Dear Reviewer 2,

We addressed your comments and suggestions point by point as follows:

Comment / Suggestion:

It is not clear how this number: “150” comes out and how much is the associated error. The comparison in fig.4 is made with different sources 241Am and 239Pu, by using very different geometry, moreover there are no numbers/scale in fig4.

Reply:

We clarified the way we calculated the number by changing the relevant part of the discussion to

> We estimated the light yield per alpha particle in the liquid by integrating the pixel intensities of the background-corrected planchet image and dividing with activity and exposure time. A similar procedure has been used for to the central parts of liquid radioluminescence image, and the resulting light yield was further scaled to take into account the total liquid area in the cuvette. The energies of alpha particles emitted by Pu-239 are 5.157 MeV and by Am-241 are 5.486 MeV, respectively[27], thus the two samples are quite similar from an alpha radiation point of view. From this analysis, we find that the light yield of an alpha particle in the liquid is roughly 150 times lower than in air.

The caption of Fig. 4 already says ”The z-scale used in the images is the same as the one used in Fig. 2”.

Comment / Suggestion:

There is no comparison with a luminescent non-radioactive liquid to exclude an optical effect of the liquid/air interface. My hypothesis is that light crossing the liquid/air interface, is crossing from high density medium to air thus there is large refraction and this can explain the observed light maximum. This is supported by dotted lines in fig 4 and fig. 5b where it is shown that maximum occurs below the interface and not above (in the air) where one should expect the air luminescence. As an example, it is possible to see this optical effect in fig.4 of the paper DOI: 10.1103/PhysRevX.9.011024

If authors want to mention the air luminescence hypothesis have to support this with a picture of non-radioactive but luminescent liquid excluding the presence of a maximum at the boundary due to the simple refraction effects.

Reply:

Unfortunately we do not have images of the proposed non-radioactive luminescent liquid. However, we do know that alpha particles are emitted
out from the liquid to air \([\text{https://doi.org/10.1016/j.nima.2004.07.290}]\), and that they produce radioluminescence in air \([\text{https://doi.org/10.1088/1367-2630/16/5/053022}]\). Thus our speculation with radioluminescence of air at the surface is well justified, but we cannot rule out the optical effect in the surface either. We decided to modify the manuscript (see below) to present the two possible explanations for the enhanced intensity at the surface, as we cannot prove the origin of it. The proposed hypotheses are the radioluminescence of air and optical reflection/refraction from the surface.

> The pixel intensities at the liquid-air boundary in Fig. 4 (d) are higher compared to the pixel intensities further down the water column. The shape of this area is coincidental with the shape of the liquid-air boundary in Fig. 4 (c), which can be even better seen in the overlay image in Fig. 4 (e). The camera is slightly tilted towards the cuvette, which makes it possible to look on top of the interface of the liquid-air boundary. The increase in the intensity might be due to an optical effect or an increased light production at the surface. The optical refraction at the liquid-air boundary can redirect more radioluminescent light towards the camera. Also, it is known that alpha particles are emitted out from the liquid[28], and thus can create radioluminescence in air [5]. The radioluminescence yield in air was previously shown to be approximately 150 times more efficient, which can result in the increased intensity at the surface in Fig. 4 (d).

Comment / Suggestion:

I cannot see the blue shaded area in the figure.

Reply:

A typographical error. We corrected it to read:

> Dotted lines indicate the position where the interface of the liquid-air boundary faces the camera.

Comment / Suggestion:

The referee appreciate the inclusion of fig. 7.a in the draft, giving a reference for Air spectra. However to support conclusions on fig. 7.b., not only the position of N2 lines have to be explained but also relative amplitudes. I suggest to superimpose to fig. 7.a. a plot with the expected. Air luminescence spectra and to evaluate the system transmission function.

Reply:

We superimposed the radioluminescence spectrum of air on Fig. 7(a). To easier comprehend the superimposed spectra we normalized them to be 1 at the peak around 337 nm. We also updated the figure caption and the relevant parts of the discussion. The superimposed spectra make it easier to verify that the transmission function of our calibrated system is flat.

2
Comment / Suggestion:

To exclude these emission processes and support other processes the author should give some quantitative analysis. At least author should show superimposed to fig. 7.b) the expected Cherenkov and bremsstrahlung spectra considering the transmission/efficiency effects obtained with calibration figure 7.a). In particular the measured spectra shown in fig 7.b is qualitatively similar to the one expected by Cherenkov production in water, having a very similar behaviour and steep decrease at the short wavelength. On the other hand if the author hypothesis of a dominant effect of air luminescence on the boundary rface i, the measured spectrum is a mixture of liquid radioluminescence spectrum and air radioluminescence spectrum.

Therefore to exclude, as an example, Cherenkov emission process, a quantitative analysis is mandatory. As an example of a typical quantitative analysis it is possible to see fig.6 of:


Reply:

Cerenkov light shows a spectrum that is proportional to $\lambda^{-2}$. We also added this information to the discussion with the sentence

> Cerenkov light has a spectrum that is proportional to $\lambda^{-2}$, where $\lambda$ is the wavelength of the emitted light [30].

We further enhanced our discussion by adding

> Further, alpha particles are not capable of producing Cerenkov light directly or through secondary mechanisms [31] and therefore we conclude that the measured radioluminescence has to have an origin other than Cerenkov radiation.

Fig. 6 in [https://arxiv.org/pdf/1809.02048.pdf] shows the Cerenkov light spectrum that is modified by absorption of acrylic by their acrylic cube water tank. Acrylic is known for absorbing just below 400 nm. This link [https://www.plexiglas.com/en/acrylic-resins/optical-and-weathering-properties/] leads to a list of documents showing typical transmission curves of various types of acrylic.

Fig. 7 (b) of our manuscript is measured at the position of the bulk liquid. We added the sentence

> When the liquid sample was spectrally analyzed, the fibre was installed as close as possible to the sample holder to make sure that only radioluminescence from the bulk of the liquid reaches the detector.

3
Sincerely,

Thomas Kerst  
Corresponding author

Tampere University  
Physics Unit  
Photonics Laboratory  
P.O. Box 692  
FI-33101 Tampere, Finland  
+358 50 300 5968  
thomas.kerst@tuni.fi