We appreciate the time and effort that the reviewers have dedicated to providing your valuable feedback on our manuscript. The reviewers' comments are all very helpful for revising and improving our manuscript. The point-by-point response below address all the points raised by the reviewers. Our responses are shown in **blue**, deleted text in **red**, and added text in **gold**. Moreover, an additional copy of the revised manuscript with tracked changes (Manuscript with tracked changes_Fu.pdf) is attached, which shows all the changes by using the yellow highlight for additions and red strikethrough font for deletions.

Comments from Review 1 (RC1)

General Remarks

The paper deals with the possibility of determining the UV-radiation dose on airborne materials using dye. During the study, an impactor was used to collect coloured DEHS aerosol. The use of a non-evaporating liquid allows a continious probe sampling and a uniform colour distribution for soluble dyes. The aerosols produced were analysed before and after UV-light illumination. A variety measuring instruments were used during this study, such as a SMPS, TEOM and Impactor. A spectrophotometer was used to determine dye degradation. Whilst claiming to have a feasible method for quantification, this paper offers no theory/calculation to determine quantitative results and compares inadequately with previous studies. A more comprehensible description on the method of determining the degradation of the dye would be helpful.

This manuscript needs additional details, clarification and further explanation to be suitable for publication in AR

We genuinely value your commitment in providing your time and effort to provide insightful suggestions and constructive feedback. Based on your detailed report, we have incorporated additional data and discussions, alongside providing more comprehensive experimental details to enhance both comprehension and clarity. Although our ability to develop a mathematical model or relationship between dye degradation under UVC irradiation was constrained by limited experimental data, we have made enhancements to Section 3.3 to provide a more thorough explanation.

"Two potential mathematical models that might offer insights into dye degradation: the classical exponential decay model (log-linear decay) and the shoulder model, which begins with a horizontal slope before transitioning into full exponential decay (Kowalski, 2009). However, the determination of which model is better suited for the selected dye solution remains inconclusive due to limited experimental data. A dataset that includes 90% dye degradation induced by UV radiation is, at a minimum, required."

Specific Questions and Comments

1. Normalisation would be useful to improve comparability. This would use the power of the light source, the irradiated volume and averaged over the aerosol size. The CMD (count median diameter) and GSD (global standard deviation) of the total aerosol population or that collected by the impactor should be used. The distance between LED and central aerosol flow would also be helpful as well.

Thank you for pointing this out. In response to the suggestion for normalization, we have included a detailed schematic (Figure 3b) that clearly illustrates the radiant power of the UVC LEDs, the effectively irradiated volume, and the distance between the LED and the central aerosol flow. The corresponding text in Section 2.2.2 and table 2 have been revised to provide comprehensive insight into the experimental setup.

"The quartz tube, with a length of 500 mm, a wall thickness of 2.5 mm, and an internal diameter of 60 mm, was purchased from GVB GmbH - Solutions in Glass (Herzogenrath, Germany)….

The distance, denoted as h, between the LED and the central aerosol flow measures 45 mm….

The slanted shaded region in Figure 3b illustrates the calculated effective irradiated volume V ($V = \pi \cdot (D/2)^2 \cdot L$) of the UV chamber."

Additionally, Table 4 has been added to summarize the calculated Count Median Diameter (CMD) and the Geometric Standard Deviation (GSD) of the aerosol populations as measured by the electrical low-pressure impactor (ELPI) and scanning mobility particle sizer (SMPS). We have deleted one sentence and revised the corresponding section to discuss the particle size distribution at various operating pressures more precisely (Section 3.1.2).

Changed from:“ ~~while the particle size distribution remained relatively unchanged~~.”

To:

“Table 4 presents a summary of the calculated count median diameter (CMD) and the geometric standard deviation (GSD) of the total aerosol population, as measured by ELPI and SMPS. While ELPI measurements show CMD values ranging from 505 nm to 639 nm, with GSD decreasing from 1.76 to 1.35 as pressure increases. SMPS results indicate consistently lower CMD values between 297 nm and 305 nm, with a similar downward trend in GSD from 2.05 to 1.81. This discrepancy could be due to the different measurement principles and sensitivities of these measure instruments.”

2. The study mentions that DEHS is used as UV-VIS spectrum baseline. Please also show this as a graph in the study.

Based on the comment of the reviewer, we have incorporated a new figure (Figure 6 in the revised manuscript) that presents the UV-Vis spectra of water and DEHS, using air as the baseline. This figure also displays baselines obtained from measurements with different batches of DEHS. Additionally, the necessity of employing a DEHS baseline and the potential errors introduced by subtracting this baseline are discussed in the revised manuscript (Section 3.1.1).

“As depicted in figure 6a, while pure water has no absorption upon UV-Vis light, the chosen carrier liquid, Di-ethyl-hexyl separate (DEHS), significantly absorbs UV light with wavelengths below 300 nm. Since the absolute absorbance of DEHS below 270 nm exceeds 1, the UV-visible spectra ranging from 270 to 800 nm were utilized as the baseline for measuring the spectra of pure dyes in this study. Variations in dye concentration were identified through characteristic absorption peaks within the visible spectrum.”

3.DEHS eliminates the evaporating complexity. But water solved dye could be dried, leaving just dye behind, without chemical interactions. Elsewise; high humidity levels reduces the UV-induced inactivation capacity (Peccia et al. 2001). Please discuss this.

The research conducted by Peccia et al. focused on the impact of relative humidity (RH) on the inactivation of bacteria by UV radiation. The study found that variations in ultraviolet spherical irradiance were minimal and not statistically significant across a range of 20-95% RH. However, a notable observation was the decrease in the UV-induced inactivation rate as RH increased, particularly beyond 50%. This decrease is hypothesized to be linked to the increased water absorption by bacterial cells at higher humidity levels. The study suggests that hydration and rehydration of these cells could alter protein structures, potentially affecting DNA repair processes or the extent of UV inactivation.

To simplify the study and avoid the complexities associated with droplet evaporation and the use of microorganisms, DEHS (Di-2-ethylhexyl sebacate) was chosen as the carrier liquid, along with a DEHS-solved dye to develop a model system. We agree the limitation of chemical dyes in simulating potential alterations in bacterial cell protein structures due to water absorption upon varying relative humidity (RH). However, this model system still offers valuable insights when using DEHS aerosols. This is particularly relevant in understanding how the suspending medium and aerosol droplet size influence the required UV dose for effective dye degradation.

We added several sentences to illustrate this (Section 2.1).

Changed from: “~~allowing for accurate online measurements~~.”

To:

“In aerosol science, DEHS is widely used to generate liquid particles with extended lifetimes due to its extremely low saturation vapor pressure. This quality is crucial for studying particle size effects (Ren et al., 2021; Li et al., 2020). Besides, DEHS aerosols, recommended for aerosol filtration testing, allow for more accurate particle size measurement of spherical liquid particles compared to non-spherical solid particles like salt and test dusts, which tend to agglomerate (Gustavsson, 2003).”

4. Instrumentation: Please also provide the exact specifications for the measuring devices used (Cut-Off Diameters for Impactor; Flow Rates for DMA just to name a few). The SMPS; Model 3938 includes the possibility to determine the size distribution and number concentrations by CPC (condensation particle counter) or OPC (Optical Particle Counter), are both used?.

Thank you for your suggestion. Based on this, we have added the calculated cutoff Stokes diameter for the designed impactor (Section 2.2.1). We have summarized the calculations in Appendix A. Additionally, we have included several sentences to clarify the specific model of the SMPS device, including the models of the neutralizer, DMA, and CPC. The flow rates for the applied DMA have also been added for further clarification (Section 2.2.3).

"The estimated cutoff Stokes diameter d_{50} for the impactor at various inlet gas flows q of 0.78 L/min, 2.26 L/min, and 8.86 L/min is approximately 1.28 µm, 0.75 µm, and 0.38 µm, respectively. For more detailed information on the calculations, please refer to Appendix A."

"The SMPS device comprises three main components: an aerosol neutralizer (TSI Model 3088), a differential mobility analyzer (DMA, TSI Model 3081) and a condensation particle counter (CPC, TSI Model 3775). In this setup, the DMA employed closed-loop system for the sheath flow, whereas the aerosol flow through the entire SMPS system was regulated by the CPC, which maintained at a low flow rate of 0.3 slm (standard liter per minute). The sheath flow rate for the DMA was set to 3 slm to achieve an aerosol to sheath flow ratio of 1: 10. In the ELPI, particles are classified into 14 size fractions using a cascade impactor. The cutoff aerodynamic sizes are 0.016 µm, 0.03 µm, 0.053 µm, 0.093 µm, 0.15 µm, 0.26 µm, 0.38 µm, 0.60 µm, 0.95 µm, 1.6 µm, 2.5 µm, 3.7 µm, 5.4 µm, and 10.0 µm, respectively. Additionally, to account for the influence of multiply charged particles on the measured signal, a multiple charge correction was used to the measurements obtained by both ELPI and SMPS."

In this study, we used only the CPC to determine the aerosol particle number concentration. We did not employ the available OPC instrumentation (Aerodynamic Particle Sizer, TSI Model 3321) in our lab, as it cannot distinguish aerosol particles below 0.5 µm. Therefore, OPC was not utilized to measure the particle number concentration. We apologize for any confusion this may have caused.

5. The use of an impactor allows the collection of larger aerosol particles. With sufficiently large droplets, shielding would be possible, preventing the dissolved dye inside (core) the aerosol from interacting with the radiation.

Yes, we acknowledge the potential shielding effect of large droplets on the dissolved dyes within the droplet particles. We have added the text to point out that a detailed study of the impact of aerosol particle size on UV-induced inactivation rates is warranted (Section 3.3). Our current study is primarily focused on assessing the feasibility of determining the UV radiation dose experienced by non-bioaerosols. In our subsequent research, we plan to classify aerosol particles and design various impactors for their collection. This will enable us to investigate the shielding effect caused by large droplets more thoroughly.

"Another possible cause for the observed nonlinear degradation might be the different degradation responses in droplets of varying diameters, resulting from the variable gas flow to the impactor, which in turn leads to different cutoff sizes for the collected droplets. Future research should also focus on optimizing the impactor to separately collect particles from different categories in order to explore potential particle size effects."

6. In Introduction; add more information on possible tracer methods (fluorescence or radiation (Talaat et al. 2021))

Accordingly, we incorporated additional information on various tracer techniques which are employed across a range of scientific and medical fields. This includes fluorescence tracing, radioactive tracing and chemical tracing techniques.

"Tracer methods have been widely used in various scientific and medical disciplines to study biochemical and biophysical processes. The tracer is chosen so that it behaves like the substance being studied but can be easily detected. For instance, fluorescence tracing has become powerful tools in biology and biochemistry for imaging cells and tissues, and tracking the movement of molecules within organisms (Kyrychenko, 2015). Radioactive tracing is particularly useful in medicine, where radioactive tracers (Talaat et al., 2019) are used to image body tissues and organs, highlighting areas of high metabolic activity. Recently, Talaat et al. (Talaat et al., 2019) developed a model to numerically assess the radiation dosimetry of inhaled radioactive aerosols, by coupling computational fluid-particle dynamics (CFPD) and the Monta Carlo (MC) methods. ~~In various studies~~ Furthermore, chemical tracers, such as UV-sensitive dyes, …"

7. Please add the chemical structures of your final chromophores (#4 and #7). Are those used in prior studies (Putt et al.)?

For our study, we utilized a UV-sensitive dye provided by the Risk Reactor company. These dyes are characterized by their oil solubility and are complex, fluorescent-based products. However, the Risk Reactor company was

unable to share the chemical structure of these dyes with us. In the earlier research conducted by Putt et al., the UV-sensitive dyes #4 and #7 were not utilized, as their investigation was concentrated on dyes that dissolve in water, rather than those like DEHS, which are known for their solubility in oily substances.

8. Missing data from UV-Vis spectra of aerosol droplets exposed to UV-light. Please show a graph with DEHS without UV exposure and DEHS with UV; together with dye-loaded DEHS with and without UV exposure, to show the amount of degradation. (One example, one concentration, one dye, is sufficient.

In the revised manuscript, we added the UV-Vis spectra of collected aerosol droplets of both pure DEHS and dye solution #4 (10 µg·mL-1) before and after UV exposure (See Figure 13b in the revised manuscript). Additionally, we have revised the relevant sections to discuss the error resulting more thoroughly from using DEHS as the spectral baseline in determining the degradation of the dye (Section 3.3).

"In addition, Figure 13b presents the UV-Vis spectra of collected aerosol droplets of both pure DEHS and dye solution #4 (10 µg·mL^{-1}) before and after UV exposure. The degradation of dye #4 after UV exposure is evidenced by noticeable changes in the maximum peak absorbance (specially at 497 nm for dye #4). While pure DEHS exhibits some spectral changes under UV irradiation, these *changes* are predominantly below 300 nm wavelength, with insignificant absorption changes above 400 nm. Therefore, the error introduced by using DEHS as the spectral baseline for determining dye degradation is considered negligible."

9. Missing approaches/estimations (qualitative and quantitative) between UV-lethality doses on viruses and UV-radiation to which the aerosols are exposed to. Provide quantitative information on your methods. Compare with prior studies (e.g. Putt et al. 2012).

The constrained capacity of our existing UV irradiation chamber resulted in limited data collection, leading to only a 10% degradation in dye concentration. Due to this restricted dataset, it's not feasible to formulate a mathematical model or draw comparisons with the virus inactivation dose, which is typically measured in terms of log-reduction in virus concentration. However, we added several sentences to discuss the similarity and difference of the measured UVC irradiation dose for dye solution (not suitable for aerosols) with previous studies (Putt et al., 2012), and additional conclusions were included (Section 3.3).

"While pure DEHS demonstrates significant absorbance under UVC irradiation, as shown in Figure 6a, dyes dissolved in DEHS undergo photodegradation upon UVC exposure, similar to that observed in water-dissolved dyes (Putt, et al. 2012). This behavior of dyes in DEHS under UVC irradiation might closely mimic the response of pathogens in saliva, where proteins also absorb UVC light."

Major Comments

Line 33-39; references to other methods that deviate from the topic are too one-sidedly negative

Adjustments have been made to ensure the neutrality of the description regarding other methods.

"The rapid and groundbreaking advancement in COVID-19 vaccine development by researchers has played a pivotal role in significantly reducing the global impact of the pandemic (2023 Chakraborty et al.).
….,which could reduce the efficacy of the developed vaccines (Kaur and Gupta, 2020; Van Dorp et al., 2020) ~~allowing the virus to evade the immune system even after vaccination~~."

Line 44; Citation missing for the statement "UV radiation presents a more environmentally friendly and energy-efficient alternative to liquid disinfectants and heat disinfection for sterilizing liquids, air, and surfaces

Thank you for pointing this out. We added the corresponding citation in the revised manuscript.

Line 54; Power of radiation device is given for an area. For comparability, it would also be important to know at what distance from the source to the object this applies.

The data provided here represent the radiation doses (mW·s·cm^{-2}) required to inactivate 90% of the virus concentration (the log-reduction doses). The radiation dose is calculated as the product of radiation intensity (mW·cm^{-2}), which varies according to the distance between the sample and the light source (s), and the exposure time. Notably, some studies did not provide the distance between the light source and the sample, providing instead

the measured radiation intensity ($mW \cdot cm^{-2}$) at the sample position. For better understanding and comparison, we have included available data regarding the radiation distance.

"For instance, even within the 254 nm results, the UV radiation doses required to inactivate 90% of the virus concentration (the log-reduction doses) ranges widely~~it is 0.6 $mW \cdot s \cdot cm^{-2}$ for bovine coronavirus while for SARS (CoV Urbani), it's as high as 11,754 $mW \cdot s \cdot cm^{-2}$~~ (Heßling et al., 2020). For the removal of bovine coronavirus using a UV24 unit (with an airflow of 85 m^3/h and a produced UV dose of 19.8 $mW \cdot s \cdot cm^{-2}$), the required dose is 0.6 $mW \cdot s \cdot cm^{-2}$ in a room of 244 m^3 volume without outside air (Kowalski, 2017). In the case of SARS-CoV (Urbani strain), it's as high as 11,754 $mW \cdot s \cdot cm^{-2}$ by applying a UVC light source of 4.0 $mW \cdot cm^{-2}$ at a distance of 3 cm to the sample (Darnell and Taylor, 2006)."

Line 65; Citation needed for the statement:" experiments involving pathogenic microorganisms due to the stringent requirement of biosafety laboratory" - Statutory text or the Ordinance on Industrial Safety and Health would be appropriate here

We added two citations, which summarizes the biosafety guidelines for working with pathogenic and infectious microorganisms and provides resources for more information.

Line 71; Citation needed for the statement:" …as been used to measure the radiation doses of UV light with a wavelength of 254 nm, serving as chemical indicators for UV sterilization processes"

In the revised manuscript, we have included the relevant citation.

Line 114; sedimentation is mentioned as an exclusion criterion. The uniform distribution within the aerosol is important here.

We have added one sentence to clarify and emphasize the importance of uniform dye distribution in aerosol droplets.

"For uniform dye distribution in the aerosol, it's essential to use a DEHS solution that reliably maintains a stable dye concentration, a key factor in producing consistent and stable aerosol particles."

Achieving this uniformity hinges on using a DEHS solution that consistently maintains a stable dye concentration over extended periods. If sedimentation is observed, it suggests poor or minimal solubility of the dye in DEHS. Thus, we avoid using such solutions with sedimentation to ensure we only utilize stable solutions, which are crucial for the continuous production of uniform aerosol particles.

Line 117; Please elaborate further on the similarities between the colour and DNA/RNA

Accordingly, we revised this sentence to demonstrate that dyes with an absorption peak near 260 nm were selected for further study.

"Given the varying susceptibility of different UV-sensitive dyes to UVC exposure, dyes with an absorption peak near 260 nm (below 300 nm in this study) were selected for further study. This is because the chemical structure, crucial for the replication of deoxyribonucleic acid (DNA) and ribonucleic acid (RNA), is more sensitive to UV irradiation near 260 nm, where it also exhibits an absorption peak (Abkar et al., 2022). ~~those with UVC susceptibility similar to deoxyribonucleic acid (DNA) or ribonucleic acid (RNA), were identified for further investigation.~~"

Table 1; Please add peak wavelength details

Peak wavelengths details were added in Table 1.

Line 140f; Please add more details of the LPI (Tube lengths; Flow rates, Distance between nozzle and impact plate; cut-off diameter)

We have added the requested details of the designed low-pressure impactor in the revised manuscript.

"The distance z between the collecting substrate and the aerosol outlet was 5 mm, and the tube length of circular jet tube was approximately 20 mm. The estimated cutoff Stokes diameter d_{50} for the impactor at various inlet gas flows q of 0.78 L/min, 2.26 L/min, and 8.86 L/min is approximately 1.28 µm, 0.75 µm, and 0.38 µm, respectively. For more detailed information on the calculations, please refer to Appendix A."

Line 163: Please add picture of the 3d-printed object or refer to figure 2

We have included a figure (Figure 2a) to display the 3D-printed fixture used for holding the quartz cuvette and UV LEDs.

Line 184; Please add length of the quartz tube or refer to table 2; Please add estimations of sedimentation losses

Details about the quartz tube have been added: "The quartz tube, with a length of 500 mm, a wall thickness of 2.5 mm, and an internal diameter of 60 mm, was purchased from GVB GmbH - Solutions in Glass (Herzogenrath, Germany)."

We agree that droplet sedimentation can occur when prolonging the particle residence time within the quartz tube. In this work, we did not account for sedimentation losses, due to the limited sampling time and the small size of the produced aerosols ($d_{p,\,max}$ = 2 µm). However, this issue raises a compelling question that we may explore in our further studies.

Line 190; add brief function description of the cyclone

A brief function description of the cyclone has been included in the revised manuscript: "Due to centrifugal force, the cyclone can efficiently separate larger particles generated from the nebulizer. Heavier particles (particles with an aerodynamic diameter > 2 µm) are collected in a liquid reservoir, while the lighter particles are released with the gas flow."

Line 194-198: Add more details for the instrumentations (flow rates, cut-off sizes)

We have added several sentences detailing the specific model and flow rates of the SMPS device. Additionally, the flow rates for the online instrumentation used are illustrated in Figure 4. Furthermore, the cutoff sizes of the ELPI are now included in the manuscript.

"The SMPS device comprises three main components: an aerosol neutralizer (TSI Model 3088), a differential mobility analyzer (DMA, TSI Model 3081) and a condensation particle counter (CPC, TSI Model 3775). In this setup, the DMA employed closed-loop system for the sheath flow, whereas the aerosol flow through the entire SMPS system was regulated by the CPC, which maintained at a low flow rate of 0.3 slm (standard liter per minute). The sheath flow rate for the DMA was set to 3 slm to achieve an aerosol to sheath flow ratio of 1: 10. In the ELPI, particles are classified into 14 size fractions using a cascade impactor. The cutoff aerodynamic sizes are 0.016 µm, 0.03 µm, 0.053 µm, 0.093 µm, 0.15 µm, 0.26 µm, 0.38 µm, 0.60 µm, 0.95 µm, 1.6 µm, 2.5 µm, 3.7 µm, 5.4 µm, and 10.0 µm, respectively. Additionally, to account for the influence of multiply charged particles on the measured signal, a multiple charge correction was used to the measurements obtained by both ELPI and SMPS."

Line 198: Add citation for the statement: "This dilution technique did not significantly alter the particle size distribution"

We have included the relevant citation to support this statement.

Figure 4; flow rate consistency inside your figure

Figure 4 has been revised to ensure consistency in the flow rate representation.

Line 206: Please add citation and another point of view, like: experiments with pathogenic microorganisms for aerosol studies are often simulated with smoke aerosols (Chen et al. 2021)

We have added the relative citations and included another point of view, as suggested by the reviewer.

"Some researchers, therefore, use smoke aerosols to simulate pathogenic microorganism in their aerosol studies (Chen et al., 2021)."

Line 207: with "stable aerosol droplets" is meant; with a low vapour pressure /nearly non evaporating aerosol (Liqiao et al. 2020)

Thank you for bringing this to our attention. We have modified the sentence to eliminate confusion for readers: "…due to its extremely low saturation vapor pressure (Li et al., 2020)."

Line 208; Do you mean Suspension?

No, we are referring to a solution. A suspension is defined as a heterogeneous mixture where solid particles are dispersed throughout a liquid without dissolving. In our work, we discuss a dye solution, indicating that the dyes are homogeneously dissolved in the DEHS solvent. We have revised this sentence for clearer understanding with "…, the ability of one substance to dissolve in a solvent,"

Line 210; Dye concentration of 100 ug/mL; Why? Virus load comparable, comparability for later UV-VIS spectra; low noise to signal ratio?

We selected dye concentration of 100 ug/mL to align with studies by Putt, who investigated the degradation of chromophores in water solutions for determining UV light radiation doses.

We added the following text: "aligning with the concentration used by Putt et al. (Putt et al., 2012), who studied chromophore degradation in water solutions for evaluating UV radiation doses."

Line 215; Table 3: Add Type of dye and colour/ absorption wavelength peak

The type of dye and its absorption peak wavelength in the visible region have been added in table 3.

Line 221: Baseline with DEHS: Please add a figure of the pure DEHS spectra; and error estimations for the subtraction method

A figure (Figure 6 in revised manuscript) presenting the UV-Vis spectra of water and DEHS, with air as the baseline, has been included. The potential errors arising from subtracting this baseline are discussed in the revised manuscript.

"Figure 6b shows three DEHS baselines obtained by various batches of liquid. As the absorbance fluctuations are smaller than ± 0.001, the error caused by subtracting this baseline should be smaller than 1 %."

Line 223; "undergo chemical reactions" observed by? Caused by (chromophore frame alterations)

We revised this sentence for better clarity and added a citation to support the explanation that potential alterations in the chromophore structure might occur.

"…, as indicated by ~~broad absorption peaks~~ their UV-Vis spectra. These spectra show absorption across the whole visible wavelength spectrum and the absorption peaks appear broadened and obscured. For a given organic chromophore, the absorption peaks or shoulders are common for the presence of the conjugated systems and might become broad when extensive conjugation occur with the solvent dependent (Hemdan, 2023; Zheng et al., 2018)."

Line 216; Line 225-229; Desired Absorption Range of around 260 nm; Please refer to Fig 6; AND Where is the Data for 260 nm Wavelength? UV-VIS Spectra goes hardly below 270 nm wavelength.

Figure 6a demonstrates that Di-ethyl-hexyl sebacate (DEHS) significantly absorbs UV light at wavelengths below 300 nm. Given that DEHS's absolute absorbance exceeds 1 below 270 nm, we utilized its UV-Vis spectra from 270 to 800 nm as the baseline for measuring the spectra of pure dyes. The spectrum below 270 nm was excluded due to substantial baseline fluctuations. In this research, dyes that showed pronounced absorption properties below 300 nm, which we assume to be around 260 nm, as depicted in Figure 7 Group C, were considered desirable for our purposes.

We added the following sentence for clarity: "Since the maximum absorbance of DNA/RNA is near 260 nm when apply UVC light above 240 nm, dyes with obvious absorbance below 300 nm, which we assume to be around 260 nm, are desired."

Line 240: change quantify to estimate; or elaborate the process with uncertainties.

The word 'quantify' was changed to estimate.

Figure 6; Desired Wavelength (260nm) is not represented here. At least one picture with higher resolution at desired wavelengths.; Add a statement for the baseline subtraction in the picture description, if this is not raw data

As previously clarified about the added UV-Vis Spectra of DEHS in Figure 6, we utilized DEHS's UV-Vis spectra from 270 to 800 nm as the baseline for measuring the spectra of pure dyes, since its absolute absorbance exceeds 1 below 270 nm. Due to significant baseline fluctuations, the spectrum below 270 nm was excluded. We have included a statement and discussion about this baseline subtraction.

"Variations in dye concentration were identified through characteristic absorption peaks within the visible spectrum. Figure 6b shows three DEHS baselines obtained by various batches of liquid. As the absorbance fluctuations are smaller than ± 0.001, the error caused by subtracting this baseline should be smaller than 1 %.....

…absorption characteristics near 260 nm (below 300 nm in this study) and sharp absorption peaks in the visible region."

Figure 7; Absorbance higher 1? Absorbance is only relatable between 0.2 and 0.8. ; Colors are hardly distinguishable

Yes, the absorbance peaks near 1 in the visible region at a maximum concentration of 20 $\mu g \cdot mL^{-1}$. Typically, absorbance values between 0.2 and 0.8 are more precise, and we appreciate you highlighting this. We plan to adjust the sample concentration values in our future research accordingly. Since we selected dye #4 for our study and produced aerosols at a concentration of 10 $\mu g \cdot mL^{-1}$ with UV radiation, this does not impact the results of our current study. Furthermore, we have adjusted the color scheme of this figure (Figure 8 in revised manuscript) to enhance its readability.

Line 248; "smaller particle sized aerosols" Please refer to your aimed CMD of your aerosol size distribution

The aimed CMD "($d_{CMD} < 1$ µm)" has been included.

Line 252: Please add information on what the mobility or stokes diameter is based on

The measurement principles of SMPS and ELPI have been added with the following sentence: "The SMPS measurement is based on the electrical mobility of particles, whereas ELPI relies on the aerodynamic properties of the particles."

Line 255: Please refer to fig 8 for the statement "size distribution remained relatively unchanged"

We added the CMD and GSD in Table 4 and corresponding text to support the statement "size distribution remained relatively unchanged".

"Table 4 presents a summary of the calculated count median diameter (CMD) and the geometric standard deviation (GSD) of the total aerosol population, as measured by ELPI and SMPS. While ELPI measurements show CMD values ranging from 505 nm to 639 nm, with GSD decreasing from 1.76 to 1.35 as pressure increases. SMPS results indicate consistently lower CMD values between 297 nm and 305 nm, with a similar downward trend in GSD from 2.05 to 1.81. This discrepancy could be due to the different measurement principles and sensitivities of these measure instruments. ~~while the particle size distribution remained relatively unchanged.~~ Regardless of the operating pressure, ~~T~~the generated aerosol droplets exhibited a size distribution similar to that of most respiratory droplets, with a large fraction is smaller than 1 µm and a peak around 0.2 to 0.8 µm (Morawska et al., 2009; Zayas et al., 2012; Fabian et al., 2011)."

Line 269: “slight fluctuation in the number concentration is likely due to the instability of the aerosol generator” – If you refer to the “jumps” between 200 and 300 nm it is most likely, this happens due to the change of the measurement system (with internal corrections applied). It looks like the SMPS change its number concentration measurement from an OPC (from 1000-300 nm; straight line”) to a CPC(more precisely) . Please state the operation ranges and flow rates of the DMA (if those changes as well)

The 'slight fluctuation' refers to the variation in particle number concentration, which can be attributed to factors inherent to the aerosol generator, such as the stability of pre-pressure regulation. As previously stated, we did not use an Optical Particle Counter (OPC) for our measurements. The 'jumps' observed between 200 and 300 nm are most likely due to the application of a multiple charge correction, which compensates for the influence of multiply charged particles on the measured signal.

“the slight fluctuation in the number concentration is likely due to ~~the instability of the aerosol generator~~ factors inherent to the commercial aerosol generator, such as the instability of pre-pressure regulation.”

Line 274: Please add a citation for statement “… size can change due to evaporation during the sampling process…”

In the revised manuscript, we added a citation for supporting this statement.

Line 279; “collected … for one hour”; Are there fluctuation in the aerosol generation? Is the Complete Aerosol size distribution used, or a fraction (DMA selected?)? If so, please describe.

Yes, we collected the aerosol droplets for one hour to guarantee a sufficient liquid volume (at least 80 µL for the submicron quartz cuvette) for UV-Vis measurements. As mentioned, the fluctuation in particle number concentration is attributed to the commercial aerosol generator. During the collection of droplets, the entire aerosol size distribution was utilized.

Line 281; “… mass output of the aerosol generator to the collected liquid…” How was the mass output determined? Please add a statement of the 10% missing aerosol mass is from lower droplet sizes, that are not “collected” by the impactor.

The mass output of the aerosol generator was determined by directly measuring the aerosol mass concentration online using a commercial TEOM device. A statement about the 10% missing aerosols has also been included.

“The mass output of the aerosol generator was determined by directly measuring the aerosol mass concentration online using a commercial TEOM device, and this data was then used to estimate the collection efficiency of the LPI. Concurrently, aerosol droplets were collected using the impactor at a defined aerosol flow rate (8.8 $L \cdot min^{-1}$) for one hour, ensuring an adequate liquid volume (minimal 80 µL for the submicron quartz cuvette) for UV-Vis measurements….The missing 10% of aerosol mass is likely from smaller droplets not collected by the impactor, given that the estimated cutoff size (Stokes diameter) d_{50} of the impactor at a flow rate of 8.8 L/min is 0.38 µm”

Figure 10; red points does not follow the curve! Discuss; With standard curve, you mean: calculated by lambert-beer law and extinction coefficient? – If so, please add this statement in the figure description.

The standard curve was statistically determined through regression analysis of five concentrations, as illustrated in Figure 8 in revised manuscript. The corresponding discussion has been revised for better clarity.

“~~This observation could potentially be explained by the dependence of the dye concentration in a droplet on the size of the droplet.~~This observation might be attributed to the differing concentrations within individual droplets, a hypothesis that can be further substantiated by collecting categorized droplets with varying diameters.”

Line 285; “… dependence of the dye concentration in the droplet…” The deviation is most likely be caused by the presence of stray radiation within the sample, resulting in a negative deviation from Beer´2 Law. There is a text by Thomas Wenzel at libretexts.org describing the Beer´s law in detail.

Thank you for providing another possible cause of concentration deviation at high concentrations. In our manuscript, we have only suggested one possible explanation. The exact cause will be explored through our further research.

Line 285; “theoretically calculated” – Please add and refer to an equation

We apologize for any confusion caused and have rephrased the sentence for improved clarity.

“~~Both the measured and theoretically calculated results revealed that the maximal collection volume of DEHS droplets, using the aerosol generator AGF 2.0 at a pressure of 1.5 bar, was at 96 µL·h^{-1}.~~ Using the AGF 2.0 aerosol generator at 1.5 bar, the mass output m, as measured by the TEOM instrument, is 88 mg·h^{-1}. Consequently, the maximum collection volume V of DEHS droplets, calculated as $V = m / \rho_P$ (with ρ_P=0.912 g·cm^{-3}), amounts to 96 µL·h^{-1}.”

Line 287; Can you add an estimated error for the consumption rate?

Here, we provide an estimate of the maximum droplet volume that can be collected, based on the average mass concentration measured online over one hour using the commercial TEOM device. The consumption rate of the aerosol generator was not considered in this estimation.

Line 295-298; Missing citation

The citations were added in the revised manuscript.

Line 304; …”prior study” please refer or use a citation

Accordingly, a citation has been included.

Line 308-310; Please elaborate further and describe similarities between those studies and what conclusions can be derived from it.

The similarity and difference with previous studies (Putt et al., 2012) have been further detailed, and additional conclusions were included.

“While pure DEHS demonstrates significant absorbance under UVC irradiation, as shown in Figure 6a, dyes dissolved in DEHS undergo photodegradation upon UVC exposure, similar to that observed in water-dissolved dyes (Putt, et al. 2012). This behavior of dyes in DEHS under UVC irradiation might closely mimic the response of pathogens in saliva, where proteins also absorb UVC light.”

Figure 11: Are this data from sampled aerosol droplets or from the dye solution within a quartz cuvette exposed to UV-light. Please add this information in the figure description. When the Power is multiplied by the amount of residue time, then the unit would be without the unit s (seconds)

The data in this figure are from the dye solution within a quartz cuvette upon UV irradiation. We revised the corresponding section to avoid further confusion.

“We initially used UV LEDs to irradiate the dye solution and confirm whether the selected dyes exhibited degradation upon exposure. This information guided us in designing the aerosol UV irradiation chamber. Figure ~~11~~12 presents examples of the measured UV-Vis spectra of dye solutions before and after irradiation with various UV radiation doses, using experimental setup displayed in figure 2.”

Line 315; Please add information on the effective volume, or normalization on the aerosol.

We've updated the relevant section to offer more comprehensive details about the UV radiation chamber and the distribution of aerosol particles.

“Figure 10 displays the aerosol particle distribution produced using dye solution #4 (10 µg·mL^{-1})….Figure 3 illustrates both the experimental setup used and the calculated effective irradiated volume. The estimated UV radiation doses, detailed in Table 2.”

Line 318; How was the 10% Determined, please at Data, method and Figures for this procedure. Could this degradation be caused by dilution, shown in Figure 7? Please show a graph with DEHS without UV exposure and DEHS with UV; together with dye loaded DEHS with and without UV exposure, to show the degradation quantity.

We revised several sentences to explain how the dye degradation was calculated. The procedure details were also included. Moreover, we added the UV-Vis spectra of collected aerosol droplets of both pure DEHS and dye solution #4 (10 µg·mL-1) before and after UV exposure.

"Figure 13b shows *an example of* the UV-Vis spectra *for* the collected droplets, which we used to calculate the fraction of dye degradation. This calculation involved converting the maximum absorbance values at 497 nm (for dye #4) after UV irradiation (using apparatus in figure 3) into a percentage of the original absorbance value before irradiation….
In addition, Figure 13b presents the UV-Vis spectra of collected aerosol droplets of both pure DEHS and dye solution #4 (10 µg·mL^{-1}) before and after UV exposure. The degradation of dye #4 after UV exposure is evidenced by noticeable changes in the maximum peak absorbance (specially at 497 nm for dye #4). While pure DEHS exhibits some spectral changes under UV irradiation, these changes are predominantly below 300 nm wavelength, with insignificant absorption changes above 400 nm. Therefore, the error introduced by using DEHS as the spectral baseline for determining dye degradation is considered negligible."

Line 323; If Sedimentation was an issue, is there a chance to alter the experimental set-up? Longer tube, vertical stand?

If this is a problem, sedimentation could be reduced through the use of vertical UV irradiation chambers (with airflow flowing in from the bottom and out from the top). We thank for this suggestion and added one sentence in the revised manuscript.

"…limiting the ~~residence time~~ sedimentation loss of aerosol droplets by using a vertical UV chamber in further study."

Line 325; Please make it clear, in table 2, conclusion and experimental set-up description, what flow rates were actually used for later examination. State the sedimentation problem earlier in the text.

We amended these parts (Table 2 and the corresponding sections in Experimental details and Conclusions) for the sake of better understanding and clarity. As the exact consequences of particle sedimentation have not been examined in detail in the current work. We would like to point out this issue for the study in future.

Figure 12; Please change "survival fraction" to dye integrity or similar. Make a statement about the reason of non-linear degradation

The survival fraction was changed to degradation fraction and a possible reason for non/linear degradation was added.

"Another possible cause for the observed nonlinear degradation might be the different degradation responses in droplets of varying diameters, resulting from the variable gas flow to the impactor, which in turn leads to different cutoff sizes for the collected droplets."

Line 327: Use citations. E.g. this Review paper: DOI: 10.1016/j.jhin.2021.05.005

This citation has been incorporated.

Line 342; please add the size of the tube and flow rate as well.

The information requested has been added for clarity.

"…through a UV irradiation chamber (with an effective UVC irradiated volume of 777 cm^3)….

…a non-linear correlation between the ~~survival~~ degradation rate of UV-sensitive dyes and the increase in UV radiation doses. Specifically, a UVC dose of 245.1 $mW \cdot s \cdot cm^{-2}$ (with an aerosol flow of 0.78 $L \cdot min^{-1}$) at 270 nm degraded approximately 10% of the dyes in DEHS aerosols, while a lower dose of 21.6 $mW \cdot s \cdot cm^{-2}$ (with an aerosol flow of 8.86 $L \cdot min^{-1}$)"

Line 344; "feasibility of quantitively" change to "approach for qualitatively determining"

This suggestion has been incorporated.

Minor Remarks

Line 41; the radiation is also capable of breaking chemical bonds. Resulting in the destruction of the genome.

This suggestion has been incorporated with: "The UVC radiation is capable of breaking chemical bonds ~~absorption results in the formation of dimeric lesions~~ in the genome of pathogenic microorganisms, …"

Line 55; Use of non-breaking space for ²

Already amended

Line 189; delete "overall"

Already amended

Line 190; Briefly

Already amended

Line 259; "2.0 bar", Can you add information for the respective slm (liter per minute) information?

We have added the corresponding aerosol flow in slm.

Figure 8: Number concentrations at the Y-Axis please in log scale.

Statistical analysis of aerosol size distributions commonly employs lognormal distributions, as also suggested by TSI and Dekati. In this approach, the Y-Axis represents the particle number concentration on a linear scale, while the X-Axis indicates particle size on a logarithmic scale. Therefore, we would not change the Y-Axis in log scale.

Figure 9; Number concentrations at the Y-Axis please in log scale

Same as above

Line 276; Please refer the "lab build impactor" to a figure or "as described in …"; Please refer to the Cut-Off size of the Impactor

This suggestion has been incorporated.

Line 279; Dye solutions #4 and #7 with….

Already amended

Line 313; Please refer to table 2

Already amended

Line 320; please change survival to degradation loss or similar.

Already amended

Line 341; please change survival rate to dye degradation or similar.

Already amended

Abstract

"UV light-based air disinfection methods"

Accordingly, we modified this sentence in abstract.

References

All the following suggested citations have been incorporated in the revised manuscript (See References in revised manuscript).

Chiappa F, Frascella B, Vigezzi GP, Moro M, Diamanti L, Gentile L, Lago P, Clementi N, Signorelli C, Mancini N, Odone A. The efficacy of ultraviolet light-emitting technology against coronaviruses: a systematic review. J Hosp Infect. 2021 Aug;114:63-78. doi: 10.1016/j.jhin.2021.05.005.

Chen B, Jia P, Han J. Role of indoor aerosols for COVID-19 viral transmission: a review. Environ Chem Lett. 2021;19(3):1953-1970. doi: 10.1007/s10311-020-01174-8 . Epub 2021 Jan 13. PMID: 33462543 ; PMCID: PMC7805572.

Liqiao Li, Eon S. Lee, Charlene Nguyen & Yifang Zhu (2020): Effects of propylene glycol, vegetable glycerin, and nicotine on emissions and dynamics of electronic cigarette aerosols, Aerosol Science and Technology, DOI: 10.1080/02786826.2020.1771270

Jordan Peccia, Holly M. Werth, Shelly Miller & Mark Hernandez (2001) Effects of Relative Humidity on the Ultraviolet Induced Inactivation of Airborne Bacteria, Aerosol Science & Technology, 35:3, 728-740, DOI: 10.1080/02786820152546770

Putt, K. S., Kernick, E. R., Lohse, B. K., Lomboy, J., O'Brien, T., and Pugh, R. B.: The use of chromophore and fluorophore degradation to quantitate UV dose: FD&C dyes as chemical identicators for UV sterilization, Journal of Microbiological Methods, 91, 215–221, https://doi.org/10.1016/j.mimet.2012.08.015, 2012.

Talaat, K., Xi, J., Baldez, P. et al. Radiation Dosimetry of Inhaled Radioactive Aerosols: CFPD and MCNP Transport Simulations of Radionuclides in the Lung. Sci Rep 9, 17450 (2019). https://doi.org/10.1038/s41598-019-54040-1

Citation: https://doi.org/10.5194/ar-2023-9-RC1

Comments from Review 2 (RC2)

The responses below address all the points raised by the reviewer. Our responses are shown in blue, deleted text in red, and added text in gold.

General comments

In this manuscript, a method using UV-sensitive dye for UV dose determination is developed. Although the amount of work is much appreciated, there is very limited data analysis and discussion. The experimental design is questionable and many methods used are not well described and need clarification.

The major concern I have is the applicability and feasibility of using the UV-sensitive dye for UV dose measurement as described in the manuscript. The main challenge for measuring the UV dose exposed to pathogens in aerosols in air disinfection studies is that it is very difficult to account for physical shielding provided by the aerosol chemical constituents, which normally from saliva or nasal fluids that contains proteins and absorb UV. The study proposed using DEHS as the dye carrier but it is unclear whether DEHS can simulate the physical shielding and other potential interactions in real pathogen containing aerosols. In addition, the authors failed to demonstrate an actionable procedure using the proposed method for UV dose determination in other experimental settings.

We are truly grateful that you have dedicated the time and effort to providing thoughtful suggestions and critical comments. We have thoroughly considered the reviewer's comments and implemented significant enhancements to this manuscript. These improvements encompass the inclusion of additional data, expanded discussions, and the provision of more detailed experimental information to enhance overall understanding and clarity.

It's worth noting that DEHS, being an oily organic substance, exhibits significant absorbance under UVC (200-300 nm) irradiation. This characteristic is clearly demonstrated in the newly added Figure 6a, which illustrates the UV-Vis spectrum of pure DEHS. The degradation of dye within DEHS droplets when exposed to UVC light may closely resemble the behavior of pathogens in saliva, as proteins in saliva also exhibit UVC light absorption.

Given that the primary goal of this research is to showcase the feasibility of estimating UV irradiation doses delivered to aerosols using dye-laden droplets, we intend to further explore various experimental parameters, including relative humidity and temperature. Additionally, we plan to develop a mathematical model to better understand dye decay upon UV radiation. These aspects will be the focal points of our future research.

Specific comments:

Line 42: Please consider using other references here. UV inactivation mechanisms were not investigated in these two studies.

We substituted the original citations with alternative references related to research on UV inactivation mechanisms.

"(Budowsky et al., 1981; Kowalski, 2009; Beck et al., 2016)"

Line 93: You mean "no observable sedimentation"?

Thank you for highlighting this. Indeed, we used "no" observable sedimentation as a criterion to maintain a stable solution with a consistent dye concentration. We have clarified this sentence in the revision.

Line 105: Need more background on DEHS and why it has been selected for droplet generation. Any previous studies? Are there any other factors that should be considered for the carrier liquid selection, such as density, viscosity, absorbance, and potential chemical reaction with the dye?

We added several sentences to provide more background about the wide application of DEHS in aerosol science.

"In aerosol science, DEHS is widely used to generate liquid particles with extended lifetimes due to its extremely low saturation vapor pressure. This quality is crucial for studying particle size effects (Ren et al., 2021; Li et al.,

2020). Besides, DEHS aerosols, recommended for aerosol filtration testing, allow for more accurate particle size measurement of spherical liquid particles compared to non-spherical solid particles like salt and test dusts, which tend to agglomerate (Gustavsson, 2003)."

Additionally, since DEHS is an oily, organic substance, it exhibits notable absorbance under UVC irradiation, a characteristic evident in Figure 6a (Section 3.1.1), which displays the UV-Vis spectrum of pure DEHS. The degradation of dye in DEHS droplets when exposed to UVC light might closely resemble the behavior of pathogens in saliva, as proteins in saliva similarly absorb UVC light.

We added following sentences in Section 3.3."While pure DEHS demonstrates significant absorbance under UVC irradiation, as shown in Figure 6a, dyes dissolved in DEHS undergo photodegradation upon UVC exposure, similar to that observed in water-dissolved dyes (Putt, et al. 2012). This behavior of dyes in DEHS under UVC irradiation might closely mimic the response of pathogens in saliva, where proteins also absorb UVC light."

Line 112: "solutions".

This word has been corrected in the revised manuscript.

Line 117: I don't understand why having a similar "UV susceptibility" to nucleic acids is necessary here. You may have different relative sensitivity to DNA/RNA across wavelengths and simply having a sensitivity spectrum documented. Also, how did you determine the "UV susceptibility"? Both absorbance the quantum yield are needed to estimate the susceptibility but it seems you only have the absorbance documented.

We apologize for any confusion caused and have revised the relevant section for improved clarity.

"Given the varying susceptibility of different UV-sensitive dyes to UVC exposure, dyes with an absorption peak near 260 nm (below 300 nm in this study) were selected for further study. This is because the chemical structure, crucial for the replication of deoxyribonucleic acid (DNA) and ribonucleic acid (RNA), is more sensitive to UV irradiation near 260 nm, where it also exhibits an absorption peak (Abkar et al., 2022). ~~those with UVC susceptibility similar to deoxyribonucleic acid (DNA) or ribonucleic acid (RNA), were identified for further investigation.~~"

In this study, we focused only on measuring the UV-visible spectra of various dyes to determine their suitability. We specifically selected dyes that show significant absorbance below 300 nm, which we assume to be near the 260 nm peak absorbance of DNA/RNA, for further investigation

Line 119: I don't understand how the solubility is calculated here. From absorbance and concentration, you may calculate the molar absorptivity according to Beer's law, no sure they are related to the solubility. Also, it is not clear why solubility should be considered for the dye selection here.

Substances can be classified as "insoluble" if their solubility falls below 100 µg/mL in a given solvent Stegemann et al. 2007). In our study, dye solutions prepared at this concentration (100 µg/mL) level showed varying degrees of sedimentation after 48 hours of observation. Therefore, we used UV-Vis spectroscopy to further estimate the solubility of "insoluble" substances. We have amended the relative section for better clarity and understanding.

"As noted by Stegemann et al. (Stegemann et al., 2007), substances are typically classified as 'insoluble' when their solubility is below 100 µg/mL in a solvent. Technique like UV-Vis spectroscopy can be used to estimate the solubility of insoluble substances, particularly with those with very low solubility (such as below 100 µg/mL).… According to Beer's law, we expect that the maximal absorbance obtained correlates with the solute concentration, ideally forming a linear standard curve. If the measured data do not exhibit a linear regression, the corresponding substance is excluded."

Line 141: Droplet evaporation likely occurs in the real-world. Does it affect the accuracy of UV fluence measurements using the dye?

In practical scenarios, when a droplet (such as water-based droplet) undergoes evaporation, it leads to a reduction in its volume, yet the quantity of dye remains unchanged. This process causes the concentration within each droplet to increase. Consequently. the dye concentration measured with the collected liquid using an impactor might be biased towards the concentration within individual droplet particles in the real. However, in our study, we utilized DEHS droplets, known for their stability in maintaining volume over several hours, even at nanoscale dimensions. Therefore, we hypothesize that the concentration we measured after droplets collection can represent the true concentration within the individual droplet particles.

Line 160: Is 275 nm the peak emission wavelength? Need to show the UV LED emission spectrum, especially considering LED tends to have a wide emission peak, covering up to 20 nm.

Yes, 275 nm is peak and dominant wavelength, as specified by the manufacturer. This information has been updated in the revised manuscript. We acknowledge that the peak emission of the LED, typically around $\lambda_{max}\pm10$ nm, is broader than that of a low-pressure mercury lamp. In this study, we did not independently measure the emission spectrum of the LEDs to verify their spectrum width. However, this question also provides an interesting aspect to consider for future research.

Line 164: What is the dimension of the cuvette. I'm assuming there is no mixing for the dye solution in the cuvette so the UV irradiance decreases across the solution depth, which could be problematic.

We have included the dimensions of the cuvette in our revised manuscript.

"The collected liquids were transferred to a quartz submicron cuvette (Part number: 6610024100, Agilent Technologies, Santa Clara, USA), having a minimum capacity of 80 µL (external dimensions: 45×12.5×12.5 mm, opening aperture: 10 mm, path length: 10 mm)."

During UV irradiation, we refrained from using any mixing techniques, like a stirrer magnet, as they might affect the irradiation process. However, the dye solution was thoroughly mixed prior to the UV-Vis measurement. Our observations indicate that the volume and depth of the liquid lead to uneven UV irradiation, resulting in different levels of dye degradation across various parts of the liquid. Therefore, the UV light dosage determined in this study is specifically used for quantitative analysis, aimed at assessing the degradation of the selected dyes to LED UV light.

Line 165: Need more details on the UV light meter. Is it calibrated specifically for this UV LED? UV light meter normally has different sensitivity across wavelengths and often needs to be calibrated for specific UV device, especially for UV LED considering the wide emission peak. Also, what is the size the meter sensor aperture and is this the same as the size of the cuvette?

The UV light meter has a spectral measurement range of 250-410 nm. We do not have additional calibration for this UV LED (with a peak wavelength of 275 nm). The detector of this UV light meter has a circular area with a 10 mm diameter, matching the opening aperture of the cuvette used in this work. The required details on the UV light meter have been added.

"The detector of this UV light meter has a circular area with a 10 mm diameter, matching the opening aperture of the cuvette used in this work."

Line 181: The measurement taken here is likely not the average irradiance across the tube. Light irradiance is proportional to the inverse square of the traveling distance (inverse square law), not a linear decrease.

We thank the reviewer for highlighting this issue. The sentence has been revised to underscore that the intensity measured along the center line might not accurately represent the intensity experienced by all particles.

"It should be noted that the intensity measured along the center line might not accurately represent the intensity experienced by all particles. Predicting the equivalent radiation intensity for aerosol droplets in the UV chamber requires developing simulation models, which was not addressed in this study. These mathematical models would need to consider numerous factors, including the optical field of UV irradiation (accounting for reflections and refractions at interfaces between air, quartz, and nitrogen) and the flow field, which affects UV scattering and shadow effects related to particle sizes and trajectories (Kowalski, 2009). Therefore, in our research, we utilized the irradiation intensity measured at the chamber's largest cross-section to inform feedback for our reactor design."

Line 210: Please explain why 100 ug/mL is tested here. A dye could still be considered as a good selection as long as it can provide sensitivity and measurable responses to UV irradiation at a lower concentration, even the solubility less than 100 ug/mL.

We selected dye concentration of 100 ug/mL to align with studies by Putt, who investigated the degradation of chromophores in water solutions for determining UV light radiation doses.

We revised the following sentence:“ To rapidly screen UV-sensitive dyes soluble in DEHS, solutions were prepared at a dye concentration of 100 μg·mL-1, aligning with the concentration used by Putt et al. (Putt et al., 2012), who studied chromophore degradation in water solutions for evaluating UV radiation doses.”

Line 218: It is not true. The 260 nm peak absorbance only applies to > 240 nm. Higher absorbance can be observed in the far UVC range (<230 nm).

Thanks for pointing this out. We have revised this sentence to enhance its clarity and comprehension.

Line 224: Please explain why board peaks indicate chemical reactions or polarization?

We revised this sentence for better clarity and added citations to support the explanation that potential alterations in the chromophore structure might occur.

“…, as indicated by ~~broad absorption peaks~~ their UV-Vis spectra. These spectra show absorption across the whole visible wavelength spectrum and the absorption peaks appear broadened and obscured. For a given organic chromophore, the absorption peaks or shoulders are common for the presence of the conjugated systems and might become broad when extensive conjugation occur with the solvent dependent (Hemdan, 2023; Zheng et al., 2018).”

Figure 6: Recommend just showing the germicidal UV range (200 – 320 nm). The data in the visible range is not useful in this study.

As previously clarified, we utilized DEHS's UV-Vis spectra from 270 to 800 nm as the baseline for measuring the spectra of pure dyes, since its absolute absorbance exceeds 1 at wavelength below 270 nm (see figure 6 in revised manuscript). Due to significant baseline fluctuations from 200-270nm, the UV-Vis spectra below 270 nm was excluded. We have included a statement and discussion about this baseline. Furthermore, the data in the visible range exhibits a distinctive peak that represents the specific fine structure of a particular dye and exclude the error caused by the DEHS baseline. We utilize the variations in the absorbance peak to calculate the changes in dye concentration caused by degradation after UV exposure.

Figure 7. I don’t understand why the relationship between concentration and absorbance is needed here. It is well known that the absorbance is proportional to concentration (Beer’s law).

Beer Lambert law is valid only when the sample being analyzed is a homogeneous solution, which we cannot know by sedimentation observation. We used UV-Vis spectroscopy to further quantitively estimate the solubility of the selected dyes. According to Beer’s law, we expect that the measured maximal absorbance correlates with the solute concentration, ideally forming a linear standard curve. If the measured data do not exhibit a linear regression, the corresponding substance exhibits inhomogeneous distribution in DEHS liquid and is excluded for further study.

Line 245: Please include references.

Accordingly, the corresponding references have been included in the revised manuscript.

Line 295: Move to introduction.

After careful evaluation, we concluded that this section is not well-suited for the introduction. However, we have revised this section for improved clarity and to prevent any potential confusion.

“~~Currently, most UV disinfection systems for water, air, and surfaces continue to utilize conventional low- or medium-pressure mercury lamps. The primary concern with these lamps is their fragility and their containment of toxic mercury, which poses environmental hazards and requires proper disposal. UV LEDs, on the other hand, are emerging as a popular, environmentally friendly alternative. Their compact size simplifies their integration into sterilization systems, and they offer a diverse range of wavelengths (Kim and Kang, 2018; Song et al., 2016). Considering the above reasons, this study employed UVC LEDs to investigate dye degradation upon UV radiation.~~ In this study, UVC LEDs were utilized to explore the degradation of dyes under UV radiation. Traditional low- or medium-pressure mercury lamps, although effective, raise concerns due to their fragility and the presence of toxic mercury, posing environmental hazards and necessitating proper disposal. Conversely, UV LEDs are emerging as

a favored and eco-friendly alternative (Chiappa et al., 2021). Their compact size facilitates easy integration into sterilization systems, and they provide a wide range of wavelengths (Kim and Kang, 2018; Song et al., 2016)."

Figure 11. Why is 2000 to 6000 mJ/cm2 selected here? This range is way too high for disinfection application, and the sensitivity at reasonable UV fluence range is unclear from this study. Also, a mathematical relationship between the UV fluence and absorbance needs to be developed here.

In this study, we irradiated the dye solution with doses ranging from 0 to 8000 mJ/cm^2. Only a few examples are presented in the illustration, as the spectral changes between 2000 and 6000 mJ/cm^2 can be visually identified and compared. The higher irradiation dose required could potentially depend on the species and is also influenced by factors such as the irradiated sample volume and UV absorption of the carrier liquid DEHS. The sensitivity comparison is evident through the peak changes at the same UV dose.

We did not develop a mathematical model for the relationship between UV irradiation dose and dye solution degradation in this study. This decision is because the UV irradiation dose depends on various factors, such as the experimental equipment, solution volume, and the medium used. Initially, we used UV LEDs to irradiate the solution to verify if the selected dyes exhibited degradation upon irradiation. This step provided valuable feedback for designing the aerosol UV irradiation chamber. The related section has been revised as follows.

"We initially used UV LEDs to irradiate the dye solution and confirm whether the selected dyes exhibited degradation upon exposure. This information guided us in designing the aerosol UV irradiation chamber. Figure ~~11~~12 presents examples of the measured UV-Vis spectra of dye solutions before and after irradiation with various UV radiation doses, using experimental setup displayed in figure 2. The higher irradiation dose required could potentially depend on the species and is also influenced by factors such as the irradiated sample volume and UV absorbance of the carrier liquid DEHS. As the UV radiation dose increased, the maximal absorbance values in the visible region decreased noticeably. Moreover, despite a similar degradation trend observed for the two tested dye solutions, at the same UV irradiation dose of 6000 mW·s·cm^{-2}, the degradation fraction for dye 4 was 90% while dye 7 was 50%. It can be concluded that dye solution #4 demonstrated a higher sensitivity to UVC irradiation at 275 nm."

Figure 12: How was the "dye survival" determined, absorbance? Again, a mathematical relationship between the UV fluence and "dye survival" needs to be developed here. This relationship needs to be compared with the results in Figure 11.

We have revised several sentences to explain how the calculation of dye degradation was performed, including the procedure details.

"Figure 13b shows an example of the UV-Vis spectra for the collected droplets, which we used to calculate the fraction of dye degradation. This calculation involved converting the maximum absorbance values at 497 nm (for dye #4) after UV irradiation (using apparatus in figure 3) into a percentage of the original absorbance value before irradiation…."

Although our ability to develop a mathematical model or relationship between dye degradation under UVC irradiation was constrained by limited experimental data, we have made enhancements to Section 3.3 to provide a more thorough explanation. The development of a mathematical relationship between UV radiation dose and dye degradation will be a focus of our future research.

"Two potential mathematical models that might offer insights into dye degradation: the classical exponential decay model (log-linear decay) and the shoulder model, which begins with a horizontal slope before transitioning into full exponential decay (Kowalski, 2009). However, the determination of which model is better suited for the selected dye solution remains inconclusive due to limited experimental data. A dataset that includes 90% dye degradation induced by UV radiation is, at a minimum, required."