

The responses below address all the points raised by the topical editor and the reviewers. Our responses are shown in blue, deleted text in red, and added text in green.

Topical Editor

The reviewer's comments have been addressed and the article can now be published subject to some minor revisions. Together with the final few comments provided by the two reviewers, please consider the following minor corrections needed:

We thank all the reviewers and editors for reviewing this manuscript and allowing us to revise it again. Our point-by-point responses can be found in the text below.

Line 71; Comment by reviewer 1: Citation needed for the statement: "...has been used to measure the radiation doses of UV light with a wavelength of 254 nm, serving as chemical indicators for UV sterilization processes": Please add a relevant citation.

In the revised manuscript, we have added a relevant citation.

Changed to "...the degradation of chromophores and fluorophores has been used to measure the radiation doses of UV light with a wavelength of 254 nm, serving as chemical indicators for UV sterilization processes (Putt et al., 2012)."

Line 334: Please correct to "ELPI measurements show CMD values ranging from...".

Thank you for pointing this out, we have revised this sentence.

Changed to "~~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."

Figure 6: UV-Vis Spectra of dye solutions, where only group C is suitable due to their absorption characteristic in the vicinity of 260 nm and in the visible region.: Please correct Figure 6 to Figure 7.

Accordingly, the figure number in the revised version was corrected.

Figure 7: Effect of LPI collection on the dye concentrations by using various concentrations of dye solutions. Please correct Figure 7 to Figure 11.

Accordingly, the figure number in the revised version was corrected.

Figure 8: Dye solution degradation upon UVC irradiation using a UVC LED (275 nm) with various UV radiation doses ($\text{mW}\cdot\text{s}\cdot\text{cm}^{-2}$), using experimental setup displayed in figure 2. Please correct Figure 8 to Figure 12.

Accordingly, the figure number in the revised version was corrected.

Figure 9: (a) Effect of UVC radiation dose on the dye survival degradation within aerosol droplets and (b) UV-Vis spectra of collected aerosol droplets of both pure DEHS and dye solution #4 ($10\ \mu\text{g}\cdot\text{mL}^{-1}$) before and after UV exposure. Please correct Figure 9 to Figure 13.

Accordingly, the figure number in the revised version was corrected.

Lines 432-435 "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.": I think this part should be moved/merged with the part in Lines 450-455, where Figure 13b is discussed again.

Thank you for bringing this to our attention. We modified the first part to avoid repetitive discussions about Figure 13b.

Changed to “The influence of UV dose on the degradation of dye-laden aerosols can be demonstrated in Fig. 13. ~~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~~ The calculation of the fraction of dye degradation 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.”

Reviewer Report #1

General

The paper presents initial results from laboratory measurements of a design to measure the effect of ultraviolet light on UV-sensitive on UV-sensitive dye dissolved in DEHS aerosols. Using a custom-built impactor and irradiation chamber, dye-loaded aerosols are exposed to UV light. A spectrophotometer was used to determine dye degradation. The quality of the presentation of the results has improved dramatically. However, there are few small things to consider.

Thank you for your time and effort in reviewing this revised manuscript. We have considered all your comments, and the detailed responses can be found in the text below.

Specific Comments

Line 405- Please add a statement about the shielding issue in the conclusions as well.

Accordingly, we have rewritten this sentence to include a statement about the shielding effect in fluids containing pathogens.

Changed to “~~This behavior of dyes in DEHS under UVC irradiation might closely mimic the response of pathogens in saliva, where proteins also absorb UVC light.~~ The behavior of dyes in DEHS under UVC irradiation may closely mimic the response of pathogens in saliva or nasal fluids, which contain proteins and also absorb UVC light. Using DEHS as a dye carrier could simulate the physical shielding and other potential interactions found in fluids containing pathogens.”

Figure 12 - Don't the high radiation doses with the same minimal decrease in peak absorption height show that the dyes are UV sensitive but still too stable to be used as a suitable model? Suitable enough for initial tests, but better dyes need to be found.

Yes, this work presents our initial tests designed to demonstrate the feasibility of estimating UV irradiation doses delivered to aerosols using dye-laden droplets. The high radiation doses required for dye degradation could be attributed to the shielding effects of the DEHS carrier liquid or the UV sensitivity of the selected dyes. As clarified in our manuscript (Lines 428-436 on page 16), further research will be conducted to explore the potential effects of carrier liquid and droplet sizes. We will also seek to find dyes with higher UV sensitivity.

Major Comments

Line 45 - Is it environmentally friendly to run ventilation and uv lamp 24/7?- The Reference to Heßling et al. 2020 claims just to be more energy-efficient.

We apologize for any confusion caused and have modified this sentence for improved clarity.

Changed to “UV radiation presents a more environmentally friendly and more energy-efficient alternative to liquid disinfectants and heat disinfection for sterilizing liquids, air, and surfaces (Heßling et al., 2020).”

Figure 7- It is difficult to understand the reasoning that substances absorbing below 300 nm wavelength are interesting when the DEHS already outperforms/overshadows everything there. Data would be clearer without DEHS to show, that :(Line 297) “dyes with obvious absorbance below 300 nm, which we assume to be around 260 nm, are desired”

We agree that detailing absorption characteristics below 300 nm is essential to clarify the photostability and degradation of the tested dyes. Since these dyes are insoluble in pure water (exhibiting no background effects below 300 nm), we will consider measuring the absorption spectra of pure dye powders in future research. This approach will help eliminate interference from DEHS liquid.

Minor Comments

Line 31 – personal protective equipment (PPE) - This abbreviation is used only once; swap order or omit abbreviation

The abbreviation 'PPE' has been deleted from the revised manuscript.

Figure 2 – a higher resolution is recommended

Figure 2 has been revised with higher resolution.

Line 278- “no absorption upon UV-VIS-light”- change “to almost non-existent...”

The suggestion was considered, and the corresponding modification has been made.

Changed to “As depicted in figure 6a, while pure water has ~~no~~ almost non-existent absorption upon UV-Vis light, the chosen carrier liquid, ...”

Line 315 – Droplets exhaled, experiences fast evaporation and shrink in size

The suggestion was considered, and the corresponding modification has been made.

Changed to “Previous research has suggested that exhaled droplets, including aerosols produced by breathing, speaking, and coughing, which experience fast evaporation and shrink in size, are smaller than 5 μm, including aerosols produced by breathing, speaking, and coughing ...”

Reviewer Report #2

This work shows an option to determine the UV dose by means of aerosols and UV-photosensitive dyes. The method is suitable for aerosol and aqueous solutions. The photochemical reactions followed by HPLC could be an interesting contribution, although not mandatory. Please see the analysis for calculating the absorbed irradiance (absorbed photon flux density) and recalculating it: A. Ipiña et al. 2014, Solar Energy 109 (2014) 45–53, and Braslavsky, S.E., 2007. Glossary of terms used in photochemistry, Pure Appl. Chem. 79, 293–465.

Thank you very much for taking the time and effort to review our manuscript. We are also grateful for the literature you provided on calculating the absorbed irradiation. We will make a detailed estimate of the irradiation intensity after optimizing the design of the irradiation chamber.

1. Line 40 Definition of UV ranges according to CIE Position Statement on Ultraviolet (UV) Radiation to Manage the Risk of COVID-19 Transmission:

Based on the biological impact of ultraviolet radiation on biological materials, the ultraviolet spectrum is divided into regions: UV-A is defined by CIE as radiation in the wavelength range between 315 nm and 400 nm; UV-B 280 nm to 315 nm; and the UV-C between 100 nm and 280 nm.

Thank you for this clarification. Radiation with wavelengths between 100 and 200 nm, also called vacuum ultraviolet radiation (VUV), is usually not used for air disinfection purposes because it is strongly absorbed by air

(Heßling et al., 2020). The most studied UVC spectral ranges for air disinfection are 200–280 nm. We have modified this sentence for better clarity.

Changed to “Ultraviolet (UV) germicidal irradiation, specifically UVC irradiation within the wavelength range of 2100-280 nm (UVC), is known as an effective method for inactivating all known microorganisms and viruses (Abkar et al., 2022; Inagaki et al., 2020; Reed, 2010; Biasin et al., 2021). UVC radiation in the range of 200-280 nm is widely used in air sterilization research because radiation at wavelengths below 200 nm is absorbed by the air (Heßling et al., 2020).”

2. Line 109, a period is absent.

Accordingly, we added a period.

3. Line 189-190 This is only the source's emitted UV radiation dose at 10 mm. The absorbed irradiance should consider the path length through quartz cells.

The primary objective of this study was to determine the UV radiation dose to aerosols, rather than liquids, in a quartz cell. Here, the estimated UV radiation dose was utilized to investigate whether degradation of selected dyes occurs after exposure. Since our dye solution in the quartz cells was dosed at 1 ml, overlooking the radiation length did not affect our experimental results.

4. Please, for calculating the absorbed photon flux density, see: Braslavsky, S.E., 2007. Glossary of terms used in photochemistry, 3rd edition. (IUPAC Recommendations 2006). Pure Appl. Chem. 79, 293–465.

Thank you very much for providing this document. As clarified in our manuscript (lines 210-217 on page 7), the intensity measured along the centerline might not accurately represent the intensity experienced by aerosol particles. We will make a detailed calculation in further research after optimizing the design of the irradiation chamber.

5. Figure 8 Why did you calculate absorbance at 542nm? The maximum values are between 420nm and 510nm.

Thank you for pointing this out. The absorption peaks of dyes #3 and #6 were confounded. We have made corrections where relevant (Table 1, Table 3, Figure 7, and Figure 8).