Development of Topological Method for Nuclear Forensics Image Data

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INTRODUCTION:

The Japan Atomic Energy Agency (JAEA) partnered with the U.S. Department of Energy's Office of Nuclear Smuggling Detection and Deterrence (NSDD) in the fields of safeguards, nuclear non-proliferation, and nuclear security under an agreement signed in 1988 between the U.S. and Japanese governments. Since 2011, JAEA and DOE-NNSA cooperatively carried out five projects as part of the nuclear forensics technology development in JAEA-ISCN (Integrated Support Center for Nuclear non-proliferation and Nuclear Security).

The most recent project in nuclear forensics involves a comparative study designed to improve the abilities of automated morphology software packages to achieve similar results. The ability of automated software to quantify morphological characteristics including particle size, aspect ratio, diameter, and circumference is an important tool in a nuclear forensic examination in support of a nuclear security investigation. Since these morphological parameters can be characteristic of material process history or origin, they elucidate potential signatures of the nuclear material under investigation. However, further method development is required to ensure consistency between image analyses protocols and computational software packages.

The four tasks of this on-going project include meetings to exchange procedures and plan the inter-laboratory comparison; the development of JAEA's computational tool for quantification of particle images; automated particle analyses and comparisons between the JAEA-developed image analysis tool and the Morphological Analysis for Material Attribution (MAMA) software, an export-controlled program developed at Los Alamos National Laboratory (LANL) and used by the U.S. laboratories. Included in this effort is the development of protocols for quantifying overall uncertainty of morphological measurements and general reporting. This manuscript will summarize the initial results from this on-going project that involved analysists from JAEA, using the JAEA-developed software, and LANL and Lawrence Livermore National Laboratory (LLNL), using the MAMA software.

EXERCISE DESIGN AND OBJECTIVES

The initial in-person meeting and subsequent virtual meetings were used to share particle analysis protocols, particle image data set requirements, and discuss powder sample preparation procedures. The resulting set of particle analysis and particle image data collection protocols have been utilized throughout the project. Four quantifiable particle morphology characteristics were identified for comparison: pixel area (particle size), aspect ratio, equivalent circle diameter (ECD), and pixel perimeter (circumference). The project plan facilitated the evaluation of these protocols in a stepwise manner with each task building upon the one prior. All tasks in this project were completed using National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 1984 particles adhered to a carbon substrate and imaged in a scanning electron microscope (SEM).[1]

The first project task compared the quantified particle morphology results from the newly developed JAEA software with that from the MAMA software. This was completed by sharing a set of ten SEM images collected at LANL with analysts at JAEA and LLNL. All analysts (JAEA, LLNL, and LANL) were to complete quantitative particle analyses compiling the results from all ten images so that the four particle characteristics could be compared. All analysts followed the image analysis protocol described here.[2]

- Exclude particles that overlap the edge of an image.
- Exclude particles that overlap each other unless one particle is clearly sitting on top of others. The particle sitting on top that has a clearly visible boundary may be included in the analysis.
- Exclude particles that have less than 100 pixels in an image.
- Measure particles at all magnifications.
- No particle may be included in the analysis more than once.
- Ideally, count greater than 700 particles. A minimum value is 500 particles.

The second task enabled study of the variability introduced in morphologic analyses due to multiple analysts imaging the same sample. In this task, a new SEM stub was prepared with SRM 1984 and imaged at LANL. The image set was shared with LLNL and JAEA. The stub was then sequentially shipped to and imaged at LLNL, and then shipped to and imaged at JAEA. The analysts at LLNL and JAEA endeavored to capture and analyze the same set of particles as originally imaged at LANL. The previously described image analysis protocol would be employed to complete the quantitative particle analysis and provide a second opportunity to compare the two software packages.

The third task studied the influence of sample preparation, and continued developing image collection and software analytical practices and capability. The task did not define required sample preparation practices. However, the analysts discussed sample preparation best practices. This discussion included topics such potential artifacts introduced by different preparation methods and equipment currently available, and what might be useful in future particle analyses. Discussion focused on methods and techniques that the analysts might utilize to ensure that the final SRM 1984 SEM sample was representative of the actual SRM 1984 particle population and ensure that the particles were well distributed on the SEM stub. All analysts prepared subsamples utilizing a process analogous to cone-and-quartering using weigh paper and spatulas to sprinkle an even smaller subsample onto the adhesive carbon mounting tape.[3]

The image collection protocol to be used during task three image collection follows.

- Capture images in which particles have well defined edges.
- Capture multiple images of multiple areas using a range of magnifications. Images should not overlap.
- Capture images of between 500 and 700 particles for an analysis. Avoid areas with significant fractions of overlapping particles.
- Ensure the smallest particles are captured so that they are represented by more than 100 pixels in an image. Ideally particles are represented by 10^3 to 10^4 pixels.
- Ensure scale bars are calibrated in the X and Y directions.

RESULTS

Analyses supporting the initial primary task revealed significant progress in the JAEA's software development. Comparisons of the four morphologic parameters revealed largely consistent results between the two sets of results. The primary and secondary differences that were able to be identified upon close study was the manner in which the JAEA software treated overlapping particles and subtle differences in how it traced particle boundaries, see Figure 1. Ongoing discussion of the method employed by U.S. analysts included clarification that overlapping particles were manually segmented.

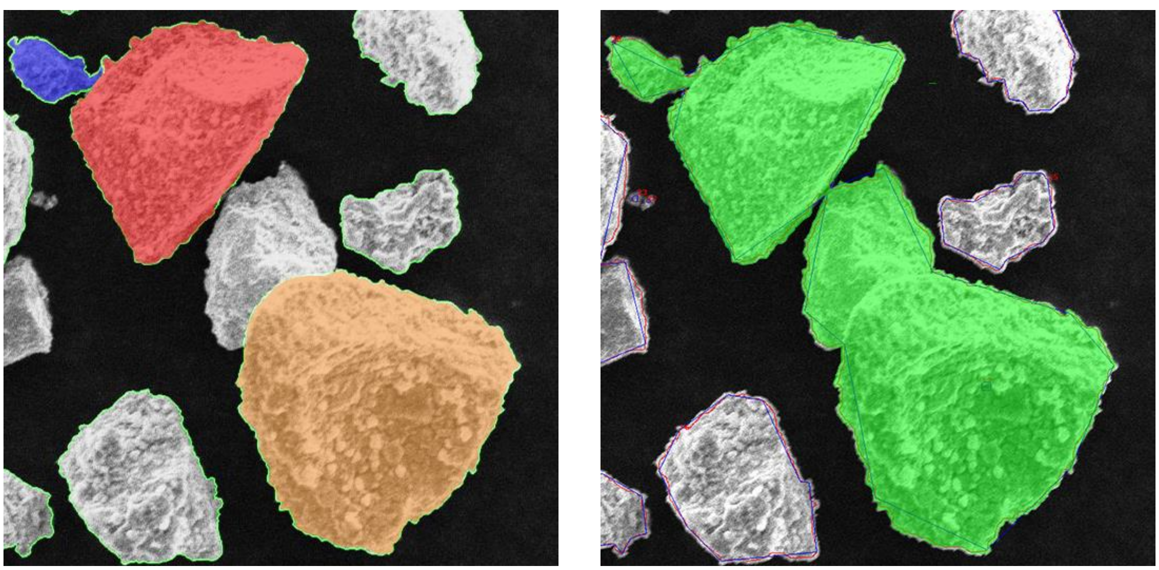

Figure 1. Example of primary task one results from the US and JAEA computational tools, left and right, respectively. [4]

The second task added and assessed variability introduced by multiple analysts imaging one sample. The analysts from LLNL and JAEA found it difficult to capture a completely identical set of images to those collected at LANL due to the challenge in finding the fiducial marks made on the sample. In spite of this challenge, LLNL and JAEA analysts were able to image and analyze the majority of the areas and particles imaged and analyzed at LANL. The morphologic parameter populations and characteristics generated from the three analysts image sets closely resembled those generated in the first task. This second data set also illustrated the increasing refinement of the JAEA software's particle tracing capability.

This set of data also revealed that there were still some differences between the JAEA and MAMA software in the process of separating overlapping particles. One image was selected and repeatedly modified with decreasing amounts of complexity, forming a set of simplified test images. The set of test images was then used to support further coding development of the JAEA software to refine its segmentation and perimeter definition algorithms. The original and unmodified image is shown in Figure 2. The plots that compare the four particle characteristics from all analysts are shown in Figure 3.

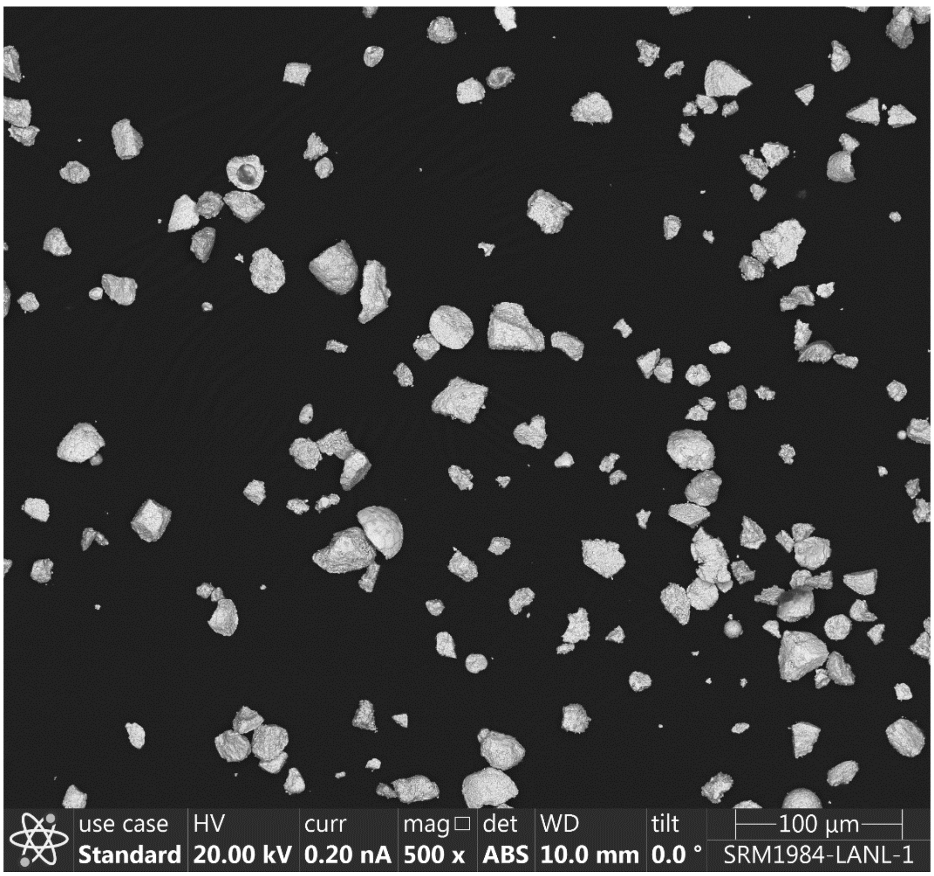

Figure 1. The base image used to provide step-wise investigation of image parameters that introduced variability into the JAEA software tool's particle characteristics. Plots containing the quantitative output from the JAEA and US DOE tools are shown in Figure 3.

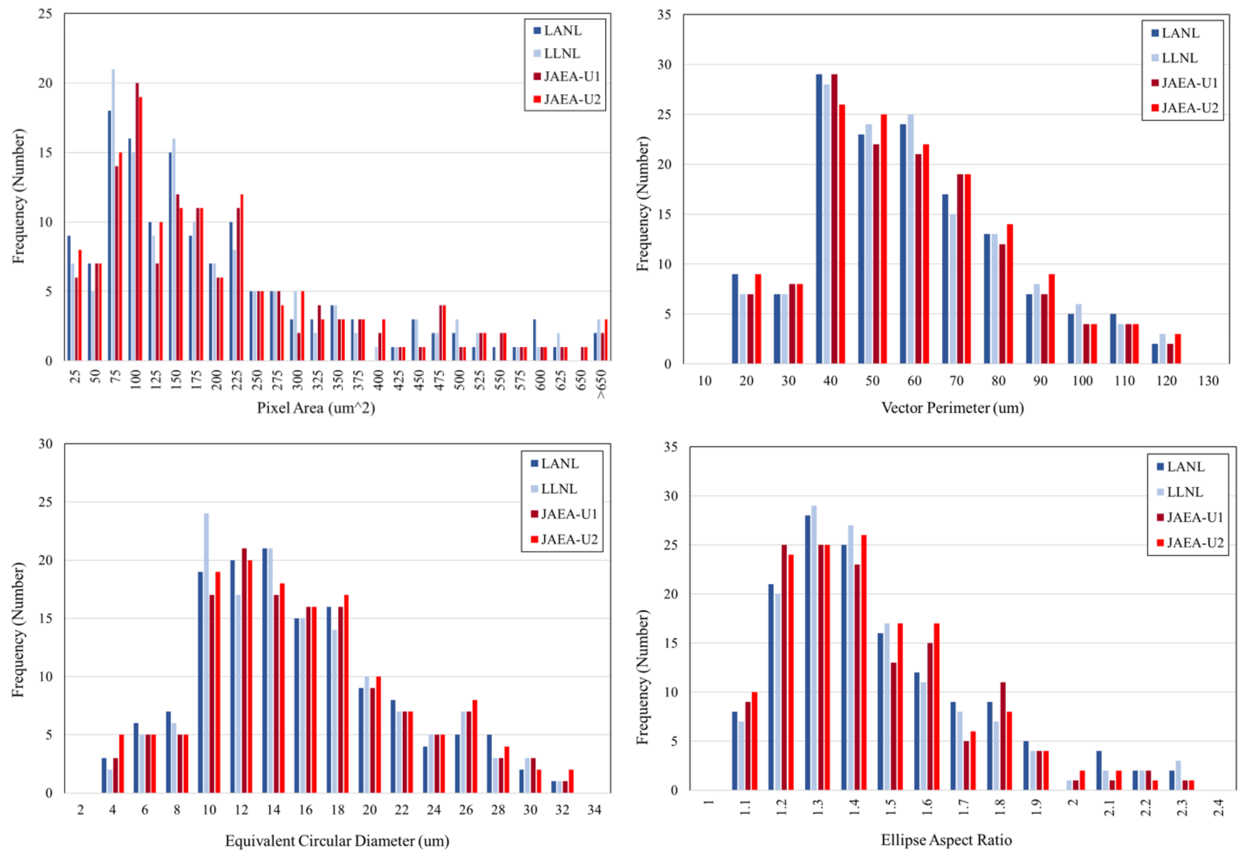

Figure 3. Comparison of the two computational tools' analytical output for the image shown in Figure 2. The number of particles analyzed and contributing to these plots by analyst is: LANL – 141; LLNL – 140; JAEA-U1 – 135; and JAEA-U2 – 143.

The final task of preparing an SEM sample and subsequently collecting and analyzing an image set suitable for particle analysis is ongoing. The plots of the initial results from the third task reveal that the individually prepared samples' morphologic characteristic are largely consistent with the first two tasks' results, see Figure 4. These plots include the full initial data set generated with LANL and JAEA images. The LANL analysis includes 1202 particles whereas the JAEA analysis includes 719 particles. The images collected by JAEA for this third task were reanalyzed in the MAMA software to provide a direct comparison of the two computational tools' output, see Figure 5. These direct comparison plots provide additional evidence that the two software packages return similar particle morphology analysis results.

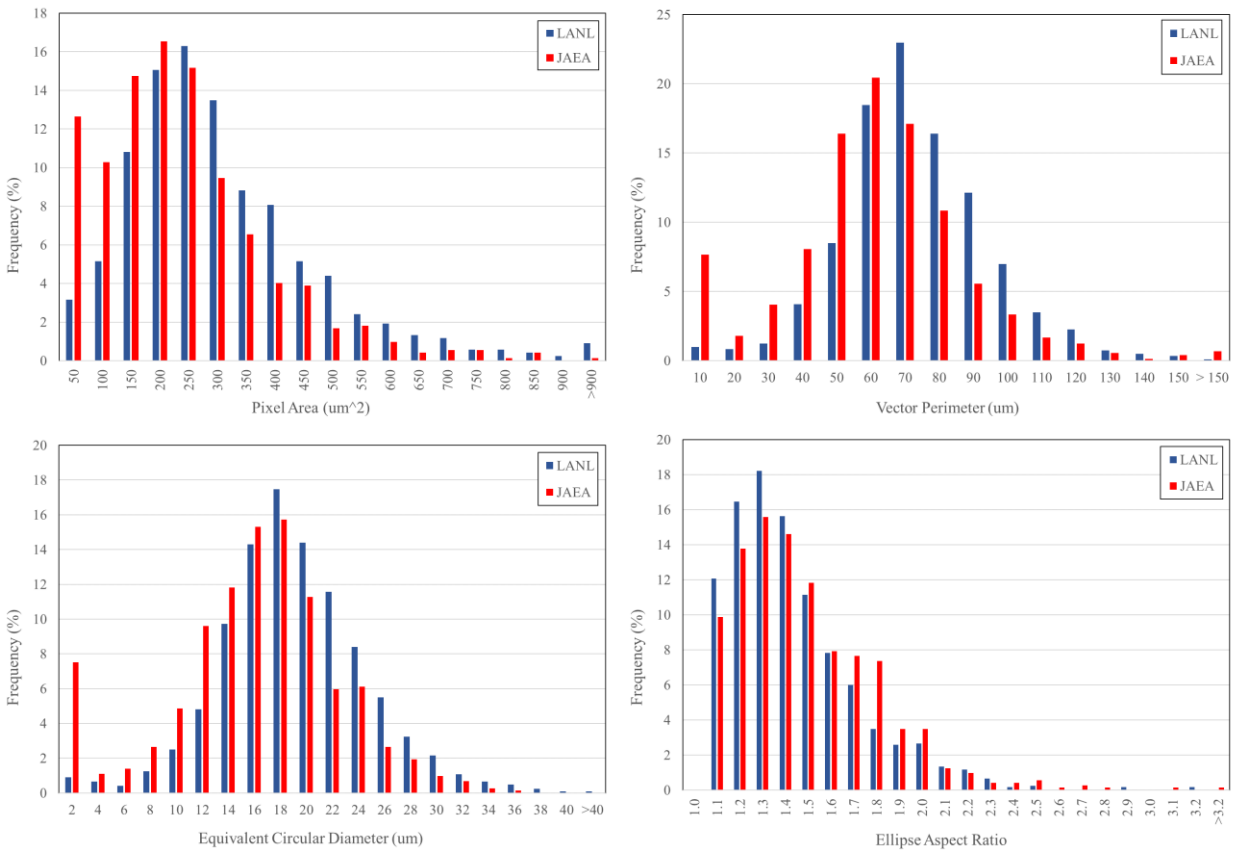

Figure 4. Initial comparison of morphology characteristics generated from the individually prepared samples of SRM 1984. Note that the distributions have been normalized as each analyst's particle set is of a different size.

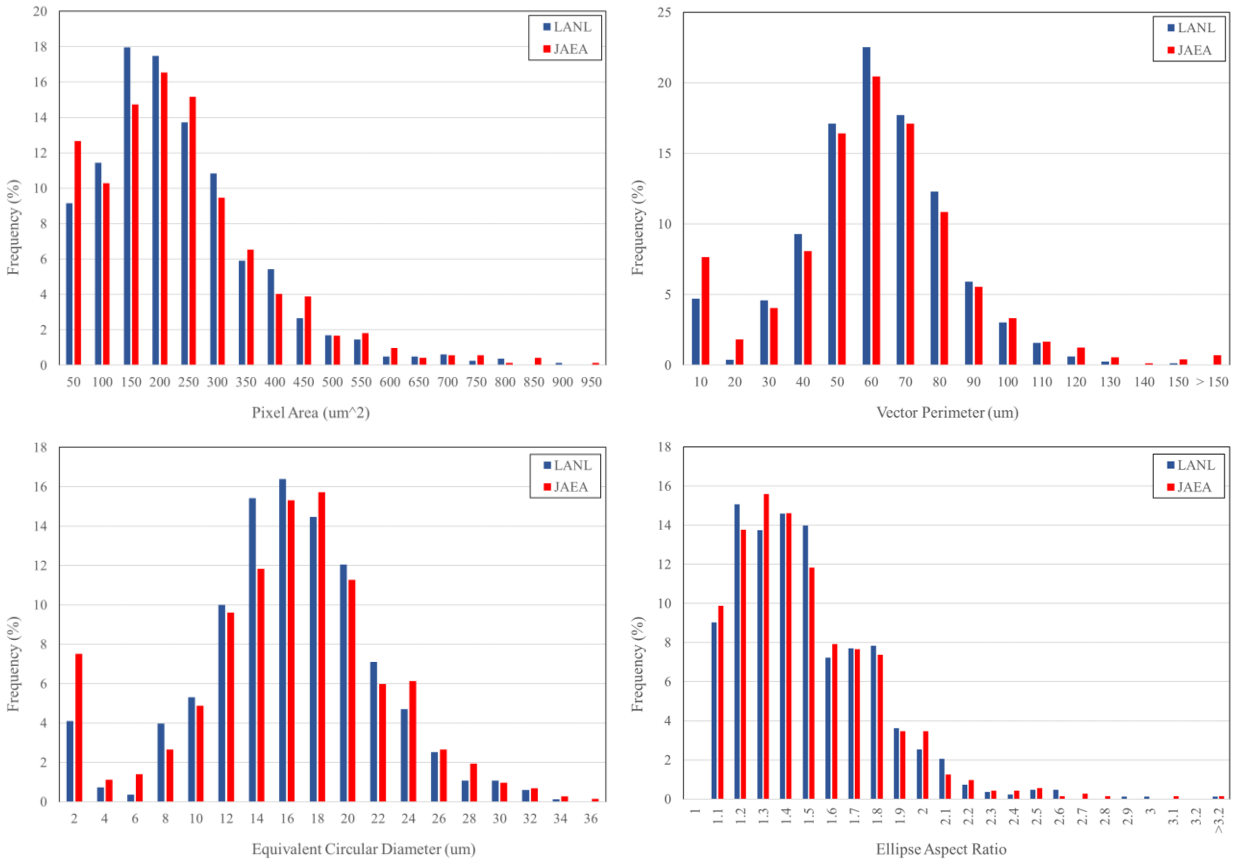

Figure 5. Images of JAEA's individually prepared sample were analyzed using the MAMA software and compared to the analysis from the JAEA software user.

CONCLUSIONS

The results from the first and second tasks of this project reveal that the new JAEA software and the established U.S. DOE MAMA software return generally similar results for the evaluated particle metrics. The third task, which was designed to compare the influence of sample preparation by different analysts as well as continue the comparison of morphological characteristics from the two software packages, suggests SRM 1984 is a reliable and robust test material, and confirms that the two software packages are converging on generating consistent results. These results encourage future work in developing enhanced sample preparation protocols based upon the results achieved with what may be considered an ideal sample, SRM 1984. Repeating tasks two and three with less ideal samples that more closely mimic samples that may be encountered in the international forensics community would provide significant benefit to the community.

The ability to objectively describe and quantify particle samples using digital images and image analysis software is crucial for national and international nuclear forensic evaluations, and will provide the international nuclear forensics community with increasing capabilities for determining sample origin and process history through its morphological characteristics. This project provides ample evidence that it is possible to develop software that returns similar results even if the codes are developed in a physically isolated manner and over a multi-year time span. Developers should anticipate that early software testing with real data will reveal opportunities to subtly refine code capabilities for particle segmentation and perimeter algorithms. All project planning and experimental design time investments made early in the project are beneficial. Initial project meetings will need to include discussion of many different aspects that contribute to particle analyses. Development of a test plan with sequentially increasing the complexity is vital to deconvolute the interaction of the software, sample, and analyst. Identification a suitable sample material (well characterized and readily available) and development data collection and analysis protocols further accelerate the successful development of the software. Utilizing the now ubiquitous virtual meeting platforms is very helpful when project participants are unable to meet frequently in person.

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REFERENCES

1. SRM 1984; *Thermal Spray Powder – Particle Size Distribution Tungsten Carbide/Cobalt (Acicular)* National Institute of Standards and Technology; U.S. Department of Commerce: Gaithersburg, MD 28 September 2015.
2. A.R. Ross, L. Tandon, and D.S. Schwartz, "Protocols and Uncertainty Determination Efforts for Morphological Analysis", LA-UR-17-29680, (2017)
3. A.R. Ross, L. Tandon, and D.S. Schwartz, "Practical Aspects of Sampling and Data Acquisition", LA-UR-17-29687, (2017)
4. M.M. Schneider, A.R. Ross, "Summary of the Results of Action A: Image Analysis Comparison", LA-CP-18-20622, (2018)