, consistent with expectations and in line with previous studies.⁹ When either air cleaner was turned on, the O_3 dropped extremely rapidly, indicating efficient removal by the cleaners. The removal rates (determined by fitting the decrease to a simple exponential) were 0.33 min^{-1} for the ozone cleaner and 0.41 min^{-1} for the VOC + PM cleaner. We note that for the ozone cleaner, due to the NO emission, there may be some O_3 removal via $\text{NO} + \text{O}_3$ reaction. The VOC + PM cleaner, which contains activated carbon filters, was able to remove O_3 as quickly as the ozone cleaner, in agreement with previous studies demonstrating O_3 removal by activated carbon.^{29,38,39} Even with the GUV_{222} lamp on, each cleaner ensured that O_3 steady-state levels were very low ($\sim 1.2 \text{ ppb}$), only slightly higher than the lamp-off baseline ($\sim 0.7 \text{ ppb}$). As shown in Figure S6, when the cleaners were turned off, O_3 immediately resumed growing in at the previous rate. These findings demonstrate that air cleaners can indeed mitigate O_3 produced by GUV_{222} lamps when run concurrently. Air cleaners are thus a promising way to counteract O_3 generation from indoor use of GUV_{222} , though as mentioned above, trade-offs with emissions must be considered for each specific use case.

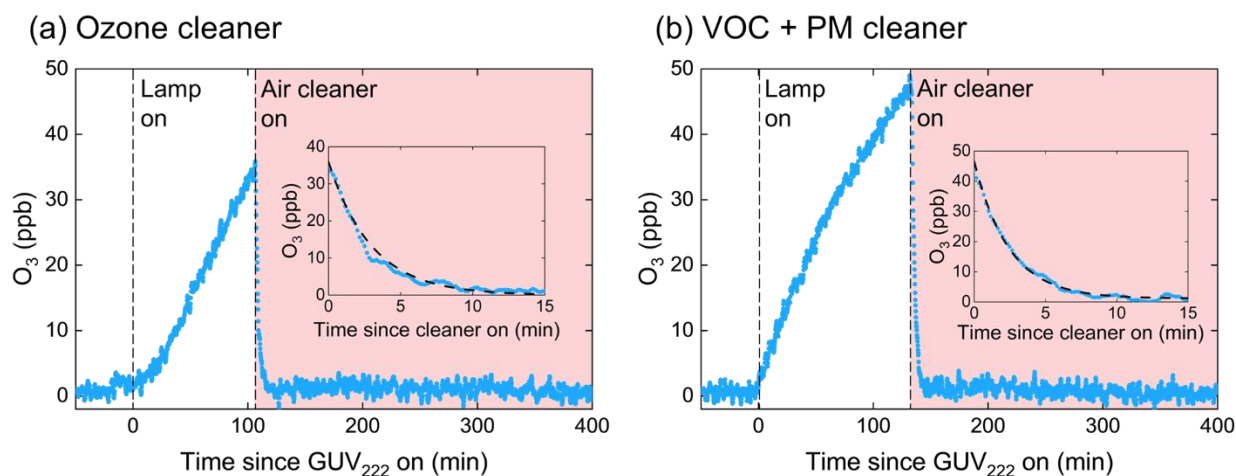


Figure 1. Time series of ozone concentration during GUV_{222} irradiation in chamber experiments with (a) the ozone cleaner and (b) the VOC + PM cleaner. Insets show first 15 minutes of the

cleaner-on periods, with dashed line marking exponential fit of the decay. Data are from Experiment 1 and Experiment 2, respectively (see **Table S1** for experiment details).

Removal of Secondary Products Formed in the Presence of VOCs

To explore the effects of air cleaners on O₃-initiated chemistry (e.g., ozonolysis of alkenes present in the indoor environment), a set of experiments were run in which limonene (a common ingredient in fragrances and cleaning products) was added to the chamber. Adding limonene and then turning on the GUV₂₂₂ lamp led to the formation of secondary products in both the gas- and particle-phase, including (but not limited to) limonaldehyde, CH₂O, and SOA (**Figure S12**). These products are consistent with the expected chemistry of limonene reacting with O₃.^{40–42} Such oxidation products have also been observed in previous experimental studies and predicted in modeling studies of limonene in the presence of GUV₂₂₂.^{16,43,17,18,20}

Both the ozone cleaner and VOC + PM cleaner were found to remove the oxidation products, as well as remove limonene (tested in separate experiments; see **Table S1**). **Figure 2** shows the removal of these species (limonene, limonaldehyde, CH₂O, and SOA) by each air cleaner, normalized by the respective species concentration when the cleaner was turned on. Gas-phase species were removed efficiently by both cleaner types. Notably, CH₂O, a major hazardous indoor air pollutant,⁴⁴ was completely removed within 10 minutes of turning on the ozone cleaner, consistent with expectations based on previous studies of CH₂O oxidation on manganese oxide catalysts. The VOC + PM cleaner reduced CH₂O concentrations by 90% within 20 minutes, and achieved complete removal on a longer time scale (**Figure S13**).

Also shown in **Figure 2** is removal of other species relevant to ozone chemistry and/or indoor air quality. Additional experiments were run with the introduction of 6-methylhept-5-en-2-one (6-MHO), a compound formed from the ozonolysis of squalene, a major component of human skin oil.⁴⁵ Both cleaners removed 6-MHO, as well as its ozonolysis products (the major product being 4-oxopentanal, C₅H₈O₂;^{46,47} see **Figure S14**), highlighting the wide range of organic compounds that air cleaning technologies can mitigate. Removal of inorganic species such as NO₂ (another indoor air pollutant) and halogenated species such as the dilution tracer, C₆F₆ (**Figure S15**), was observed as well.

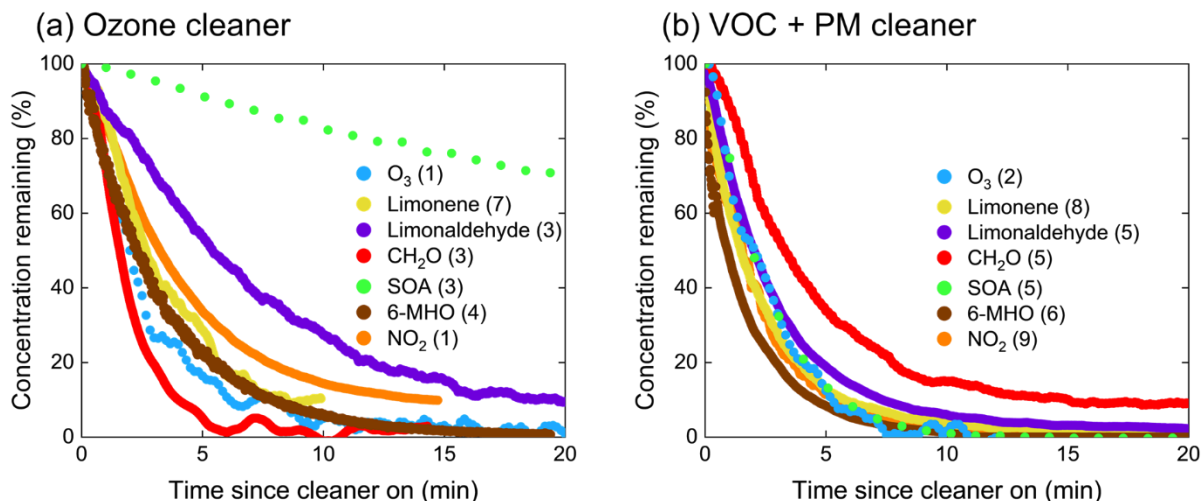


Figure 2. Comparison of cleaner removal for several species by (a) the ozone cleaner and (b) the VOC + PM cleaner. Data are corrected for dilution and wall loss, and normalized by concentration of species remaining. Numbers in parentheses signify the experiment number from which the species removal data was obtained (see **Table S1** for experiment details).

SOA removal was substantially different between the two devices, with the VOC + PM cleaner achieving ~20 times faster removal than the ozone cleaner (**Table 1**). The HEPA filter used in the VOC + PM cleaner allows for efficient removal of particles, whereas the SOA removal by the ozone cleaner occurs only via physical losses of particles within the device. Despite these vast differences in removal rates between cleaners, for each individual cleaner the rate of removal was largely independent of particle size. Both removed ultrafine (<100 nm) particles at similar rates to larger particles (**Figure S16**), indicating their ability to mitigate not only fine particulate matter but ultrafine particles (which represent a particular health hazard)¹⁵ as well.

Table 1 gives the clean air delivery rate (CADR), the rate at which a cleaner delivers air free of a given species, of each species for both cleaners. The CADRs were calculated from the baseline-corrected decay of a species, subtracting dilution and wall loss, and multiplying by chamber volume; further calculation details are given in **Section S1.2**. These CADR values are notably higher than CADRs measured for other air cleaners in our previous work,³² likely due to differences in volumetric throughput and removal technology between the various cleaners. Given the largely effective removal of particles and many gas-phase species by the air cleaners, both GUV₂₂₂ and non-GUV₂₂₂ indoor scenarios likely benefit from operation of these devices.

Species	Ozone cleaner	VOC + PM cleaner
	CADR _{species} (m ³ h ⁻¹)	CADR _{species} (m ³ h ⁻¹)
O ₃	149	184
Limonene	127	176
Limonaldehyde	56	138

CH ₂ O	291	86
SOA	9	178
6-MHO	168	213
NO ₂	86	193

Table 1. Clean air delivery rates (CADRs) of various species for the ozone cleaner and the VOC + PM cleaner.

Extrapolation to Indoor Environments

The laboratory chamber experiments described above demonstrate that air cleaners can efficiently remove a variety of species formed from GUV₂₂₂ irradiation, including both O₃ and secondary pollutants (OVOCs, SOA), as well as other species found in indoor air (other VOCs, NO₂). However, the laboratory chamber is a highly simplified system, with many factors (such as room size, ventilation, deposition, and fluence rate) differing between the chamber environment and real-world indoor environments.

Here, we applied the results of the chamber experiments to a simple photochemical model in order to investigate the implications of using air cleaners in typical indoor environments. The model, described in Barber et al.,¹⁶ characterizes key indoor air processes—VOC emissions, background O₃ in the ventilation air, loss of O₃ to the ventilation system, and O₃ deposition onto surfaces. Air cleaner removal of species was added into the existing model, using rates based on the CADRs measured in this study (see **Section S5.1** for more detailed information). The model includes two “lumped” VOCs: one that reacts only with OH, and another that reacts with both OH and O₃. All oxidation products are lumped and allowed to react with OH, and SOA is formed based on a specified yield (10%). As in Barber et al., a range of irradiation levels and ventilation rates were explored. Fluence rates spanning from 0 to 15 μW cm⁻² were used to cover a wide range of scenarios, from no GUV to high-intensity irradiation. Ventilation rates ranged from 0.1 to 10 air changes per hour (ACH), spanning common indoor values and including the minimum American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) recommendations (homes: 0.35 ACH, offices: ~2–3 ACH, and healthcare settings: 10 ACH).⁴⁸ A room size of 150 m³, equivalent to that of a small-to-medium sized classroom or office, was used to replicate a representative indoor environment in which GUV₂₂₂ might be deployed.^{49,50} Given the air flow rates of the cleaners (~216 m³ hr⁻¹, **Table S2**), their operation in a room of this size provides an increase of only ~1.44 effective air changes per hour (eACH); hence, the modeled removal of a species by a given cleaner will be substantially slower than that observed in our smaller laboratory chamber, where the cleaners provided an increase of ~28.8 eACH (**Section S5.2**).

As the ozone cleaner emits NO which may lead to NO_x chemistry, the importance of the emissions was investigated by adding NO emission into the model (using the rate measured in the chamber and scaled to the model room size). Only emission was included, with no further chemistry or loss processes besides ventilation, to provide an upper bound on the steady-state NO in the model scenarios. As shown in **Figure S18**, even in the lowest-ventilation case (0.1 ACH), the steady-state NO level from the emission is only 9 ppb. This is significantly lower than concentrations

resulting from usage of gas stoves,⁵¹ the major indoor source of NO_x. Given this, NO_x chemistry was not included in the model.

Figure 3 shows key model results. Steady-state concentrations of O₃, gas-phase oxidation products, and SOA are provided for indoor scenarios with no air cleaner (**Figure 3a**), with the ozone cleaner (**Figure 3b**), and with the VOC + PM cleaner (**Figure 3c**). The steady-state O₃ levels were reduced by 8-28% when running either cleaner type compared to the no-cleaner scenario. The O₃ concentrations in these indoor scenarios are largely governed by influx from the outdoor air via the ventilation system—hence the increasing O₃ with increasing ventilation rate—alongside O₃ production by GUV₂₂₂. Because operation of the cleaners adds up to ~1.44 eACH, there is only a small increase in fresh air delivery, and thus only modest reductions in the steady-state O₃ levels were achieved for these conditions. The model output shown in **Figure 3** is based on using one air cleaner, and so the addition of a second cleaner would provide greater reduction in O₃ levels (see **Figure S19**). Furthermore, the model operates on the assumption that the room is always well-mixed, which may not be accurate in real-world scenarios. Since there may be local areas of high O₃, particularly near the GUV₂₂₂ lamp, the placement of the air cleaner could impact its effectiveness in decreasing overall O₃ levels within the room. Real-world testing, modeling, and/or optimization of the relative placement of the two represent important areas of future work.

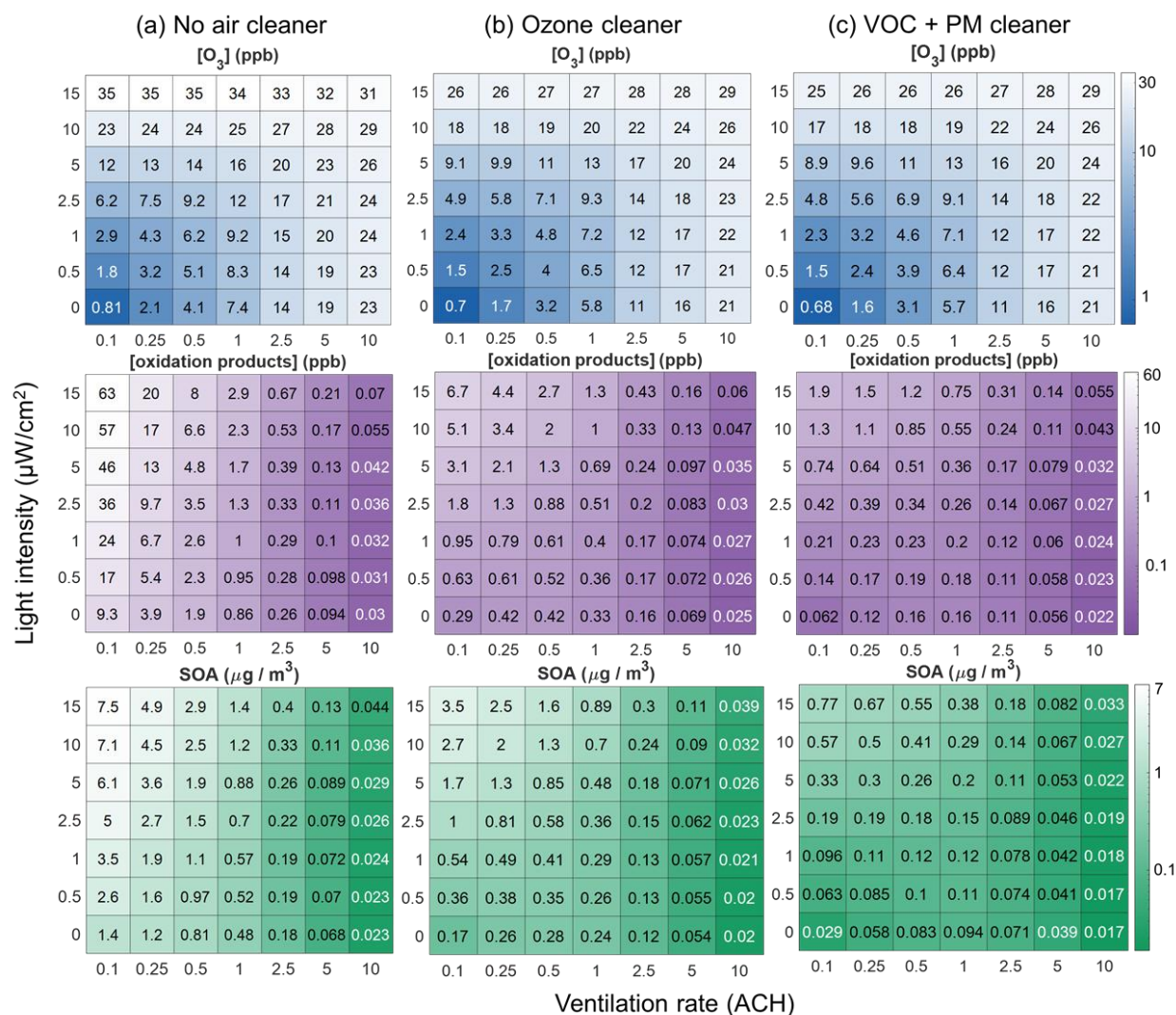


Figure 3. Effects of ventilation rate and GUV_{222} fluence rate on modeled indoor air quality across scenarios with (a) no air cleaner, (b) an ozone cleaner, and (c) a VOC + PM cleaner. Shown are steady-state concentrations of O_3 , organic oxidation products (OVOC), and SOA as predicted by the photochemical box model. Additional model results are given in **Figure S20**.

The presence of O_3 in the indoor scenarios enables OH production (model output of $[OH]$ is shown in **Figure S20**), and both of these oxidants can react with VOCs leading to the formation of gas- and particle-phase oxidation products (OVOCs and SOA, respectively). Modeled steady-state concentrations of OVOCs and SOA are shown in **Figure 3**. For these secondary products, the air cleaners were able to reduce the concentrations substantially, in some cases by an order of magnitude or more. The formation of secondary products depends on the concentrations of VOC and O_3 , and since both reactants are removed by the cleaners, there is a multiplicative decrease in the product formation rate. Additionally, the products themselves are also removed by the cleaners, leading to an even greater decrease in steady-state concentrations. Hence, the net effect of the air

cleaners is a dramatic reduction of secondary products, despite the relatively low eACH increase; the benefit is especially pertinent for the low-ventilation cases.

Implications

The power of air cleaners for removal of indoor pollutants suggests that they can be beneficial in minimizing O₃-related risks associated with indoor use of GUV₂₂₂, particularly under low-ventilation conditions. They can also be useful for improving indoor air quality in non-GUV₂₂₂ settings: in the non-irradiated (0 μW cm⁻²) scenarios, operation of the cleaners reduced OVOC and SOA concentrations by up to 97% (depending on ventilation). Though direct emissions by the cleaners themselves may be of concern and require greater characterization, these results demonstrate the general efficacy of commercially available sorption- and filtration-based air cleaners for a variety of indoor environments and irradiation levels.

Any indoor use of GUV₂₂₂ must consider the trade-offs between germicidal benefits and air quality disbenefits. Though a detailed analysis of these trade-offs is beyond the scope of this work, we show that air cleaner removal of pollutants can potentially shift the trade-off substantially, by improving air quality and thus increasing the range of scenarios where GUV₂₂₂ could provide a net health benefit. Uncertainties remain, including the longevity of air cleaner efficacy and changes in emissions over extended periods of use, as well as the effect of the placement of cleaners within a room. These are critical areas of future work in understanding and optimizing the use of air cleaners, with or without GUV₂₂₂, to promote healthier indoor environments.

Supporting Information

Experimental methods and procedures; additional results and discussion (including CADR and fluence rate calculations; air cleaner emissions; time series of precursors and oxidation products; and SMPS data); model details and results (including additional model output figures and calculation of effective air changes per hour)

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