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Visualization of low-level gamma radiation sources using a low-cost, high-sensitivity, omnidirectional Compton camera --Manuscript Draft--

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TITLE:

Visualization of Low-Level Gamma Radiation Sources Using a Low-Cost, High-Sensitivity, Omnidirectional Compton Camera

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KEYWORDS:

gamma-ray imaging, Compton camera, radioactive contamination, environmental monitoring, radiation protection, Fukushima Daiichi nuclear powerplant accident

SUMMARY:

We present experimental protocols for visualizing various low-level gamma radiation sources in the ambient environment using a low-cost, high-sensitivity, omnidirectional, gamma-ray imaging Compton camera.

ABSTRACT:

We present experimental protocols for visualizing various low-level gamma radiation sources in the ambient environment. Experiments were conducted by using a low-cost, high-sensitivity, omnidirectional, gamma-ray imaging Compton camera. In the laboratory, the position of a sub-MeV gamma radiation source such as ¹³⁷Cs can easily be monitored via omnidirectional gamma-ray imaging obtained by the Compton camera. In contrast, a stationary, wall-mounted dose rate monitor cannot always successfully monitor such a source. Furthermore, we successfully demonstrated the possibility of visualizing the radioactivity movement in the environment, for example, the movement of a patient injected with ¹⁸F-fluorodeoxyglucose (¹⁸F-FDG) in a nuclear medicine facility. In the Fukushima field, we easily obtained omnidirectional gamma-ray images concerned with the distribution on the ground of low-level radioactive contamination by radioactive cesium released by the Fukushima Daiichi nuclear power plant accident in 2011. We demonstrate clear advantages of using our procedure with this camera to visualize gamma-ray sources. Our protocols can further be used to discover low-level gamma radiation sources, in place of stationary dose rate monitors and/or portable survey meters used conventionally.

INTRODUCTION:

Medical facilities house various low-level gamma radiation sources with a surface and/or air dose

rate of just a few $\mu\text{Sv/h}$. Such sources are also present across broad areas of eastern Japan exhibiting low-level radioactive contamination by radioactive cesium from the Fukushima Daiichi nuclear power plant accident in 2011. These environments sometimes expose workers to the external irradiation exposure limit for the human body for the general population as advised by the International commission on radiological protection (ICRP): 1 mSv/year (e.g., 1 $\mu\text{Sv/h}$ for 4 h per day, 250 days per year)¹. If radiation sources are visualized from more than a few meters in advance on short timescales, the amount of radiation exposure can be reduced. One of the best solutions for visualizing these gamma radiation sources is to adopt a gamma-ray imaging Compton camera technique². In this technique, the energy and cone-direction of incident gamma-rays emitted from the radiation source are measured by the detector for each event, and then the gamma-ray source direction can be reconstructed by back-projection³. Previous studies have developed Compton camera systems aimed at the application of a new diagnostic device in nuclear medicine and/or a new gamma-ray telescope in astrophysics⁴⁻¹⁴, as well as image reconstruction techniques for Compton cone data by analytical^{15,16} and statistical¹⁷ approaches. More expensive, state-of-the-art devices with complicated electronics are often adopted to obtain high angular resolution within a standard deviation of a few degrees, but this precision makes it difficult to simultaneously achieve high detection efficiency.

Recently, we have proposed and developed a low-cost, high-sensitivity, omnidirectional gamma-ray imaging Compton camera¹⁸, based on a two-fold coincidence within a number of independent scintillators that act as either scatterers or absorbers¹⁹. The aim of this technique is to easily achieve high detection efficiency with an angular resolution σ of ~ 10 degrees or less, which is adequate for an environmental monitor. This is accomplished through the application of an image-sharpening technique^{18,20} based on the filtered back-projection algorithm, which applies a convolution filter used in image reconstruction for computed tomography to the Compton reconstruction. Furthermore, the detection efficiency, angular resolution and dynamic range of the detector can be easily optimized when the type, size and arrangement of scintillators are coordinated in accordance with a particular purpose, such as usage in environments emitting elevated radioactivity^{21,22}.

In this study, we present experimental protocols for various trials for visualizing low-level gamma-ray radiation sources using this omnidirectional Compton camera technique in a radioisotope (RI) facility, a positron emission tomography (PET) facility and the Fukushima field. We prepared and utilized the omnidirectional gamma-ray imaging Compton camera previously developed by ourselves¹⁸ but with some improvements, in order to achieve higher detection efficiency. **Figure 1** shows a schematic view of the arrangement of CsI(Tl) scintillators of eleven elements used in this study. The eleven counters consist of two layers; two counters at the center and nine counters in a half-circle, considering forward and backward scattering configurations. Each CsI(Tl) scintillator cube of 3.5 cm was read out with super-bialkali photo-multiplier tubes (PMT). The signals were fed into a flash ADC board with SiTCP technology²³ and the front end was connected to a PC via Ethernet. An online program created using Visual C++ with ROOT library²⁴ was operated on a Windows PC. A gamma-ray image was reconstructed and sharpened^{18,20} on a spherical surface with accumulating rings with a radius of θ that is a scattering angle calculated from Compton kinematics for each two-fold coincidence event. An omnidirectional gamma-ray

image can be displayed both online and offline by superimposition on the omnidirectional optical image previously taken by a digital camera. During the measurement, the trigger rate, total energy spectrum (the sum of the energy deposits for each two-fold coincidence event), and the reconstructed images of a preset gamma-ray energy can be displayed on the online PC screen. This information can be updated at a preset time interval (e.g., every 10 s). Here, we set the screen to display two types of reconstructed images: an image that is accumulated at the start of the measurement and an image reaccumulated at every preset time interval (e.g., every 1 min). Furthermore, because the raw data for each event obtained using the measurements are stored, it is possible to reanalyze the data after the measurements and then regenerate a reconstructed image for an arbitrary gamma-ray energy at an arbitrary time interval. **Table 1** shows the performance of the Compton camera system used in this study, by comparison with the previous six-counter system¹⁸. The comparison revealed that a sub-MeV gamma-ray source was successfully visualized with a detection efficiency twice that of the previous system, while maintaining the angular resolution σ of ~ 11 degrees. We also confirmed that the angular dependence of acceptance was kept to a minimum, showing differences of $\sigma \sim 4\%$. The details on the fundamental techniques of the system are described in Watanabe et al. (2018)¹⁸. Here we introduce three experimental protocols for visualizing various low-level gamma-ray radiation sources using the Compton camera described above.

PROTOCOL:

The protocol was conducted following the guidelines of the research ethics committee at the National Cancer Center Hospital East, Japan.

1. Monitoring of sealed radiation source in experiment room at RI facility

1.1. Set the Compton camera beside the dose rate monitor as shown in **Figure 2a**. Set the height of the detectors from the ground to 2.5 m. Build the dose rate monitor, which consists of a parallel plate ionization chamber, into the upper part of the entrance of the experiment room at the RI facility to monitor the air dose rate of the position at intervals of 1 min.

1.2. Turn on the power of the Compton camera and online computer.

1.3. Start the simultaneous measurement with Compton camera and the dose rate monitor.

1.4. Set a ¹³⁷Cs sealed source (3.85 MBq) at a position labelled 'A' in **Figure 2a** and leave it for 30 min. Set the distance between the detector and the sealed source to 3.6 m.

1.5. Move the sealed source to a position labelled 'B' and leave it during 30 min. Set the distance between the detector and the sealed source to 6.7 m.

1.6. Move the sealed source at a position labelled 'C' and leave it during 30 min. Set the distance between the detector and the sealed source to 6.7 m.

1.7. Move the sealed source at a position labelled 'D' and leave it during 30 min. Set the

distance between the detector and the sealed source to 1 m.

1.8. Move the sealed source outside the room. After 30 min, stop all measurement.

2. Environmental monitoring in PET facility

2.1. Set the Compton camera in front of reception desk in PET facility as shown in **Figure 2b**. Set the height of the detectors from the ground to 1 m.

2.2. Set the online computer in the staff room.

2.3. Turn on the power of the Compton camera and online computer.

2.4. Start Compton camera measurement early in the morning before patients arrive at facility.

2.5. After all patients leave for the day, stop all measurement.

3. Outdoor measurement in Kawamata-machi, Fukushima, Japan

3.1. Set the Compton camera near a private house as shown in **Figure 2c**, where the existence of some radiological Caesium hot spots with surface dose rates of 1 $\mu\text{Sv/h}$ or less are suspected. Set the height of the detectors from the ground to 1.5 m.

3.2. Turn on the power of the Compton camera and online computer.

3.3. Start Compton camera measurement.

3.4. After 30 min, stop all measurement.

REPRESENTATIVE RESULTS:

Monitoring of sealed radiation source in experiment room at RI facility

Figure 3a shows the time variation of trigger rate measured by the Compton camera (black solid line), after applying a time-lag selection of two-hit counters less than 1 μs . The trigger rate changed every 30 min depending on the position of the sealed source (i.e., distance from the position to the camera). This variation was confirmed from the data measured by the stationary dose rate monitor (blue dashed line); the behavior remained constant (i.e., background level) other than between 5750 s and 7800 s. Here, we tentatively set five periods labelled (i), (ii), (iii), (iv) and (v), representing the five positions of the sealed source (**Figure 3a**). **Figure 3b** shows the total energy spectra for each such period (30 min for each), the horizontal axis representing the sum of energy deposits for each two-fold coincidence event. We note 662 keV photo-absorption peaks originating from the ^{137}Cs sealed source for (i), (ii), (iii) and (iv), while (v) shows only background levels. Peak heights for (ii) and (iii) are the same, which we attribute to the same 6.7 m distance from the camera to the sealed source. By selecting the event within 662 ± 40 keV for 662 keV, we calculated the scattering angles and reconstructed the omnidirectional gamma-ray

image. The results are shown in **Figures 3c-f**, respectively, for periods (i), (ii), (iii) and (iv). Here gamma-ray images are indicated by the red region, which indicates gamma-ray intensities in the upper half of the observed range. We find that the position of the ^{137}Cs sealed source can be successfully identified from the gamma-ray images. **Figure 4** shows the changes in the image with integration time, where the red field instead corresponds to a narrower range (the upper 30 %) of the observed range. This narrower range was adopted in order to give priority to a peak intensity. In this case, ^{137}Cs source direction could be identified after 30 s.

Environmental monitoring in PET facility

Figure 5a shows the overall time variation of the trigger rate during the daytime (5.6 h) as measured by the Compton camera (black line) in front of a reception desk in a PET facility. We observe remarkable enhancement in the trigger rate with various patterns, which could be attributed to the movement of patients injected with ^{18}F -fluorodeoxyglucose (^{18}F -FDG) around the reception desk. As an example of such patterns, we focus on the period from 6200 s to 7000 s. According to the trigger rate in this period shown in **Figure 5b**, a series of enhancements are apparent, with two plateaus labelled (i) and (ii). **Figure 5c** shows the total energy spectra for **Figure 5b**'s periods (i), (ii) and (iii). We observe 511 keV photo-absorption peaks originating from the ^{18}F -FDG. **Figure 5d,e** show the 511 keV gamma-ray omnidirectional image in periods (i) and (ii), respectively, in which we selected events within 511 ± 40 keV for image reconstruction. The directions of gamma-ray peaks in both figures correspond respectively to the directions of the sofa and the restroom behind the wall. Considering the trigger rates of both (i) and (ii), we interpret the gamma-rays in (i) as leakage penetrating the shield of the wall from the restroom; we presume that a patient entered the restroom and spent two minutes, and after that sat on the sofa a few minutes before the PET scan.

Outdoor measurement in Kawamata-machi, Fukushima, Japan

Figure 6a shows the time variation of the trigger rate for 30 min of outdoor measurement. The stability of the trigger rate implies that our Compton camera system operates stably even for measurements conducted outdoors over a long period. To demonstrate how the extended gamma-ray source was reconstructed, we set four different integration periods labelled (i) (1 min), (ii) (10 min), (iii) (20 min) and (iv) (30 min), as shown in **Figure 6a**. **Figure 6b** shows the total energy spectra for each period, depicting the structures superimposed on the photo-absorption peaks of gamma-rays emitted from radioactive nuclides at 605 keV and 796 keV for ^{134}Cs and 662 keV for ^{137}Cs . To reconstruct the gamma-ray image, we selected events within 565-622 keV for 605 keV, 662 ± 40 keV for 662 keV and 796 ± 40 keV for 796 keV. The gamma-ray omnidirectional images for 605, 662 and 796 keV are shown in **Figures 6c-f** for integration periods (i), (ii), (iii) and (iv), respectively. In this case, we find that the reconstructed gamma-ray distribution is stable as long as the integration time exceeds 20 min. The slope of a hill in front and the lower part of rain gutter are clearly contaminated, while the area covered with uncontaminated soil in the right part of image is demonstrably not contaminated. The gamma-ray intensity is in good agreement with dose rate values measured by a scintillation-type survey meter, the values of which are shown in yellow in **Figure 6f**.

FIGURE AND TABLE LEGENDS:

Figure 1: Omnidirectional Gamma-ray imaging Compton camera system. (a) Geometrical arrangement of scintillators with eleven elements used in this study. Two scintillators were arranged at the center of a circle, with nine more arranged in a half-circle, staggered vertically. **(b)** Photograph of the detector without housing. The counters were fixed inside an expanded polystyrene.

Figure 2: Experimental setup. (a) Monitoring of a sealed radiation source in the experiment room at the RI facility, where a ^{137}Cs -sealed source was sequentially set at the positions labelled 'A', 'B', 'C' and 'D'. **(b)** Environmental monitoring in front of a reception desk in the PET facility. **(c)** Outdoor measurement in the Fukushima field, Japan. The Compton camera was fixed on a stepladder.

Figure 3: Representative results of the monitoring of a ^{137}Cs -sealed source in the experiment room. (a) Time variation of the trigger rate as measured by the Compton camera (black solid line) and of the air dose rate as measured by the stationary dose rate monitor (blue dashed line). **(b)** Total energy spectra (the sum of energy deposits for each two-fold coincidence event) in **Figure 3a**'s periods (i) (red line), (ii) (blue line), (iii) (green line), (iv) (pink line) and (v) (black line), with the result of (iv) was scaled by 0.15. **(c)** 662 keV gamma-ray omnidirectional image superimposed on the optical image in period (i) (30 min). The red field indicates gamma-ray intensities in the upper half of the observed range. **(d)** Same as (c) but for period (ii) (30 min). **(e)** Same as (c) but for period (iii) (30 min). **(f)** Same as (c) but for period (iv) (30 min).

Figure 4: Same as Figure 3c, but with various measurement times: 3 s, 5 s, 10 s, 15 s, 30 s, and 60 s. Here gamma-ray images are identified by the red region, which indicates gamma-ray intensities in the upper 30 % of the observed range.

Figure 5: Representative results of environmental monitoring in front of a reception desk in the PET facility. (a) Time variation of the trigger rate as measured by the Compton camera (black line) during the daytime (5.6 h). **(b)** Trigger rate detailed for a period between 6200 s and 7000 s in (a). **(c)** Total energy spectra for **Figure 4b**'s periods (i) (red line), (ii) (blue line) and (iii) (black line). **(d)** 511 keV gamma-ray omnidirectional image superimposed on the optical image for period (i) (2 min). **(e)** Same as (d) but for period (ii) (2 min).

Figure 6: Representative results of outdoor measurement in Kawamata-machi, Fukushima, Japan. (a) Time variation of the trigger rate as measured by the Compton camera (black solid line). **(b)** Total energy spectra for **Figure 5a**'s periods (i) 1 min (blue line), (ii) 10 min (green line), (iii) 20 min (red line) and (iv) 30 min (black line). **(c)** Omnidirectional image of 605, 662 and 796 keV gamma-rays superimposed on the optical image for period (i) (1 min). **(d)** Same as (c) but for period (ii) (10 min). **(e)** Same as (c) but for period (iii) (20 min). **(f)** Same as (c) but for period (iv) (30 min). The dose rate values measured by a scintillation-type survey meter at a height of 1 cm from the ground are shown in the figures for comparison.

Table 1: Performances of present and previous Compton camera systems. *The angular resolution was estimated from 511 keV omnidirectional gamma ray images obtained during

measurement of a ^{22}Na sealed source (0.8MBq) placed 1 m ahead of the detector.

DISCUSSION:

We presented three experimental protocols for visualizing various low-level gamma radiation sources using the omnidirectional Compton camera that we developed. The representative results demonstrated that gamma-ray imaging at low radiation levels permits derivation of novel and useful information on the surrounding environment. In the RI facility, the protocol revealed that our Compton camera system successfully discovers the position of the gamma-ray source, as well as the counting rate at the given position relative to the source. This means that the proposed method can serve as a next-generation technology for environmental radiation monitoring, replacing conventional stationary dose rate monitors already mounted on the walls of almost any RI facility. In this paper, we depicted gamma-ray intensity as a red field mapping the region experiencing intensities in the upper half of observed values (**Figure 3**, **Figure 5**, and **Figure 6**), so as to suit various purposes without bias. An approach that rather gives priority to a peak intensity, rather than to the distribution of gamma-ray sources, would adopt a narrower range of the red field, perhaps the upper quarter of observed values, in order to enable directive findings at shorter timescales. Indeed, in **Figure 3c**, the peak direction could be identified with a measurement time of 30 s for case (i) as shown in **Figure 4**, for which the peak position's intensity was around 20 counts.

As for environmental monitoring in the PET facility, the protocol demonstrated the possibility of visualizing the radioactivity movement through the facility, which in this case is considered to be the movement of a patient injected with ^{18}F -FDG. In **Figure 5d,e**, the direction of the patient can be identified in less than 10 s by adopting the narrower red field range as mentioned above. In the future, the environmental monitoring of gamma-ray sources by animation would be useful for various situations, not only for the movement of patients as in this study, but also for monitoring the transfer of nuclear fuel materials such as in airports for purposes of terrorism, by taking advantage of the high-sensitivity and low-cost characteristics of the system, although the energy resolution of a system that uses a scintillator is inferior to that of more expensive semiconductor detectors, such as high purity germanium (HPGe) and CdZnTe (CZT).

In the Fukushima field, the protocol successfully visualized the extended gamma radiation source with surface dose rates of much less than 1 $\mu\text{Sv/h}$, which is an order of magnitude lower than that in a recent report^{25,26}. Our Compton camera system was found to be capable of operating stably and robustly for outdoor measurement. We have already confirmed that the system can be operated by using WiFi and portable battery for more convenient use in various situations, especially for outdoor measurement. The Ministry of Environment in Japan has set the air dose rate minimum at 0.23 $\mu\text{Sv/h}$ to designate areas to be decontaminated. We believe that our system and protocols will be a great help for the decontamination procedure in areas of low-level radioactive contamination in broad areas of eastern Japan where radioactive cesium was released by the Fukushima Daiichi nuclear power plant accident in 2011.

The Compton camera used in this study has high sensitivity for gamma rays with energies between 300 keV and 1400 keV, attributable to the use of 3.5 cm CsI(Tl) scintillator cubes¹⁸.

Scintillator type and size can be optimized for environmental monitoring of low-level gamma radiation sources below 300 keV, such as ^{99m}Tc (141 keV) and ^{111}In (171 keV, 245 keV), which are frequently used in scintigraphy. This work will be presented in another paper in the near future. The detector can be manufactured at a low price. In fact, the cost of the detector materials used in this study was no more than \$20,000, and this amount was dominated by the price of the counter consisting of CsI (TI) and PMT; this configuration is significantly less expensive than the GAGG scintillators and HPGe semiconductor detectors that are used in other Compton cameras. Furthermore, the system used in this study should be made more compact for the sake of versatility and convenience. The size of the system produced in this study was 30 cm x 25 cm x 40 cm, which is larger than the existing portable gamma camera^{5,27}. The main reasons for such large system size are the large size of the PMT attached to CsI (TI) ($\phi 4\text{ cm} \times 12\text{ cm}$) and the large electronics handmade by us. In the future, portability will be improved by replacing the PMT with a metal package PMT or Silicon Photomultiplier (SiPM) as well as by repackaging the electronics at small size.

ACKNOWLEDGMENTS:

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DISCLOSURES:

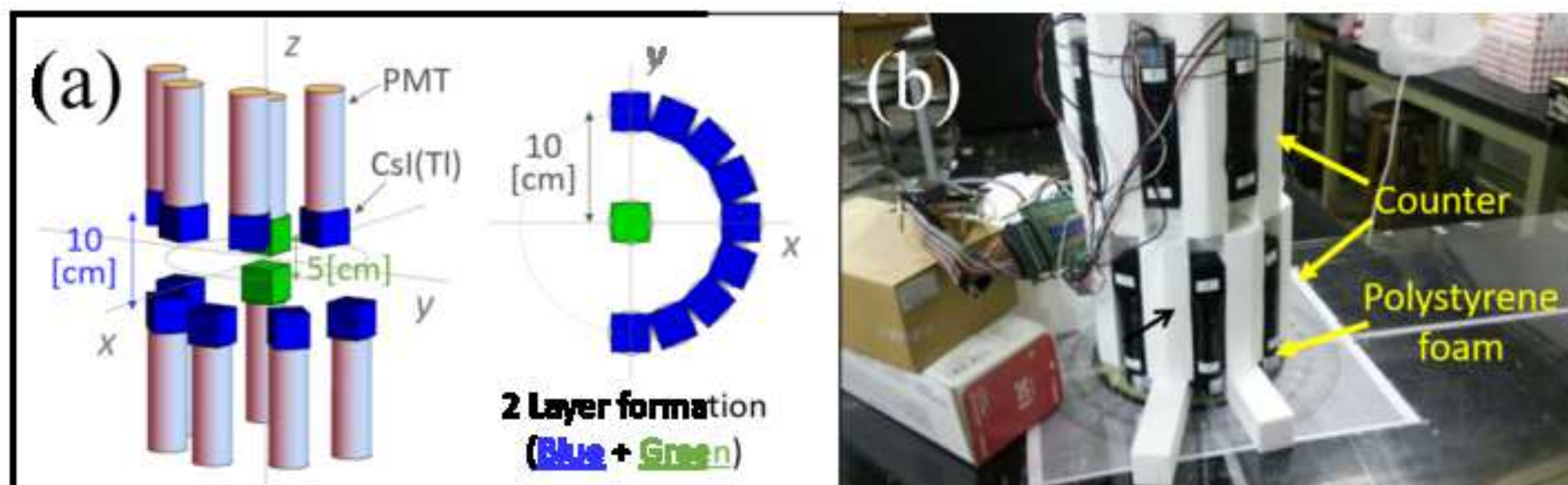
The authors have nothing to disclose.

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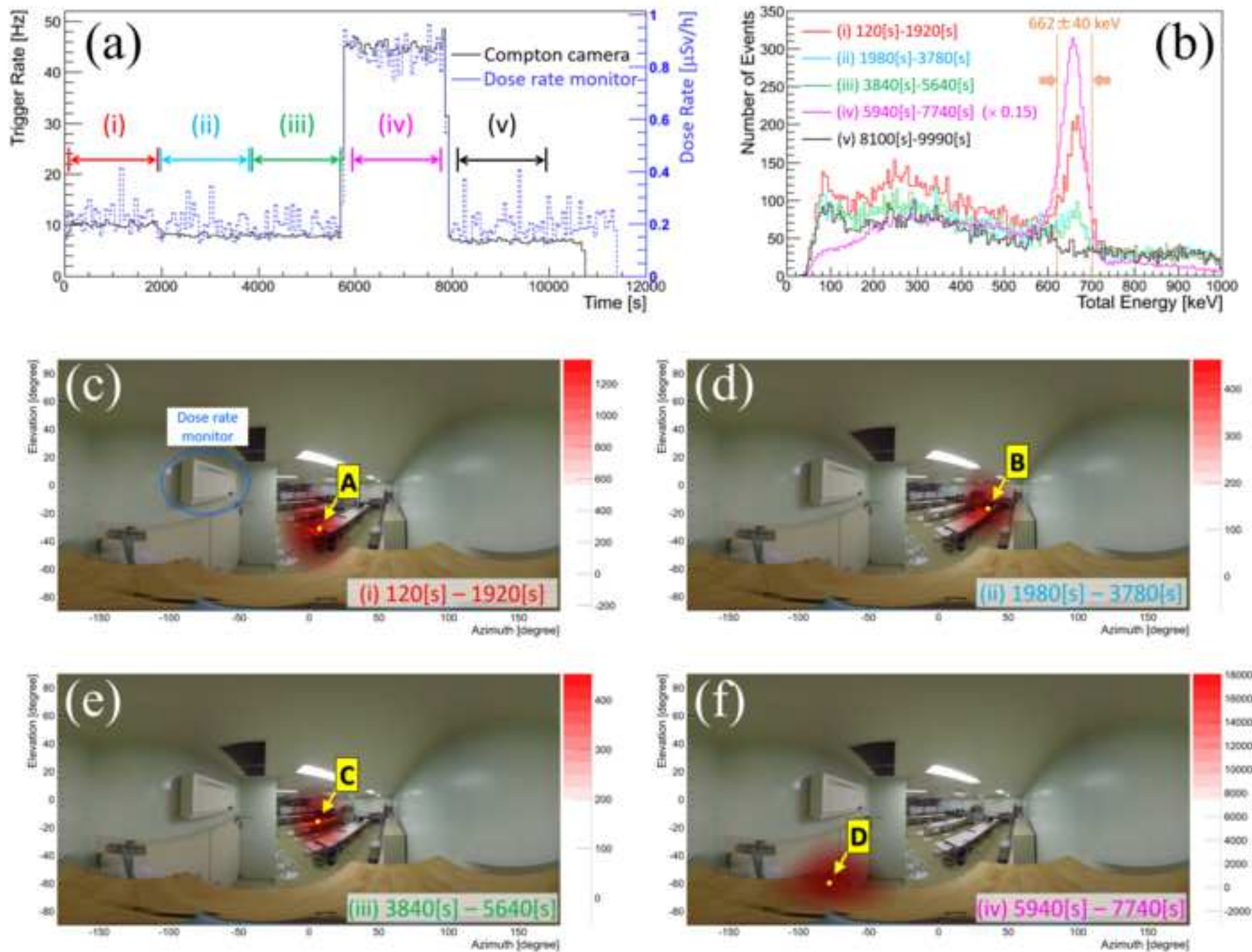
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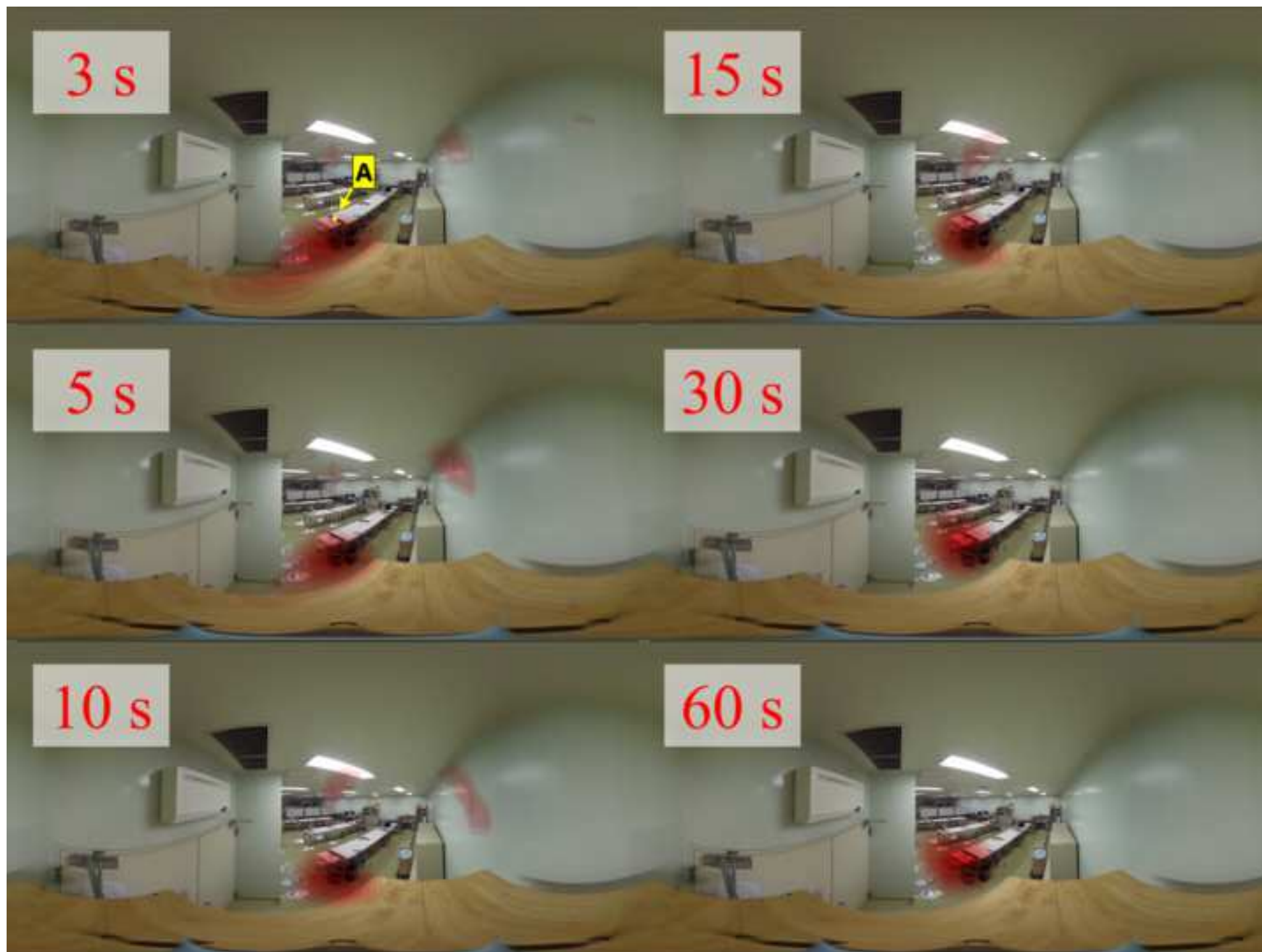
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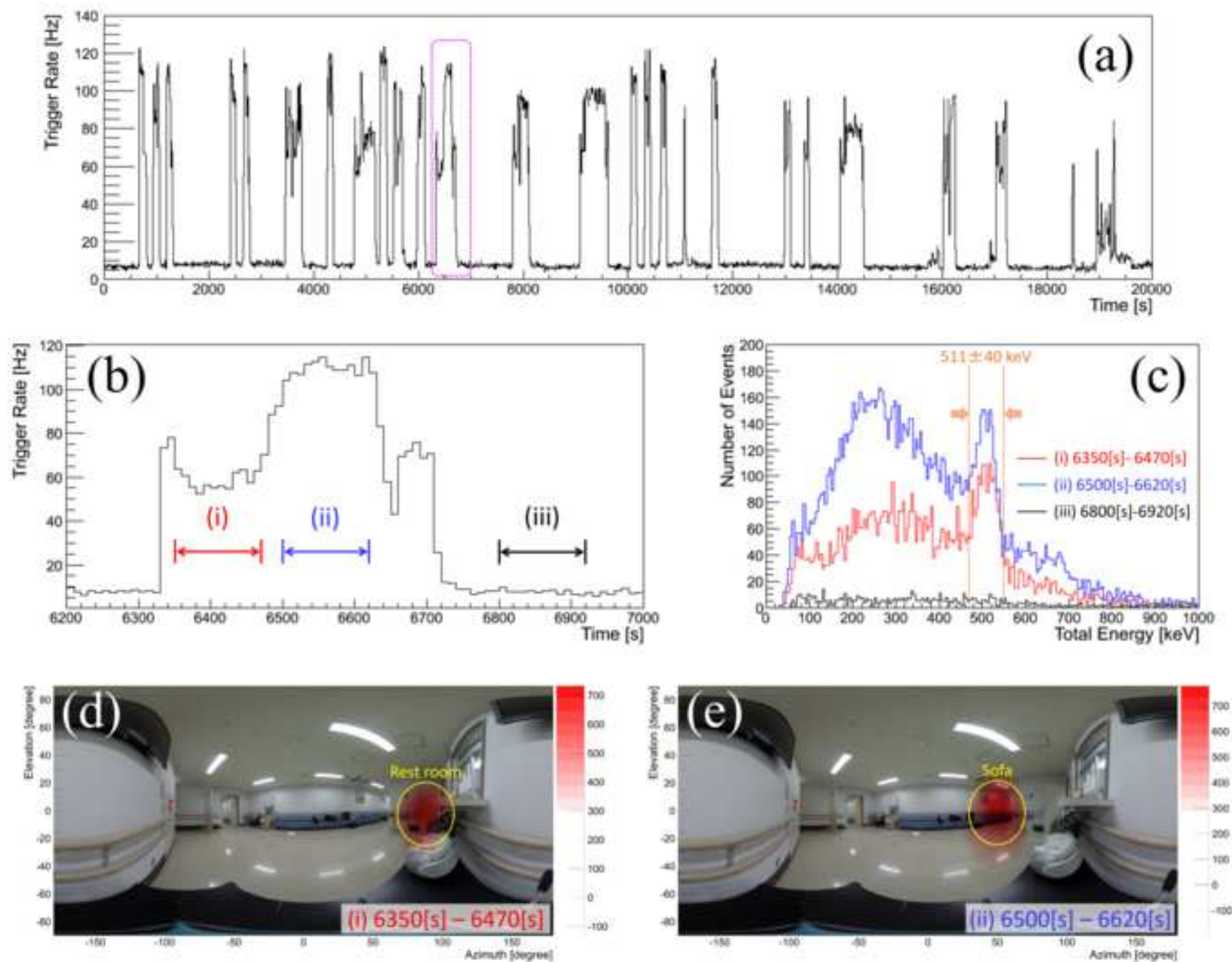
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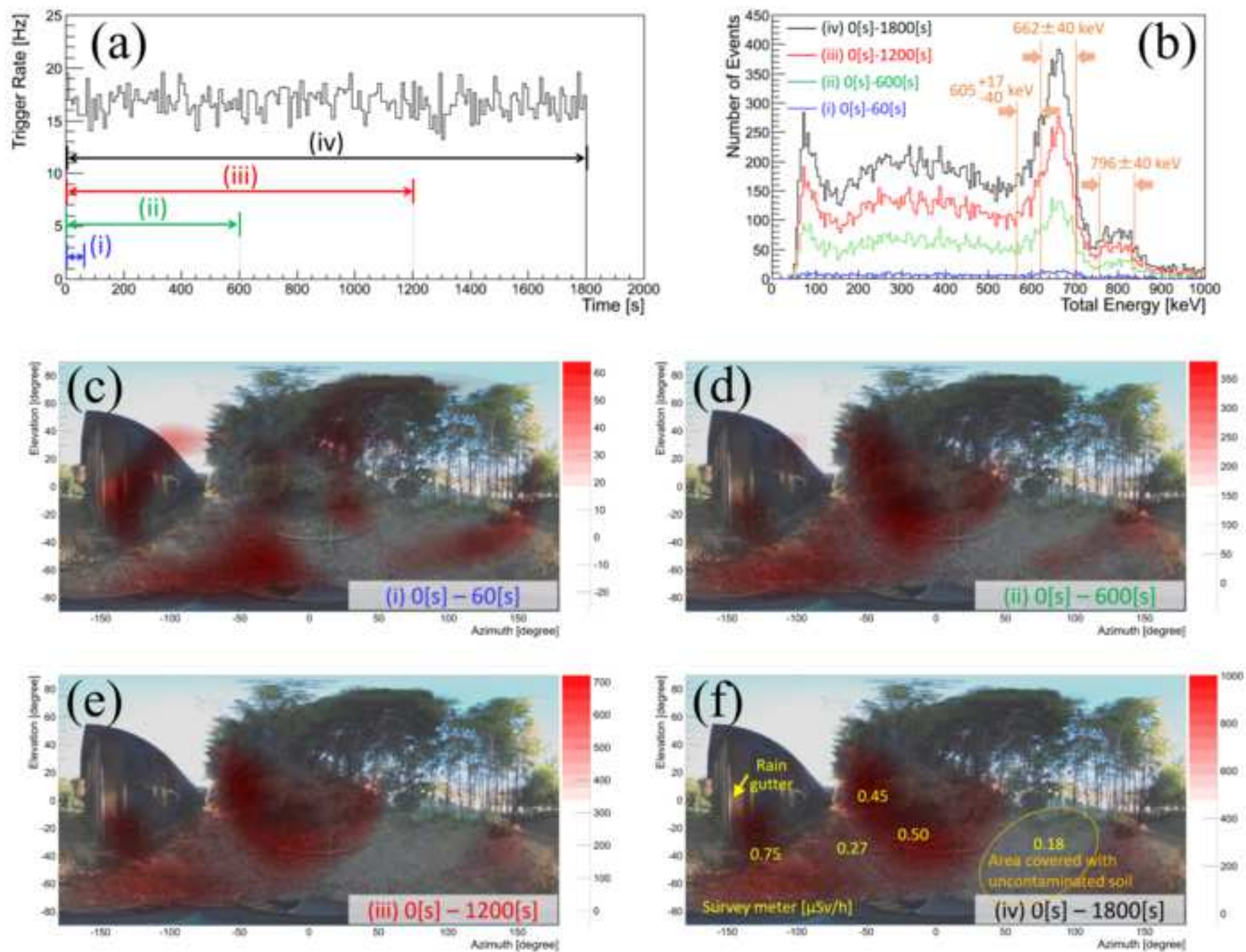












	This study	Previous study ¹⁸⁾
Number of counters	11	6
Detection efficiency (cps/(μ Sv/h)) for 511 keV gamma-rays	36	18
Angular resolution σ (deg)*	11	11

>*****

>Reply to Editorial comments:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

An English editing service has proofread the entire paper.

>2. Please include an ethics statement before the numbered protocol steps, indicating that the protocol follows the guidelines of your institution’s human research ethics committee.

In line 138, we inserted a new statement: “The protocol was conducted following the guidelines of the research ethics committee at the National Cancer Center Hospital East, Japan.”

>3. For in-text referencing, the superscripted reference number should be inserted before a comma or period. Please also remove the parentheses after the reference numbers.

We have made modifications by following your suggestion.

>*****

>Reply to Reviewer #2:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>Major Concerns:

>Thank you for making the suggested changes. The new figure is helpful and the paper is more informative now. I see that this may be a useful instrument. Though it might not be the state of the art, it's low cost and simplicity might make it good for a field instrument. The examples you have presented show that you can get reasonable results. However, I still am having a hard time

imagining that the protocols you present are useful to others. What you are presenting are examples of uses, not generally applicable procedures.

Low-dose γ -ray source imaging of 1 $\mu\text{Sv/h}$ or less in the environment has been overlooked so far in the field of radiation protection, and has not been reported or achieved elsewhere so far. Therefore, we hope you understand that we have presented new protocols, not examples of uses.

>Minor Concerns:

>Table 1: The correct way to measure resolution is to use two sources and determine the minimum separation required to resolve both. I'm suspicious that your algorithm might be shrinking spots beyond what you can actually resolve. Can you convince me otherwise?

For the evaluation of angular resolution, we simply followed the method by Watanabe et al. (2018) for comparison with previous studies. As described in this paper, the angular resolution of this detector (11 ° for 511keV gamma rays) is consistent with the expected intrinsic angular resolution which was dominated by the geometrical effect due to the crystal size and the interval of crystals. For further details on the novelty of this measurement technique, please refer to our previous paper (Watanabe et al. JJAP (2018)).

>Lines 92-93: In the section of the angular dependence, what angles did you compare? Does this include directly above and directly below as well? What exactly varies by ~4%? Is it the total efficiency, imaging efficiency, resolution,...?

The angle dependence of the efficiency was evaluated by comparing the peak values of the reconstructed images of the ^{22}Na sealed source installed in various directions for each according to the method by Watanabe et al. (2018).

>*****

>Reply to Reviewer #5:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>Major Concerns:

>- The paper lacks additional background on detector construction and setup.

The purpose of this paper is to show the procedure and representative results for visualizing low-level gamma radiation sources in various environments not previously demonstrated elsewhere “according to the aim and scope of JoVE.” Therefore, for further details on the novelty of this measurement technique, please refer to our previous paper (Watanabe et al. JJAP (2018)). We remark that in the previous revision, we moved the description of the detector overview from a first paragraph in “Protocol” to the last part of “Introduction” according to the editor's instructions.

>- The quality of the images are low. Higher quality optical images are advised.

The low resolution of the image is due to the resolution of the pdf file that is automatically generated on the JoVE web submission system when posting. The resolution of the original image is high enough.

-> A more rigorous description of data processing in this section should be included as the reader might not understand what criteria of time binning is used. In the source localization completed in the lab, time binning is completed with a priori knowledge, which might not be possible in a real world scenario. Perhaps expand on the technique by creating a new image for a 5 minute integration for example.

We apologize for our poor explanation of this point. In line 91, we inserted the following new statement: “During the measurement, the trigger rate, total energy spectrum (the sum of the energy deposits for each two-fold coincidence event), and the reconstructed images of a preset gamma-ray energy can be displayed on the online PC screen. This information can be updated at a preset time interval (e.g., every 10 s). Here, we set the screen to display two types of reconstructed images: an image that is accumulated at the start of the measurement and an image reaccumulated at every preset time interval (e.g., every 1 min). Furthermore, because the raw data for each event obtained using the measurements are stored, it is possible to reanalyze the data after the measurements and then regenerate a reconstructed image for an arbitrary gamma-ray energy at an arbitrary time interval.”

>Minor Concerns:

>- Usefulness of monitoring in PET facility is not highlighted.

In this study, we introduced an example of the possibility of visualizing the dynamics of remote radioactivity using gamma ray imaging technology from environmental radioactivity measurement data in a PET facility. In the new manuscript, we have made modifications with respect to the

movement of a patient as described below:

- (a) In line 34, we deleted a statement “Furthermore, we successfully visualised the movement of a patient injected with ^{18}F -fluorodeoxyglucose (^{18}F -FDG) in a nuclear medicine facility.”, and we inserted a new statement: “Furthermore, we successfully demonstrated the possibility of visualising the radioactivity movement in the environment, for example, the movement of a patient injected with ^{18}F -fluorodeoxyglucose (^{18}F -FDG) in a nuclear medicine facility.”
- (b) In line 190, we modified “attribute” to “could be attributed”.
- (c) In line 288, we deleted a statement “movement through the facility of”, and we inserted a new statement: “radioactivity movement through the facility, which in this case is considered to be the movement of”.

>*****

>Reply to Reviewer #6:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>Major Concerns,

>1. It said omnidirectional camera but, in fact, it is a semi-sphere or 2π FOV, not 4π (pi). The word 'omnidirectional' should be changed for over all. Check " 4π FOV compact Compton camera for nuclear material investigations", NIMA, 652(1):33-36 as a reference.

As described in the Introduction, the “omnidirectional” Compton camera technique used in this study is based on the successful development of a high-sensitivity “omnidirectional” Compton camera technology that can easily visualise low-dose γ -ray sources in the environment (Watanabe et al., JJAP, 57, 026401, 2018). In the new manuscript, in order to explicitly cover the background of this study, we added the following new references which were cited in line 58:

- 13) Lee, W., Lee, T. 4π FOV compact Compton camera for nuclear material investigations. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*. 652, 33-36 (2011).
- 14) Yamaya, T. et al. Concrete realization of the whole gamma imaging concept, *Proceedings of 2017 IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC)*. 3pp (2017).

>2. This paper used CT reconstruct. However, CT reconstruction using line projection is different from Compton imaging using cone projection. This should be discussed in the paper. Check "L. Parra, IEEE Trans. Nucl. Sci. 47, 1543 (2000)" for analytic Compton reconstruction.

In the new manuscript, we have made modifications to specify these points as described below:

- (a) In line 59, we inserted a new statement: “, as well as image reconstruction techniques for Compton cone data by analytical^{15,16} and statistical¹⁷ approaches.” We also added the following new references:
- 15) Parra, L. Reconstruction of cone-beam projections from Compton scattered data, IEEE Transactions on Nuclear Science. 47, 1543-1550 (2000).
- 16) Xu, D., He, Z. Filtered Back-Projection in 4π Compton Imaging With a Single 3D Position Sensitive CdZnTe Detector, IEEE Transactions on Nuclear Science. 53, 2787-2795 (2006).
- 17) Wilderman, S., Clinthorne, N., Fessler, J., Rogers, W. List-mode maximum likelihood reconstruction of Compton scatter camera images in nuclear medicine, Proceedings of IEEE Nuclear Science Symposium, 3, 1716-1720 (1998).
- (b) In line 68, we deleted a statement “The technique accomplishes this by applying an image-sharpening technique^{18,20} based on the filtered back-projection algorithm used in image reconstruction for computed tomography (CT).”, and we inserted a new statement: “This is accomplished through the application of an image-sharpening technique^{18,20} based on the filtered back-projection algorithm, which applies a convolution filter used in image reconstruction for computed tomography to the Compton reconstruction.”

>3. Line 92: How 11 degrees are measured? In order to evaluate angular resolution, not a single source but at least two sources should be used since the FWHM of a reconstructed single source is affected by filter or iteration number. Therefore, two sources with various positions (such as 5', 10', 20'....) should be reconstructed to estimate the angular resolution of the system. The minimum angle in which two sources are discriminated from each other, can be the real angular resolution of the system.

We understand your opinion. On the other hand, in this study, the angular resolution was evaluated according to the method in the previous study (Watanabe et al. 2018), which is already described in the caption of Table 1 (line 265) as follows: “The angular resolution was estimated from 511 keV omnidirectional gamma ray images obtained during measurement of a ²²Na sealed source placed 1 m ahead of the detector.”

>4. How could the authors determined the sequence of radiation interactions which at least more

than two? There are a few methods to determine the sequence, and hence, the method for determine the sequence of the interaction should be described.

Please refer to our previous paper (Watanabe et al., JJAP, 57, 026401, 2018). In this technique, we don't need to distinguish which counter is a scatterer or an absorber.

>5. Since the energy resolution of the system using scintillator is at least ten times lower than that of semiconductors such as HPGe and CZT. It is hard to measure multi-energy sources like nuclear fuel materials. This should be described in the text.

In line 293, we inserted the following new statement with respect to this point: “, by taking advantage of the high-sensitivity and low-cost characteristics of the system, although the energy resolution of a system that uses a scintillator is inferior to that of more expensive semiconductor detectors, such as high purity germanium (HPGe) and CdZnTe (CZT).”

>6. Line 265-267: What is the meaning of "upper half of observed values"?

It means the range of the red gradation field in the palette on the right side of Fig. 3(c)(d)(e)(f), Fig.5(d)(e) and Fig.6(c)(d)(e)(f).

>What does "a narrower range of the red field" mean?

For example, it means the following description in the caption of fig.4: "gamma-ray intensities in the upper 30% of the observed range."

>7. Line 283: Two reference is not enough to say "state-of-the-art device". Did the authors compare the system with other Compton imager using semiconductors or difference structures?

In line 300, we simply delete the statement “using a state-of-the-art device” by following your suggestion. We have never made a direct measurement comparison with a semiconductor Compton camera.

>8. Line 296: The performance of Compton imagers is significantly degraded in low energy such as Tc-99 or In-111, and hence, coded aperture or pinhole is much better choice to reconstruct low energy radiation sources.

The possibility of visualizing low-energy gamma rays using this technique will present in another paper in the near future.

>*****

>Reply to Reviewer #7:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>Minor Concerns:

>On REPRESENTATIVE RESULTS 1 (line 163), the same peak heights for period (ii) and (iii) should be attributed to the same distance of 6.7m, not 3.6m, according to the experiment protocol mentioned above.

Thank you very much for your suggestion. In line 177, we corrected “3.6 m” to “6.7 m”.

>On REPRESENTATIVE RESULTS 3, the reviewer did not understand why the selecting energy range for 605 keV and 662 keV is (565-622), (622-702), but not the midpoint of them?

The event selection of 605 keV gamma rays for ^{134}Cs should be 605 ± 40 keV (565-645 keV), but the range of 622 keV-645 keV overlaps the range of 662 ± 40 keV (622-702 keV) for ^{137}Cs . Here, as can be seen from the energy spectrum in Fig. 5 (c), the activity of ^{134}Cs in the Fukushima field was sufficiently lower than that of ^{137}Cs in this measurement. This is because the half-life of ^{134}Cs (2 years) is sufficiently shorter than that of ^{137}Cs (30 years). Therefore, in this paper, the range of 622 keV-645 keV was treated as 662 keV gamma rays.

>On DISSCISION (line 273), figure 5 just shows the two gamma-ray images by acquiring 2min for two periods, but gives a conclusion that this protocol can visualize the movement of a patient injected with 18F-FDG. However, the ability to track the moving path of the patient by using this Compton camera is not demonstrated.

As described in the second paragraph of “Discussion”, the movement of the peak position of the gamma ray image considered to be a patient can be identified in the time frame of 10 seconds or less

in the case of Fig. 5 (d)(e). Further research on tracking the patient's path in the facility using multiple detectors will be discussed in other papers. In the new manuscript, we have made modifications with respect to the movement of a patient as described below:

- (a) In line 34, we deleted a statement “Furthermore, we successfully visualised the movement of a patient injected with ^{18}F -fluorodeoxyglucose (^{18}F -FDG) in a nuclear medicine facility.”, and we inserted a new statement: “Furthermore, we successfully demonstrated the possibility of visualising the radioactivity movement in the environment, for example, the movement of a patient injected with ^{18}F -fluorodeoxyglucose (^{18}F -FDG) in a nuclear medicine facility.”
- (b) In line 190, we modified “attribute” to “could be attributed”.
- (c) In line 288, we deleted a statement “movement through the facility of”, and we inserted a new statement: “radioactivity movement through the facility, which in this case is considered to be the movement of”.

>*****

>Reply to Reviewer #8:

>*****

Thank you very much for your comments. We have provided responses to each comment. In the new manuscript, we have used red font to indicate where we have made modifications.

>It is a description of results. So the results are nice but we must to confident about results presented (reconstructed spectra and localisation deduced about each time) and it is difficult to certified the quality of work because we don't know exactly the details of analysis

We apologize for our poor explanation of this point. In line 91, we inserted the following new statement to specify these points: “During the measurement, the trigger rate, total energy spectrum (the sum of the energy deposits for each two-fold coincidence event), and the reconstructed images of a preset gamma-ray energy can be displayed on the online PC screen. This information can be updated at a preset time interval (e.g., every 10 s). Here, we set the screen to display two types of reconstructed images: an image that is accumulated at the start of the measurement and an image reaccumulated at every preset time interval (e.g., every 1 min). Furthermore, because the raw data for each event obtained using the measurements are stored, it is possible to reanalyze the data after the measurements and then regenerate a reconstructed image for an arbitrary gamma-ray energy at an arbitrary time interval.”

>Remarks: referee hungry because don't have access to reference 13 to find more detail of reconstruction principle and performance.

For details on the image reconstruction method used in this paper, please refer to the following reference:

13) Watanabe, T. et al. Development of an omnidirectional gamma-ray imaging Compton camera for low-radiation-level environmental monitoring. Japanese Journal of Applied Physics. 57, 026401 (2018).

<https://iopscience.iop.org/article/10.7567/JJAP.57.026401/meta>

14) Kagaya, M. et al. Development of a low-cost-high-sensitivity Compton camera using CsI(Tl) scintillators (γ I). Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment. 804, 25-32 (2015).

<https://www.sciencedirect.com/science/article/pii/S0168900215010669>

**>it is presented like low cost but we don't have any information about the price
Material software and also the facility of use in practical.**

In line 314, we inserted the following new statement with respect to this point: “The detector can be manufactured at a low price. In fact, the cost of the detector materials used in this study was no more than \$20,000, and this amount was dominated by the price of the counter consisting of CsI (Tl) and PMT; this configuration is significantly less expensive than the GAGG scintillators and HPGe semiconductor detectors that are used in other Compton cameras.”

**>In general they don't give the activities of sealed sources used for several tests
Don't find in figure 3 picture associated at period V.**

The radioactivity of the ^{137}Cs sealed source used in “Protocol 1” is 3.85MBq as stated in the text. On the other hand, we forgot the description of the radioactivity of the ^{22}Na sealed radiation source used to measure angular resolution. Therefore, we inserted “(0.8MBq)” in line 267. The reconstructed image of period V was simply omitted because it was at the background level and there was no significant peak.

>Minor Concerns:

>More explanations about process about localization for accredited the method and validate the work

The capability of source localization was firmly confirmed by a lot of comparisons between the position of a radioactive source on the optical image and the peak of a gamma-ray reconstructed image.



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Author(s): Hiroshi Muraishi, Ryoji Enomoto, Hideaki Katagiri, Mikakagaya, Takara Watanabe, Naofumi Narita, Daisuke Kano

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