Storage of nuclear waste suitable for non-invasive monitoring using muon scattering tomography

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Importance of Nuclear Waste Imaging

- Nuclear waste drums inside a concrete vault at the ANDRA CSA waste disposal facility in Aube, France. These drums contain short-lived low- and intermediate-level waste. The vault is back-filled with concrete as new layers of drums are added; when full, it is sealed for a minimum of 300 years.
- Nuclear waste is often stored in steel, concrete-filled waste drums and disposed of at surface level or shallow underground sites.

Ref: PhD thesis, Michael J Weeks, Univ. of Sheffield, March 2023

- There are historical nuclear wastes that were produced at an era when records were not necessary and the issue of disposal of nuclear waste was not as urgent as it is now.
- The waste materials kept may have undergone changes such as uranium oxidizing to form hydrogen gas. In addition, their original composition may not be known.
- Nuclear wastes being generated in recent times, and temporarily stored, are adequately characterized, with records being kept, as mentioned above. However, they also need routine examinations.
- Due to constant emission of heat and radioactivity from these wastes, it is not logical to monitor and maintain these nuclear wastes with human intervention.
- There is a need for methods to characterize nuclear waste that ensures both that its records are up to date, and that historical waste can be carefully characterized, and properly evacuated for final disposal, or long-term storage.

Muon imaging can be considered as one of the possible solutions

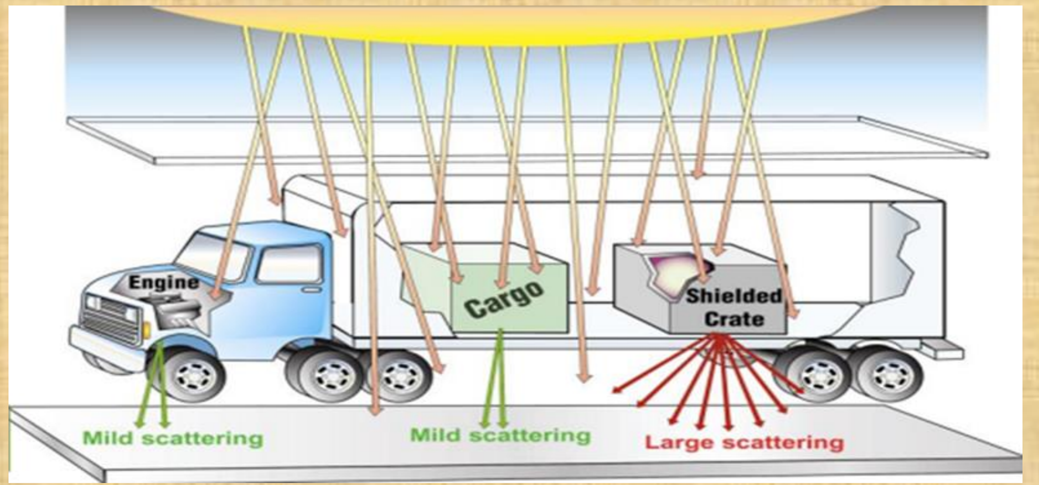
- ➢ Muon tomography or muography is a technique that uses cosmic ray muons to generate two or three-dimensional images of volumes using information contained in the Coulomb scattering or absorption of the muons.
- ❖ Application: Examining cargo containers, nuclear waste, archaeological / civil structures, monitoring volcano eruptions etc.

Muography

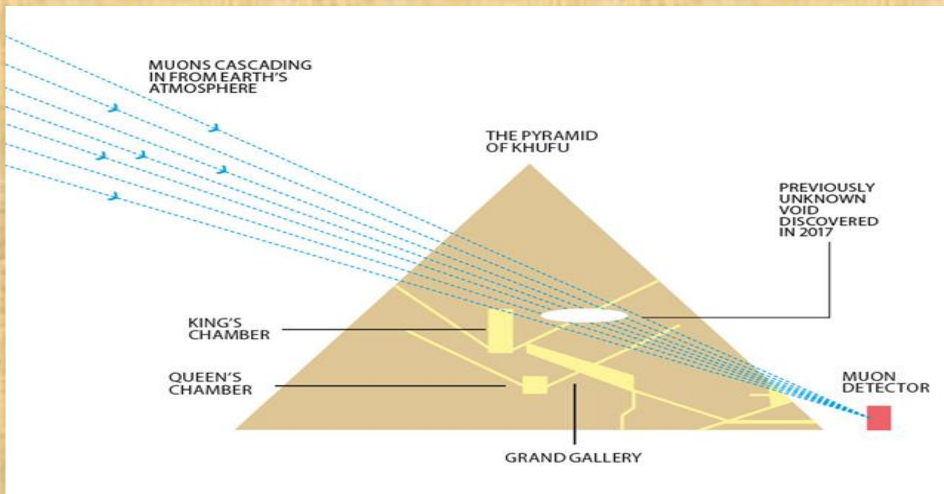
Scattering Muography | Absorption Muography

- **Muon Scattering Tomography (MST)**
  - ➢ Muon deviates due to multiple Coulomb scattering while passing through intervening matter. Deviation of muon is obtained by placing detectors on either side of the target region. This method is known as MST.

- **Absorption Muography(AM):**
  - ➢ Muons lose significant energy, leading to their absorption, traveling large distances inside matter. Comparing muon flux for 'free-sky' and target, image map can be constructed. This method is known as AM.

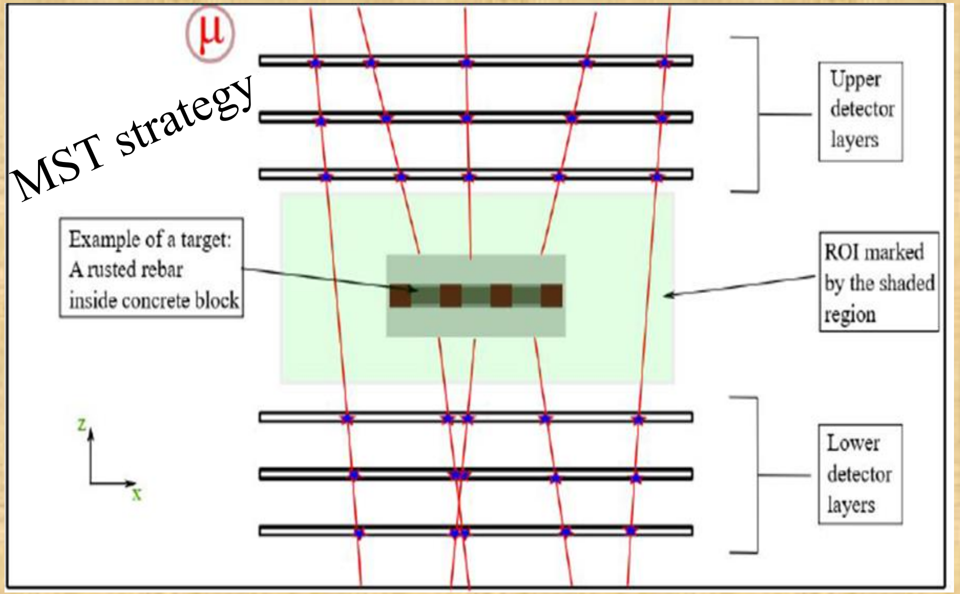


Schematic of MST employed in cargo inspection



Schematic of AM to scan pyramids

MST employed to image nuclear waste storage

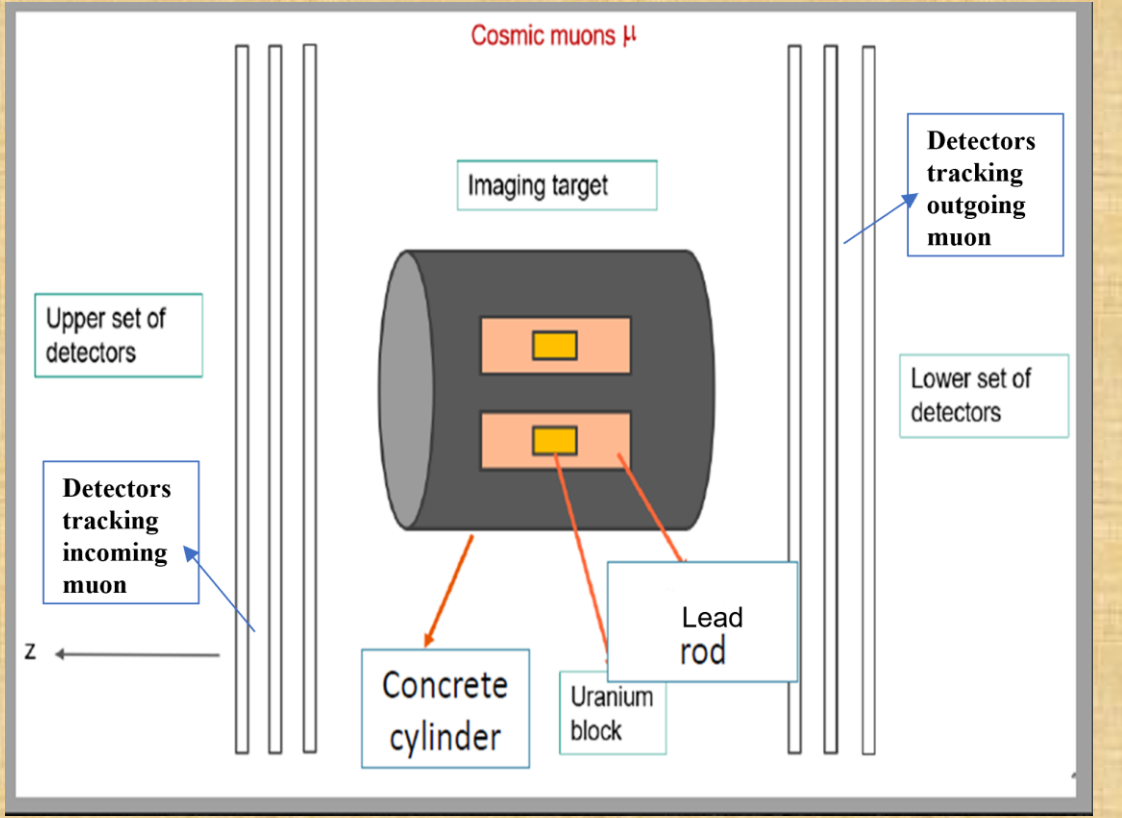


$$X = \frac{716.4\,\mathrm{g/cm^2}\,A}{\rho Z(Z+1)\ln\left(287/\sqrt{Z}\right)}$$

$$\theta_0 = \frac{13.6\,\mathrm{MeV/c}}{p\beta}\sqrt{\frac{L}{X}} \times \left(1 + 0.0038\left(\frac{L}{X}\right)\right)$$

$$\frac{dN}{d\theta_x} = \frac{1}{\sqrt{2\pi}\theta_0} e^{-\theta_x^2/2\theta_0^2}$$

p = Momentum
$\beta = v/c$
X = Radiation Length
L = Thickness
A = Atomic Mass
Z = Atomic Number
ρ = Density



Schematic of imaging of nuclear waste storage

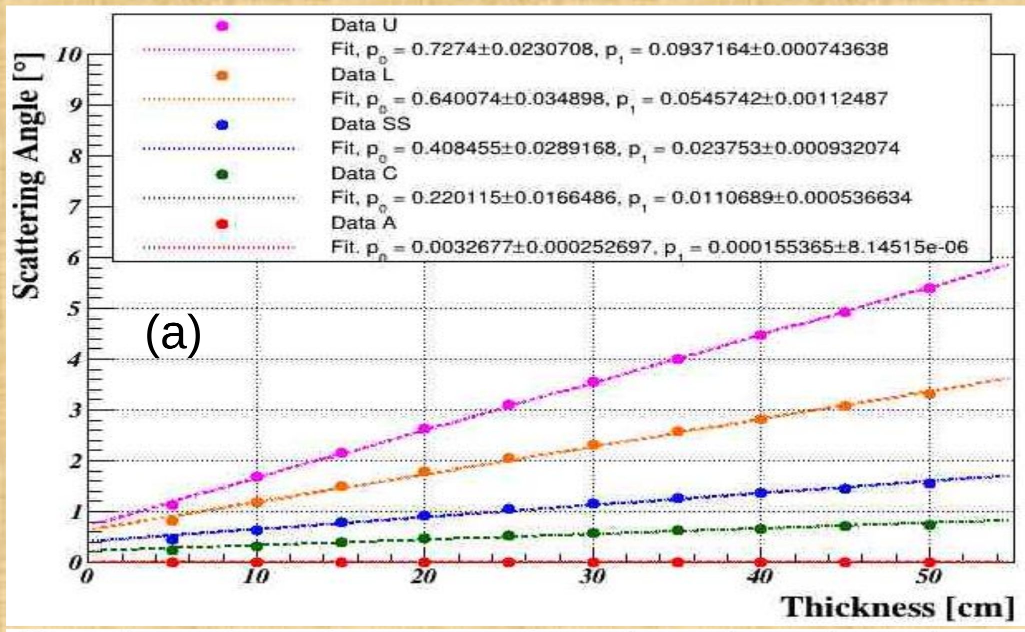
Uranium, lead, stainless steel, air (instead of hydrogen gas) enclosed by a concrete cylinder.

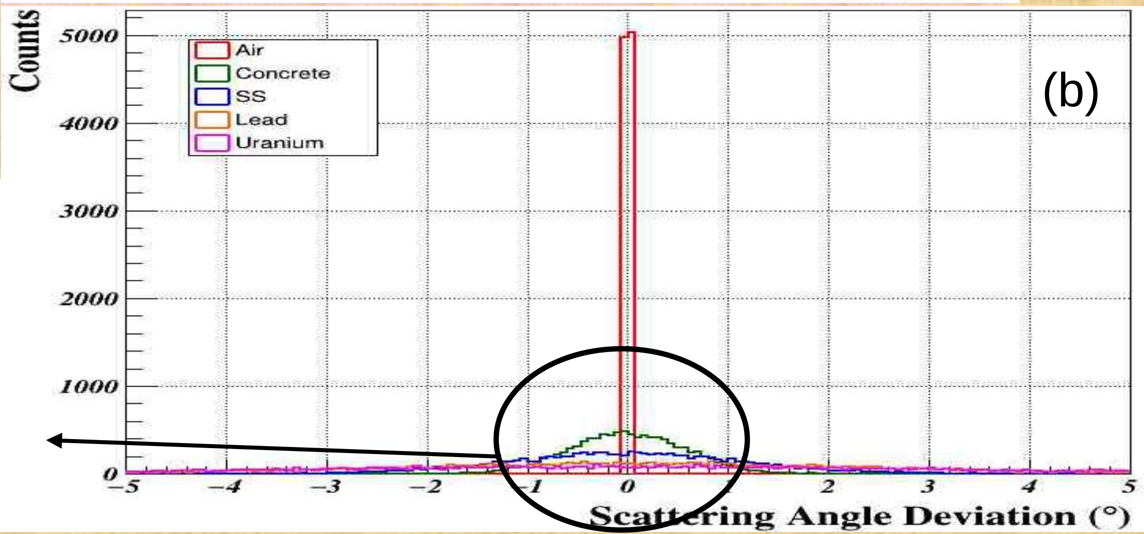
Scattering angle distribution w.r.t material thickness

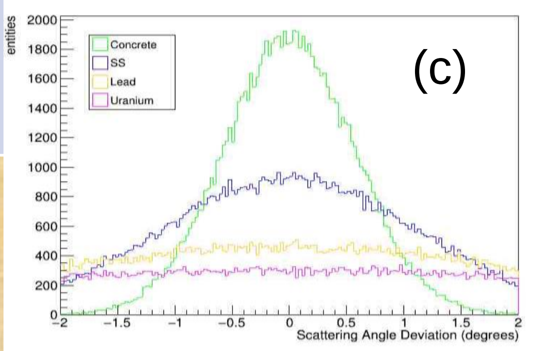
Material thickness = 30 cm

Material	Atomic Mass	Atomic Number	Density (g/cm^3)	Radiation length (cm)	Scattering angle (degree)
Uranium	238	92	19.1	0.308	3.55
Lead	207	82	11.348	0.56	2.31
SS (Fe:Cr:Ni)	Fe = 56, Cr = 52, Ni = 59	Fe = 26, Cr = 24, Ni = 28	8.05, (Fe = 7.86, Cr = 7.2, Ni = 8.907)	1.76	1.15
Air	14	7	0.001256	30558.25	0.0082
Concrete (CaO: Si O_2,: Al_2O_3: Fe_2o_3: H_2O)	H=1, O=16, Fe=56, Al=27, Si=28, Ca=40	H=1. O=8, Fe=26, Al=13, Si=14, Ca=20	2.30, (H=0.07, O=15.1, Fe=7.86, Al=2.71, Si=2.33, Ca=1.55)	6.55	0.571

p = 3000 MeV/c, β = 0.99



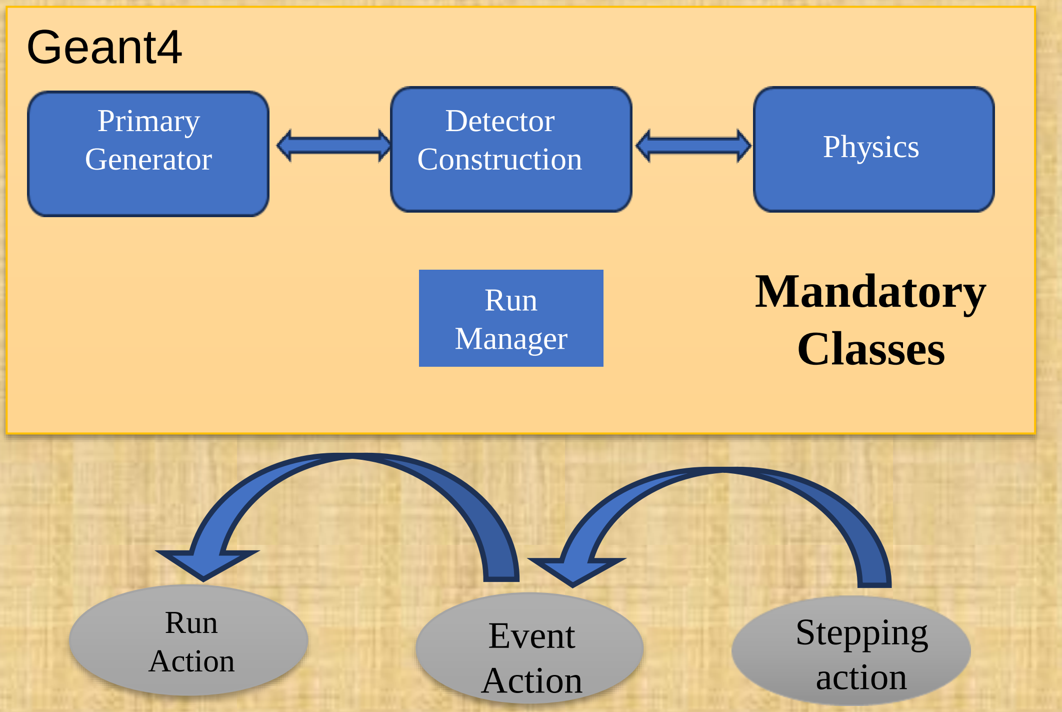


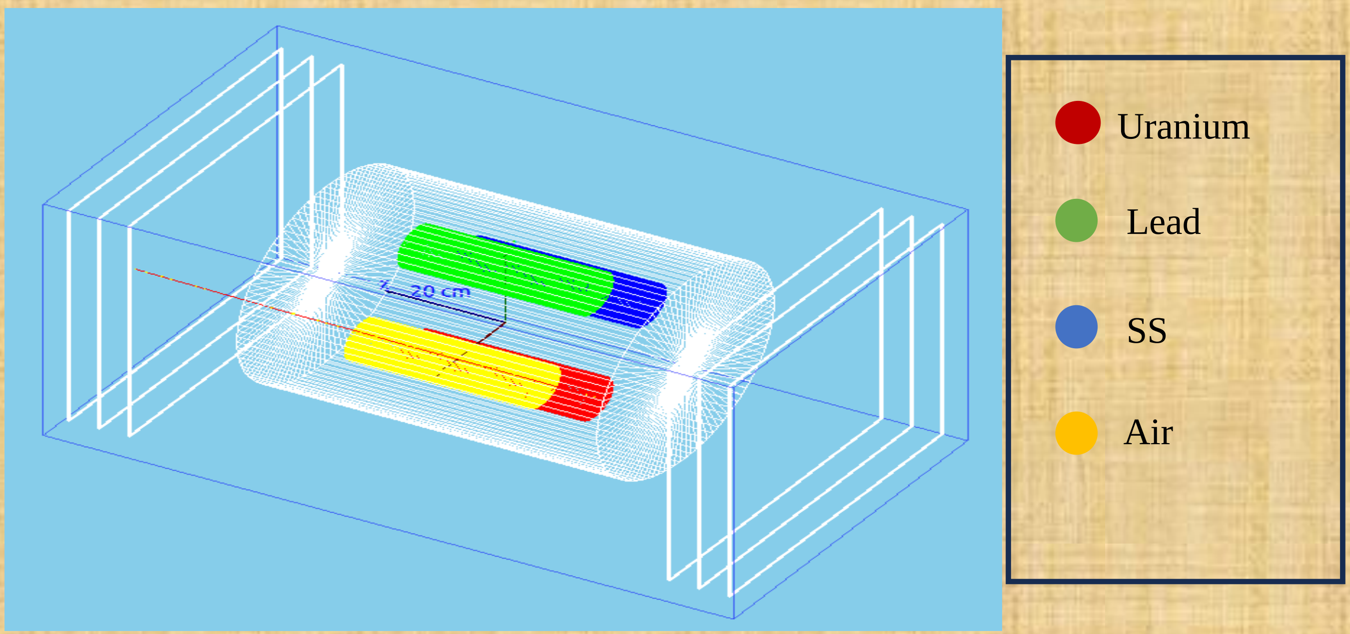


(c) Scattering angle distribution of various materials except air

Conclusion: A linear curve is observed for the scattering angle in the plot (a) for each material with the increase of their thickness. The largest scattering angle deviation is observed for Uranium and an almost scattering-free curve is observed for air. Plot (b) and plot (c) show the distribution of scattering angle deviation w.r.t counts for different materials.

Simulation details





3D view figure of cylindrical nuclear waste with detectors (60cm in size) generated by Geant4 simulation

Event generator: Cosmic Ray Generator (CRY)

- Monte Carlo based package developed by LLNL.
- Generates cosmic muon flux with exact momenta distribution and zenith angle varying from 0 to 90 degrees.
- Provides muon flux at three different altitudes: 0 m, 2100 m, 11300m.
- Energy range: 1-100 GeV.

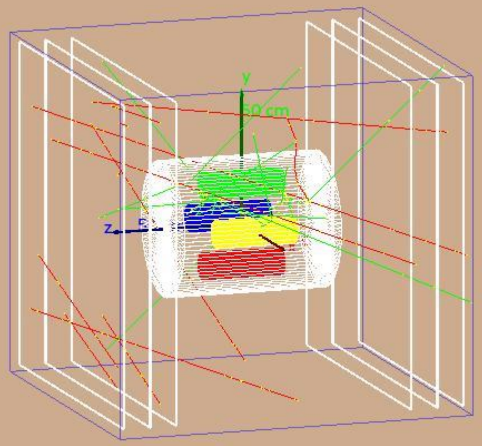
```
returnNeutrons 0
returnProtons 0
returnGammas 0
returnPions 0
returnKaons 0
returnElectrons 0
returnMuons 1
date 02-25-2024
latitude 22.74
altitude 0
subboxLength 1
```

Computational resources:
Desktop with Intel 12th Gen i5
12 CPUs
32 GB RAM
Multithreading with 8 threads used for Geant4.

Configurations / orientations

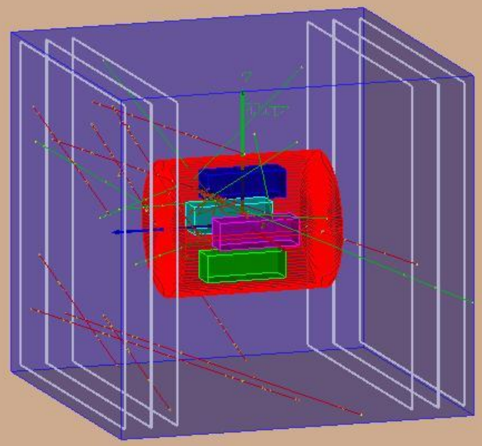
Run 0 (10 events, 10 kept) Sat Oct 5 20:45:43 2024

Circular cylinders within a circular cylinder



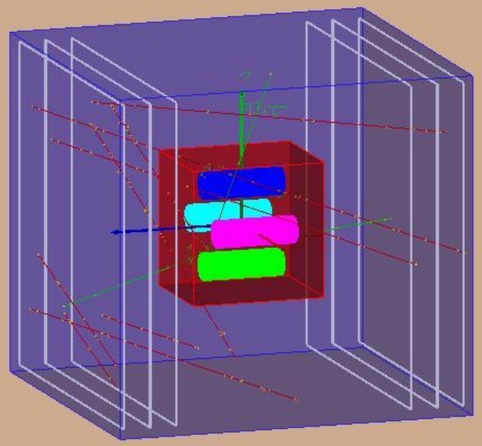
Set 1

Rectangular cylinders within a circular cylinder



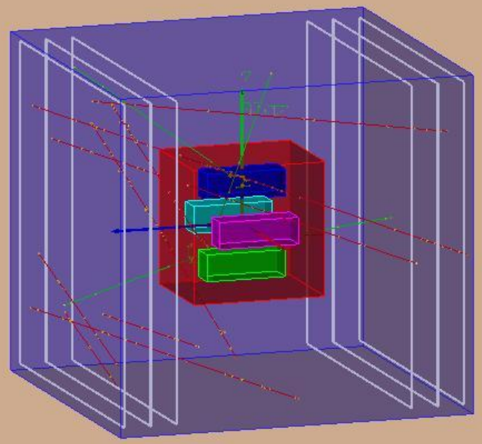
Set 3

Circular cylinders within a rectangular cylinder



Set 5

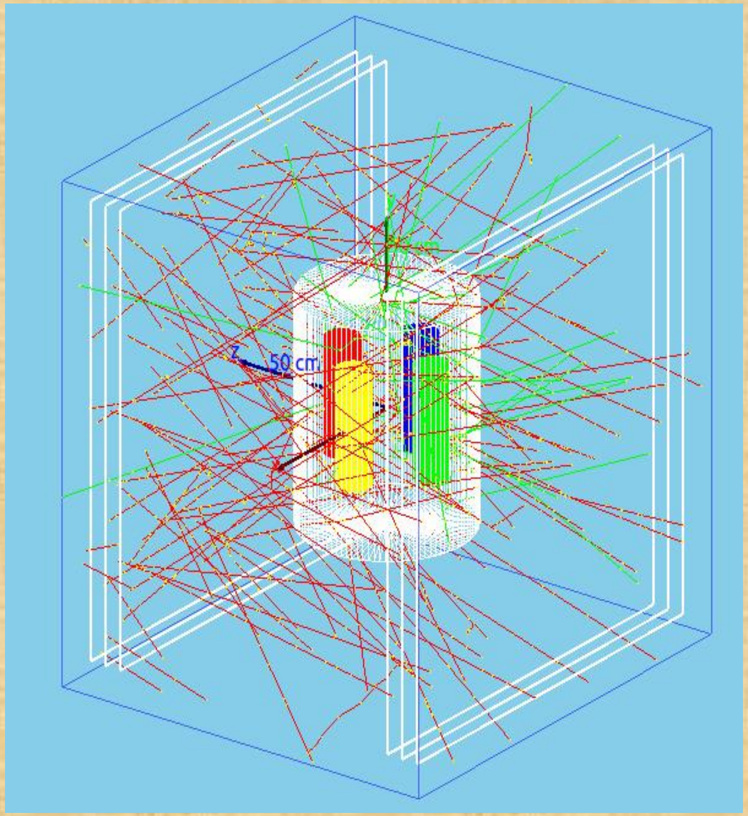
Rectangular cylinders within a rectangular cylinder



Set 6

Geant4 Simulation parameters

- Four storage configurations are considered for this presentation, as shown earlier. On the right, we show few events from the "Set1" configuration (rotated).
- The no. of detectors is 6, and their area needs to be adjusted according to ROI.
- Detector sizes considered: 60 cm, 120 cm and 150 cm.
  - Studies indicate that 120 cm detector size is optimum for the present purpose.
- Detector separations considered: 5 cm, 10 cm.
  - Experimental convenience dictates the use of larger gap. Numerical studies indicate no major problem with a gap of 10 cm.
- Track reconstruction algorithm: PoCA
- 2D image reconstruction.
- Analysis based on cluster density and scattering angle.
- Detector spatial resolution: ideal (0 μm), 100 μm , 500 μm, 1 mm.
- Exposure time: 1 day, 1 week, 1 month.
- Binning in X and Y: 60, 120, 240

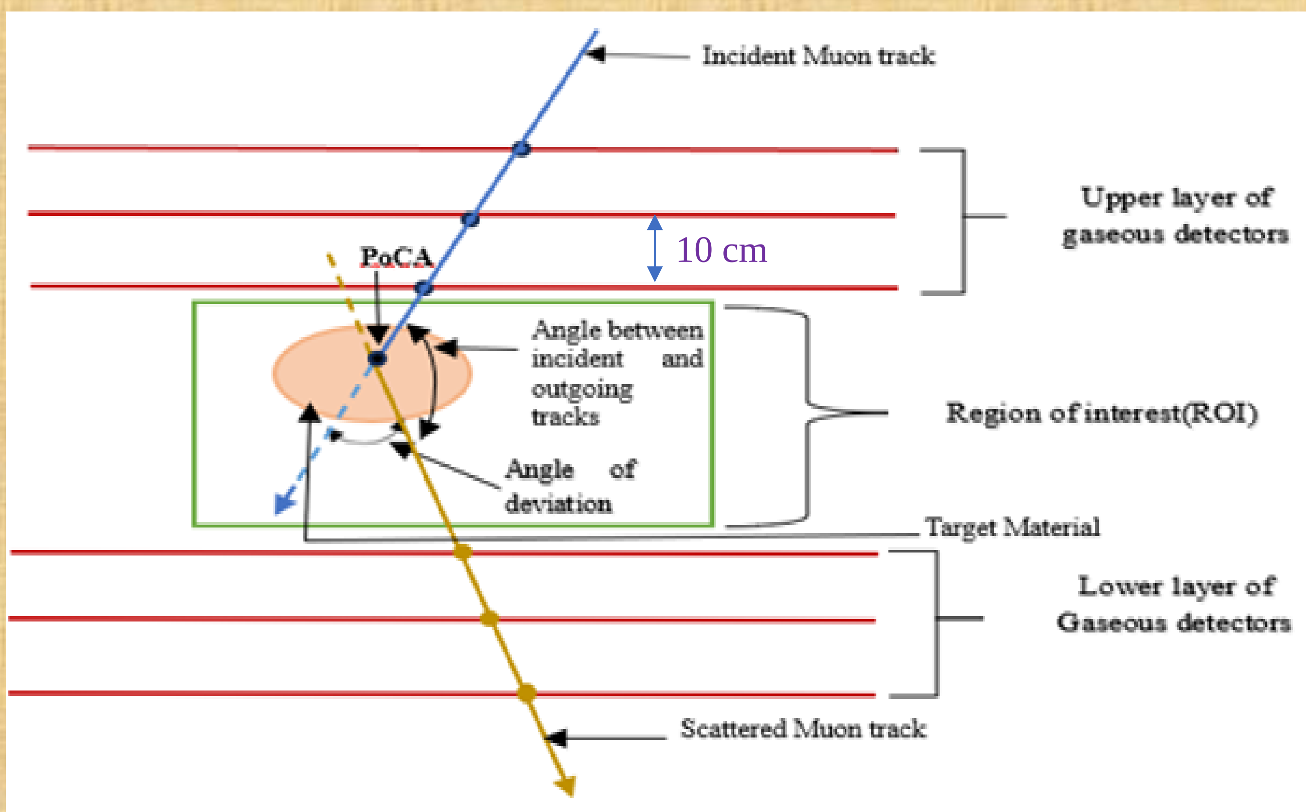


Representative configuration exposed to cosmic ray muons (z points towards the sky)

Reconstruction of muon tracks

Point of Closest Approach(PoCA):

- Identify incoming and outgoing tracks.
- Least square fit method is implemented to obtain the equation of the line passing through three points.
- PoCA is the midpoint of the shortest line joining incoming and outgoing tracks.
- Binned clustering algorithm was also used to identify clusters with RoI. This did not improve the analysis significantly. So, only PoCA was used for further analysis.
- However, PoCA does seem to lead to a large number of scattering vertices outside the RoI. This leads to lack of efficiency and its remedial possibilities are under study.
- Multithreading has been used for PoCA, as well.

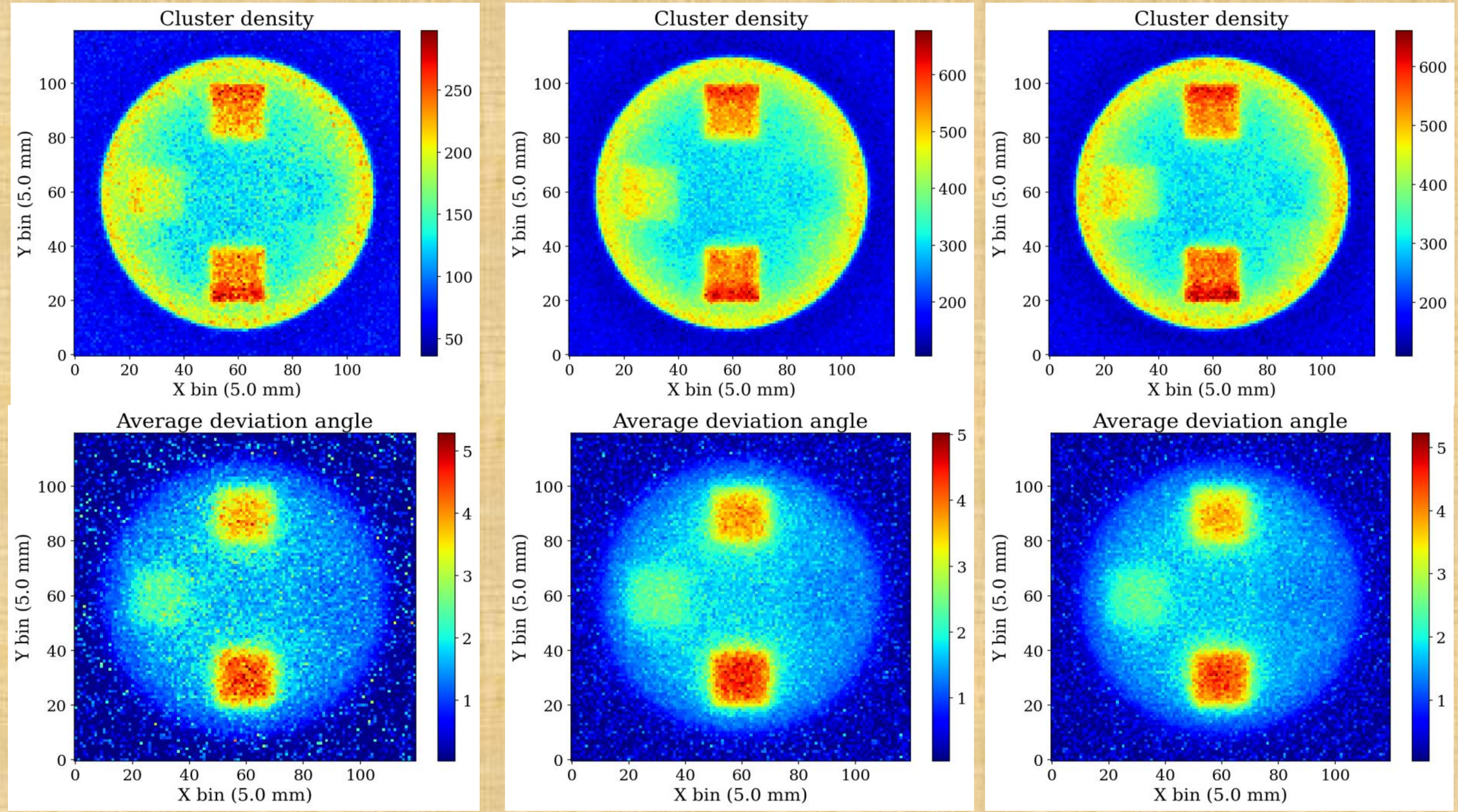


Schematic diagram of simulation setup and PoCA reconstructed point

Selection criteria of an event

- Track hits all 6 detector layers.
- Scattering point of reconstructed tracks lie within ROI.
- Scattering angle between incoming and outgoing track greater than 10 mrad.

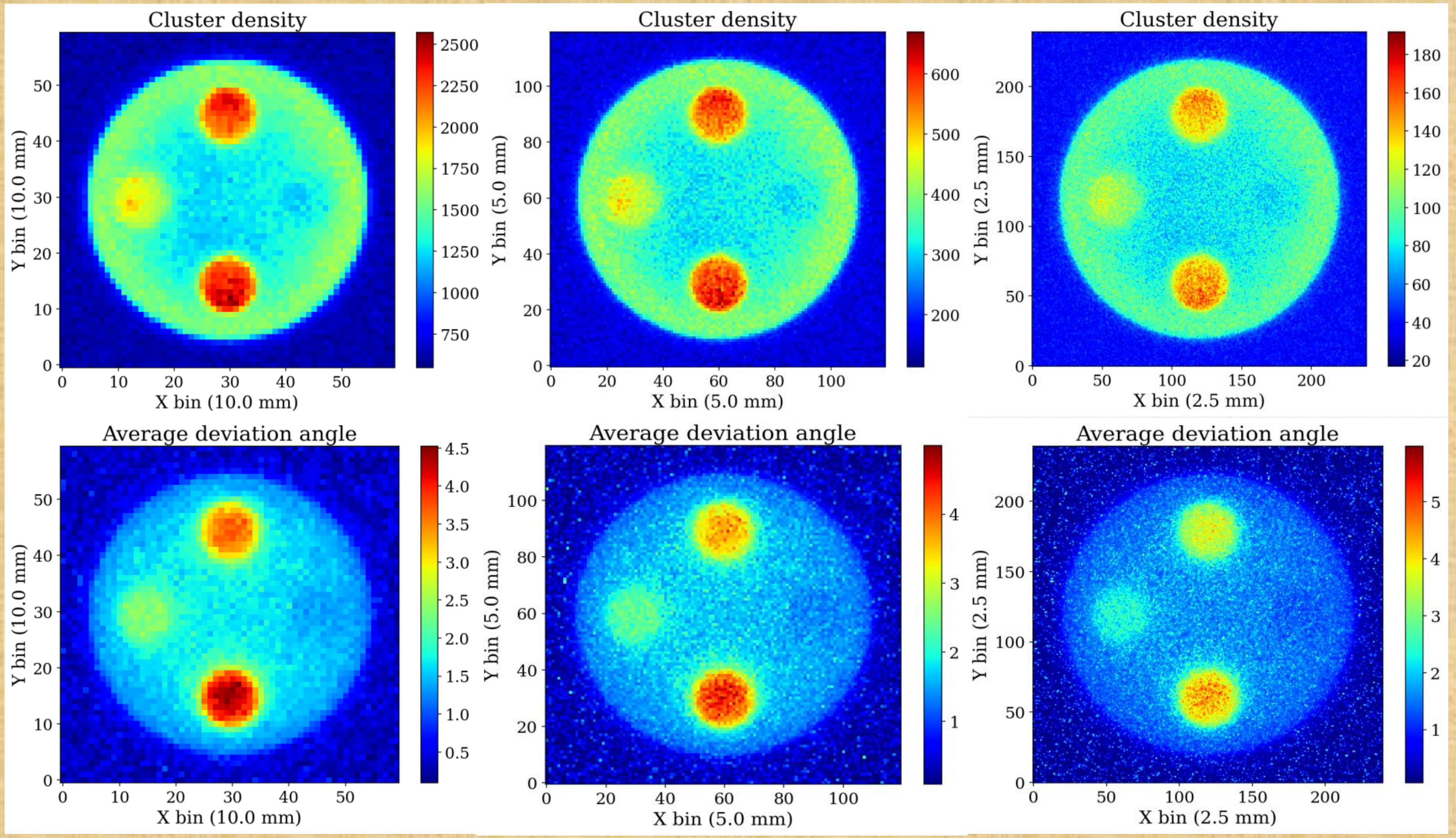
Cluster density and deviation using 120 bins



Set 3

- Top row depicts cluster density.
- Bottom row depicts scattering angle.
- Exposure from left to right: 1 day, 1 week, 1 month.
- As expected, with exposures of longer duration, the images become clean, less noisy and relatively more distinct edges.
- Here onwards, we focus on an exposure of 1 week.

Effect of binning on exposure of 1 week



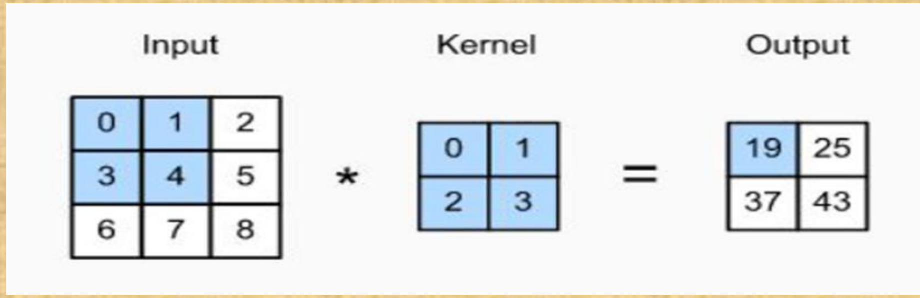
Set 1

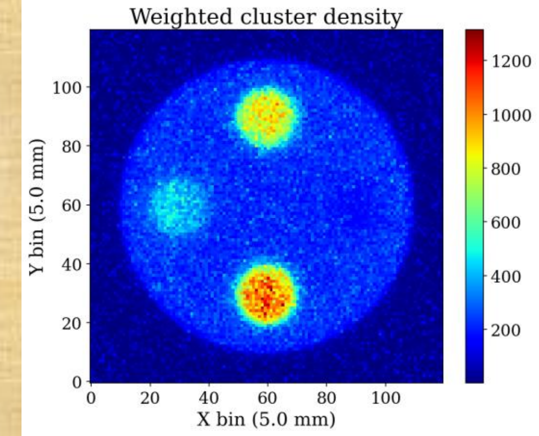
- Top row depicts cluster density.
- Bottom row depicts scattering angle.
- Binning from left to right: 60 (10mm sq), 120 (5mm sq), 240 (2.5mm sq).
- While 60 bins is found to be incapable reproducing curved surfaces properly, both 120 and 240 perform reasonably well.
- Considering the fact that, for a given exposure time, 120 bins allows more samples per bin than 240 bins, we proceed with 120 bins for the rest of the presentation.

Shape Analysis Using Pattern Recognition Method (PRM):

Essentially, convolution of a chosen kernel with the histogram of
- 2D histogram of scattering vertices density.
- 2D weighted histogram of scattering vertices density.

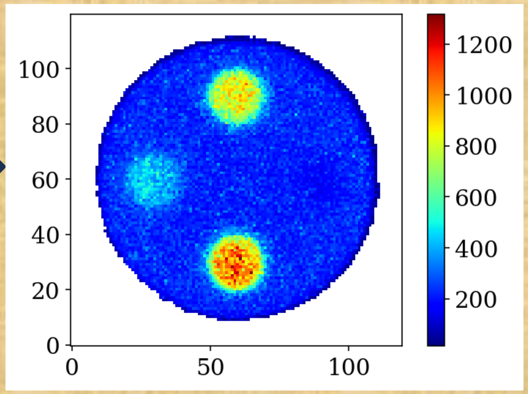
PRM has been successful in removing background noise, detecting edges and identifying clusters.



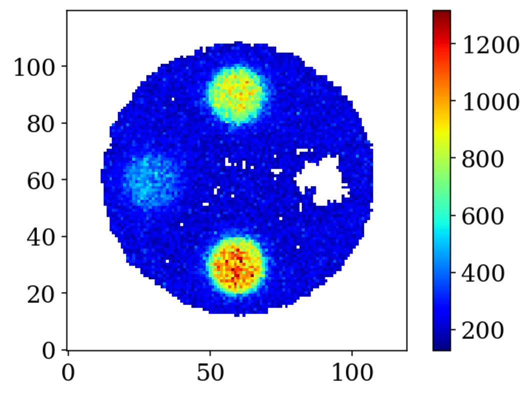


Start with the XY projection of weighted scattering vertices density

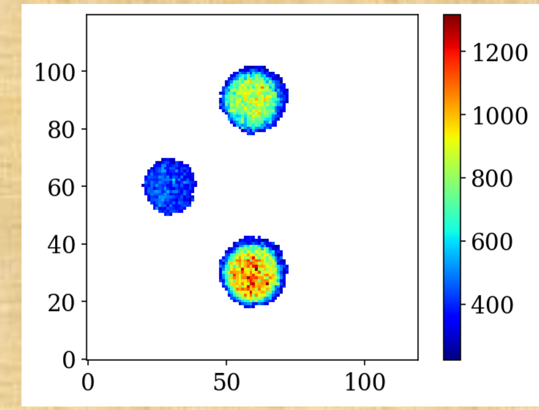
Remove air outside concrete container



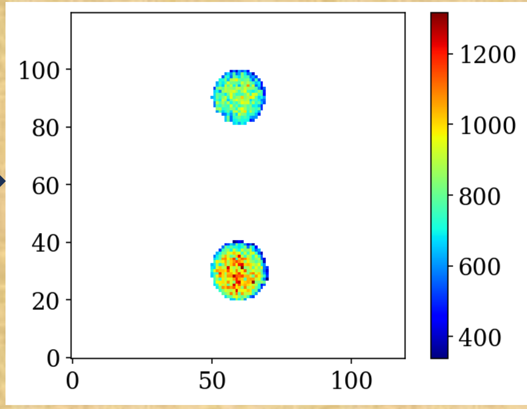
Remove air inside concrete container



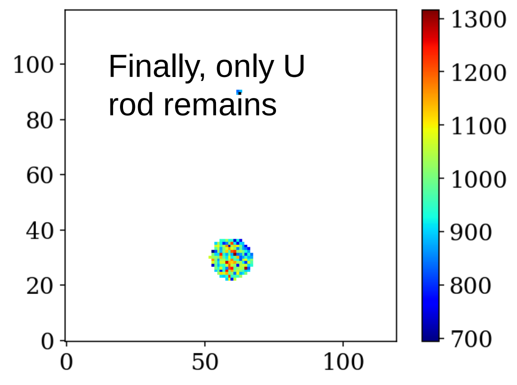
Remove concrete container



Remove SS rod



Remove Pb rod



- Each material has been successfully identified with good edge detection.
- This helps in further processing of the data.

Material discrimination: Supervised ML

- Given a training set of materials, the aim is to classify different materials of a test set.
- Classification based on PoCA points that yields density of scattering points and the amount of scattering within the ROI.
- Classification algorithms used: Linear Regression (LR), K Nearest Neighbour (KNN), Support Vector Machine (SVM), Decision Tree (DT), Random Forest (RF) and Extreme Gradient Boost (XGBoost).
- Hyper-parameter tuning of each algorithm has been attempted.

Accuracy of classification algorithms

Set 3, exposure of 1 month, detector position resolution 0.5mm.

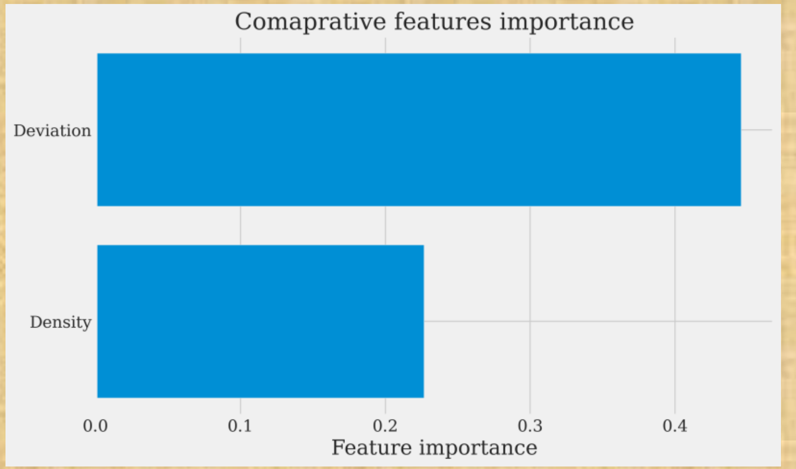
Accuracy	LR	KNN	SVM	DT	RF	XGBoost
Training data	82.90	83.67	83.45	85.72	85.02	82.43
Test data	80.94	82.12	82.59	80.00	81.41	80.70
Details of hyper-parameter tuning						
Tuned parameters	C: 78.46, solver: lbfgs	Metric: Chebyshev, n_neighbors: 25, weights: uniform	C: 100, gamma: 0.5, kernel: rbf	Criterion: gini, max_depth: 6, min_smpls_leaf: 1, min_smpls_split: 3, splitter: best	Bootstrap: True, max_depth: 5, max_features: log2, n_esti: 900, min_smpls_leaf: 4, min_smpls_split: 10,	Booster: gblinear, learning rate: 0.93, n_estimators: 840, objective: binary:logistic

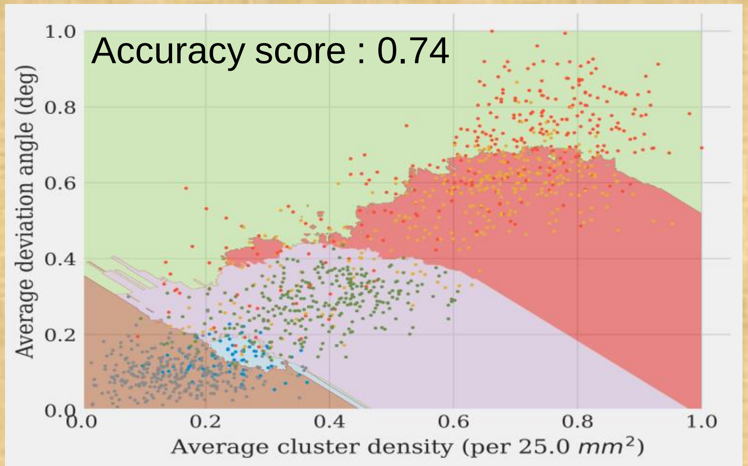
Above table is a representative one, among many configurations, exposures and resolutions.

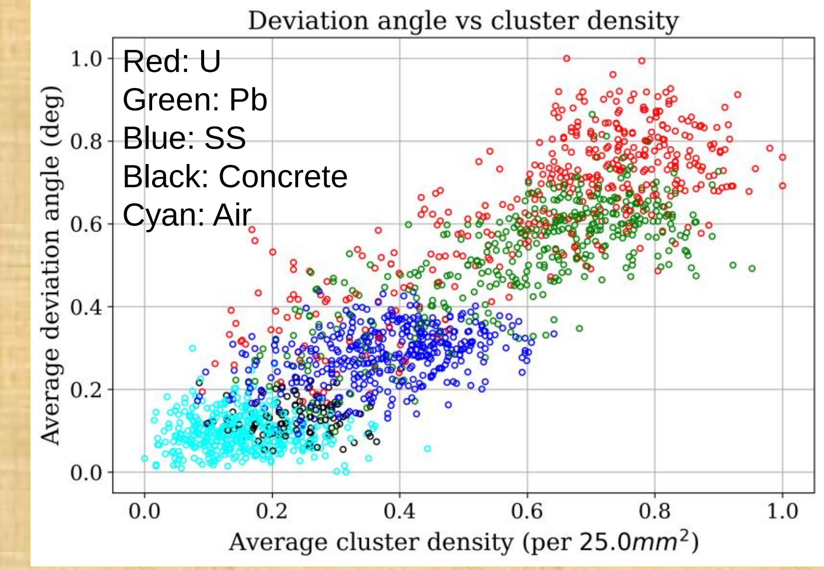
1) KNN, SVM, RF and XGBoost are found to work well under most of the circumstances. LR and DT seem to be relatively weak. RF and XGBoost is more computation intensive.
2) An exposure of a week and position resolution of 0.5mm is found to give satisfactory classification of more than 80%.
3) In an apparently counter-intuitive show, 1mm resolution seem to work fine till an exposure of 1 week. With the extended exposure of 1 month, its classification accuracy drops significantly.

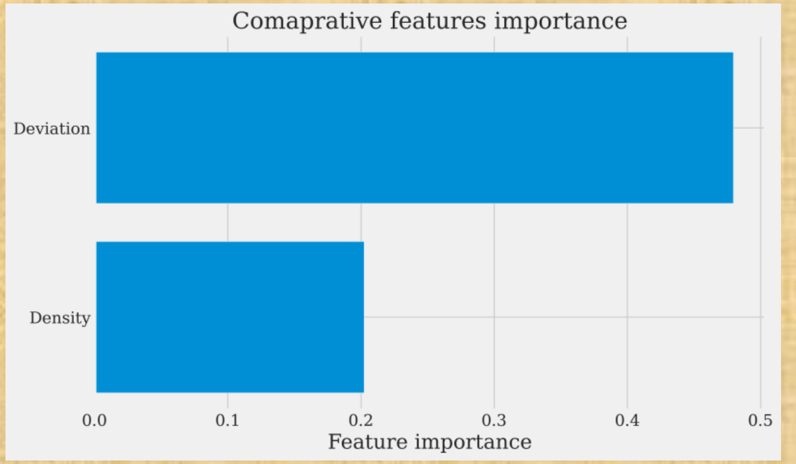
Set1, 1 week data using detectors of 100 μm resolution; Bins 120

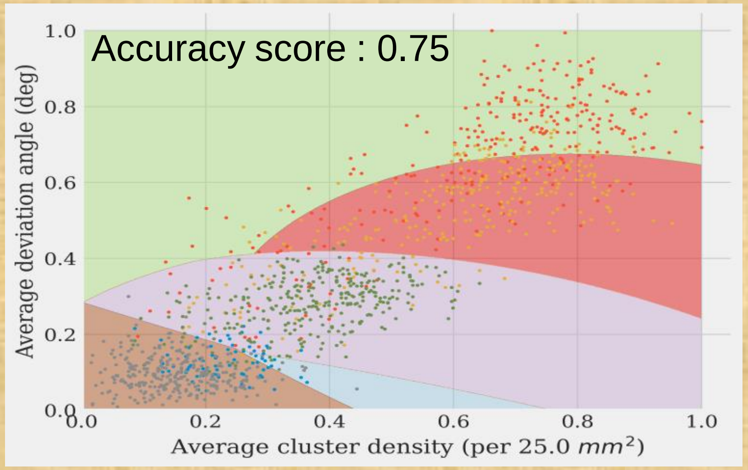
Typical KNN results







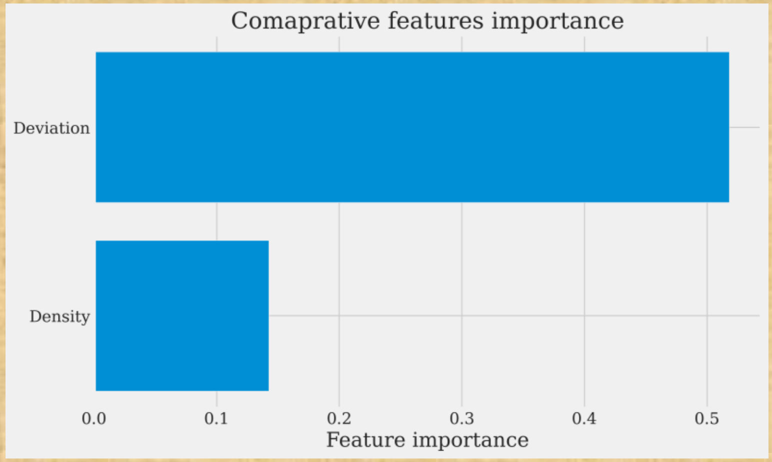


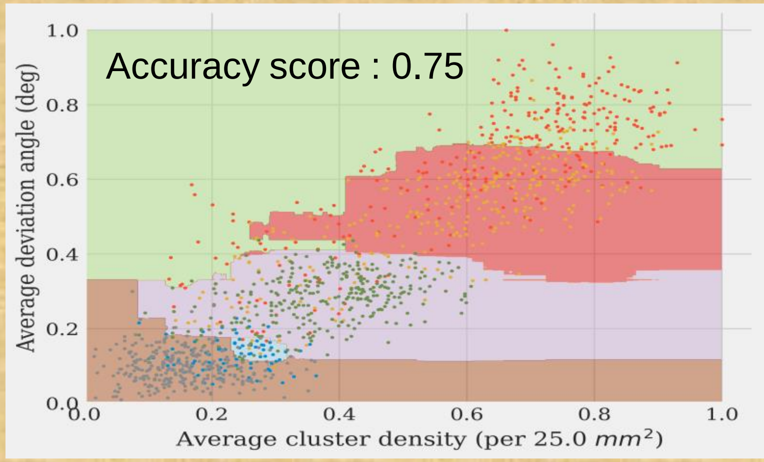


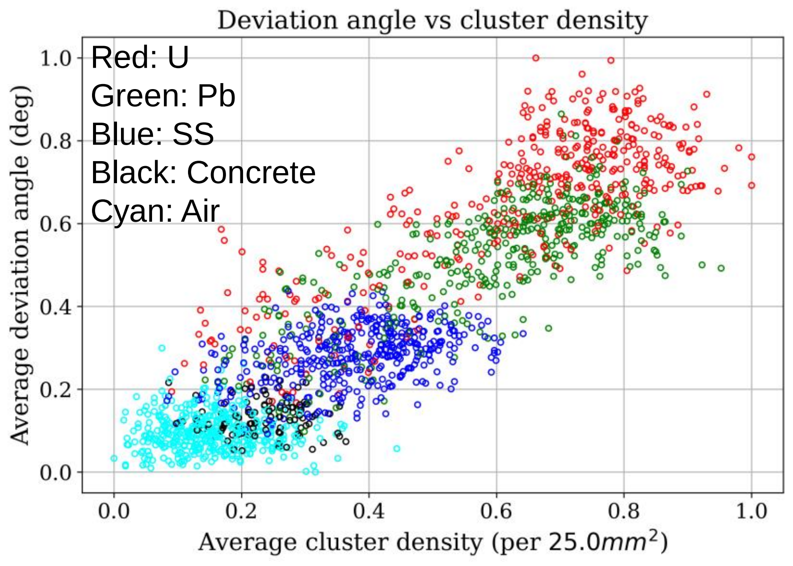
Typical SVM results

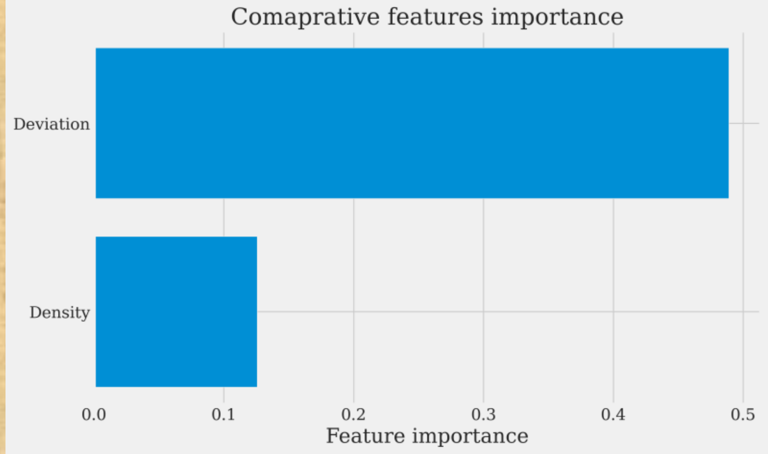
Set1, 1 week data using detectors of 100 µm resolution; Bins 120

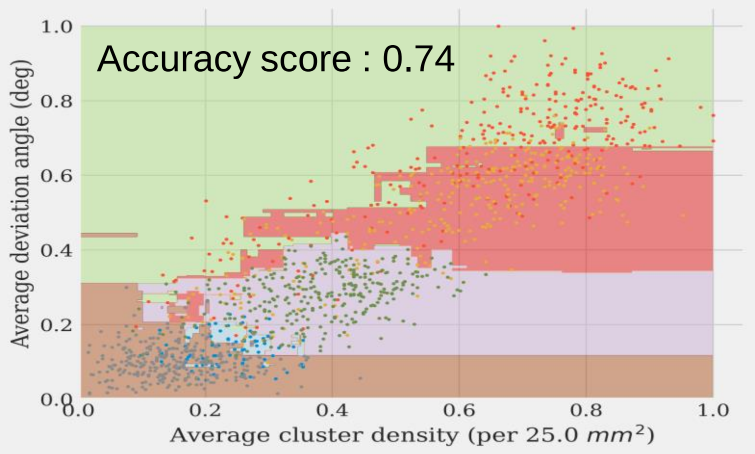
Typical RF results







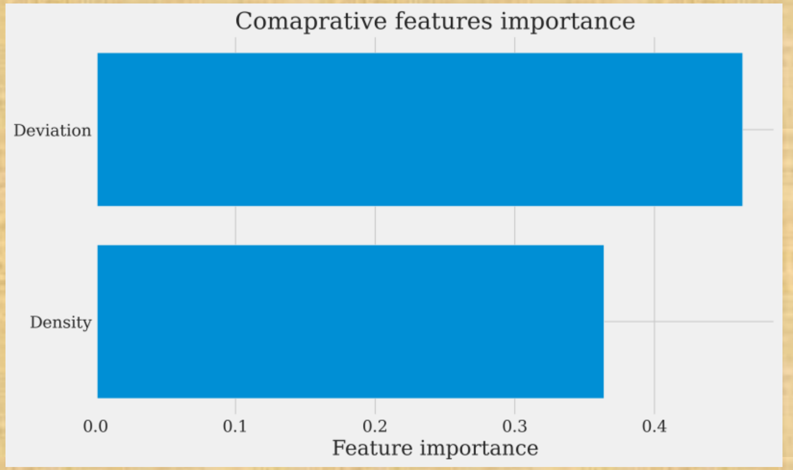


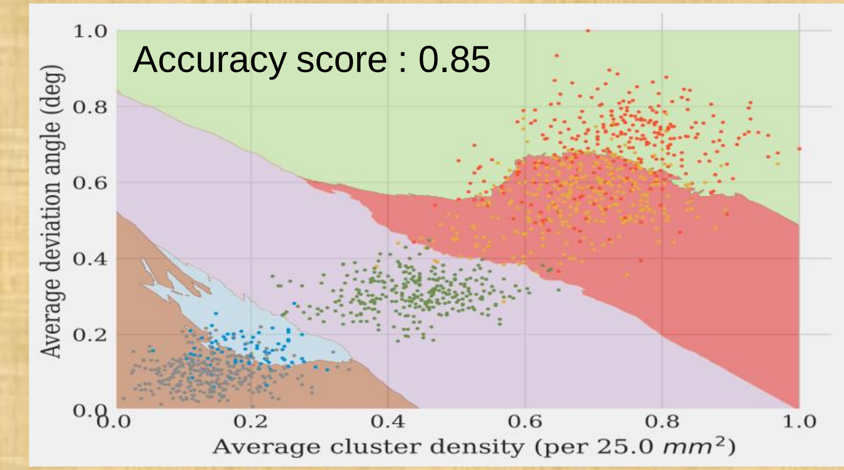


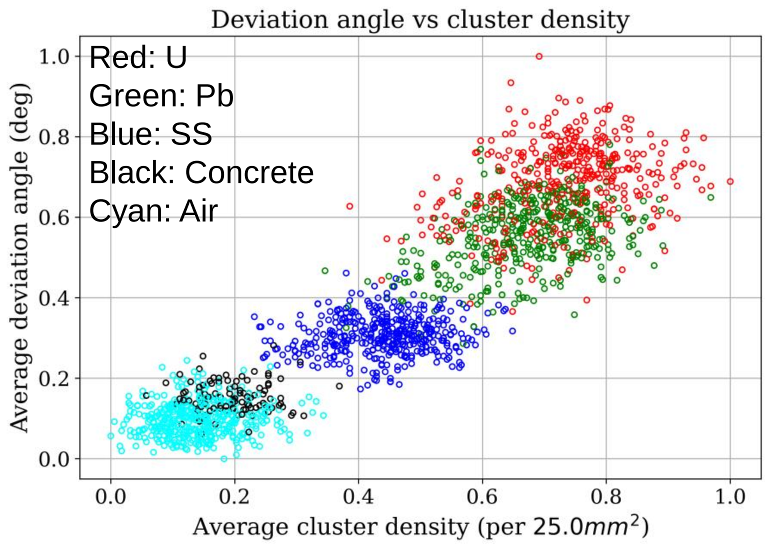
Typical XGB results

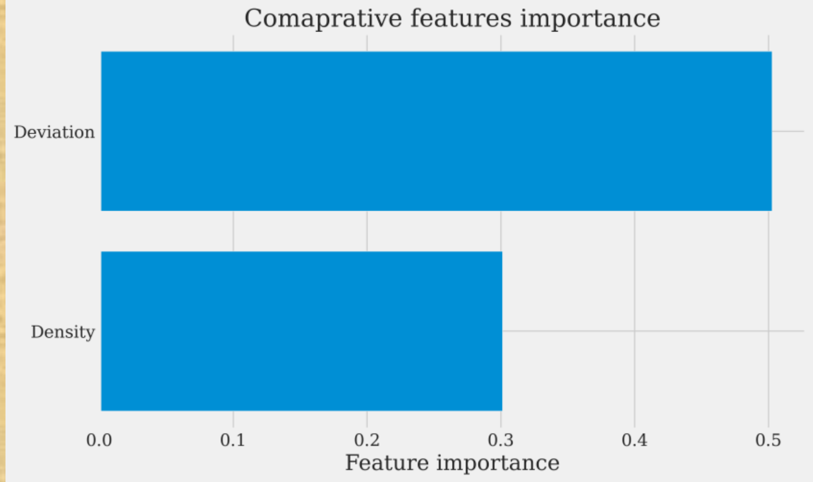
Set6, 1 week data using detectors of 100 μm resolution; Bins 120

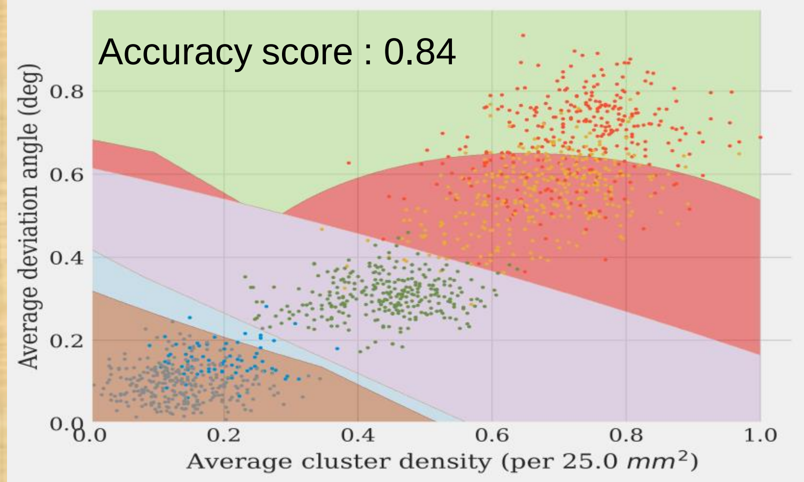
Typical KNN results







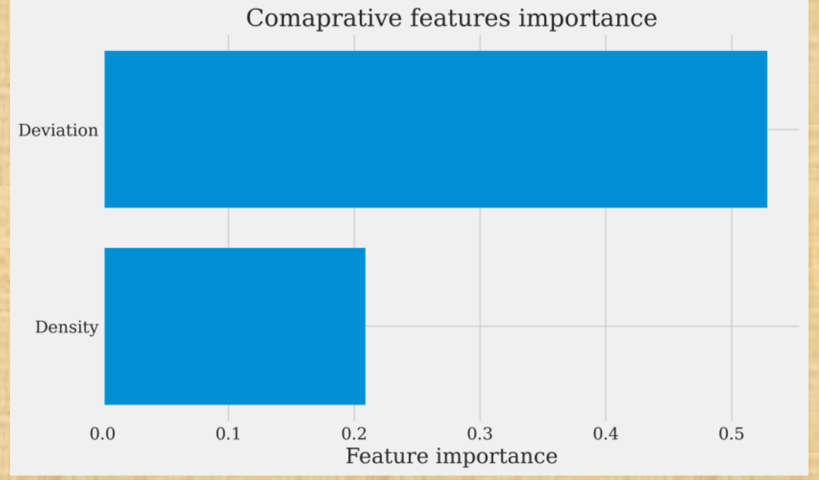


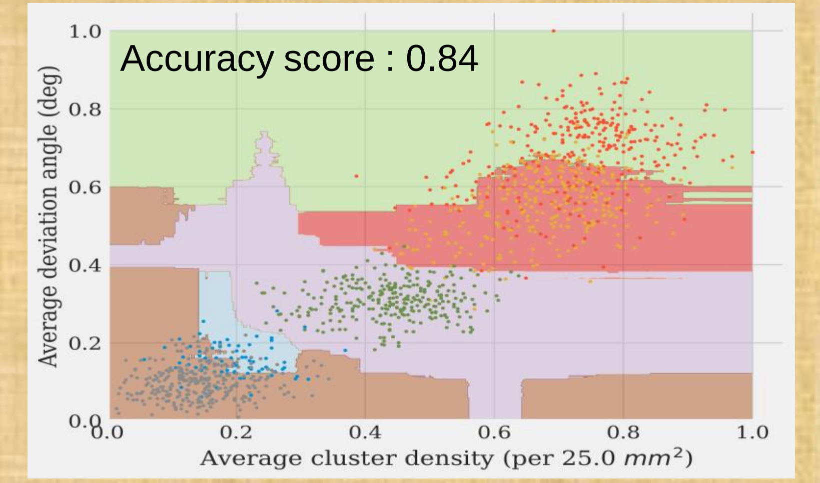


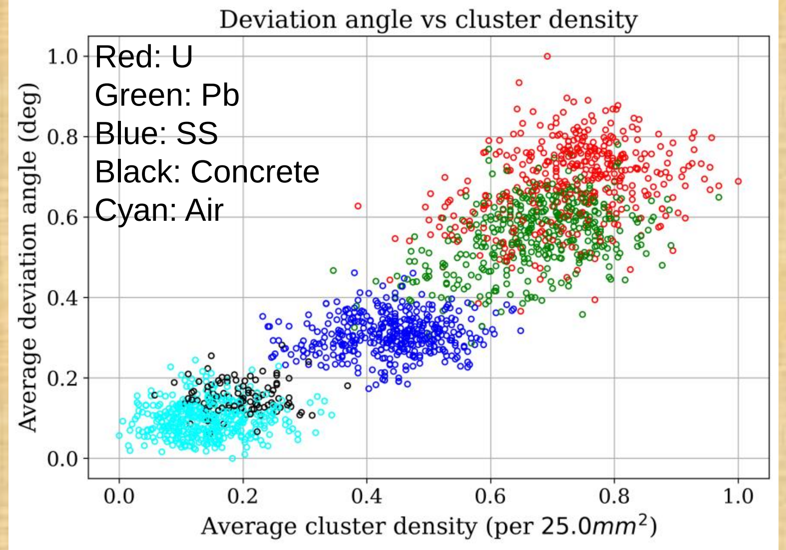
Typical SVM results

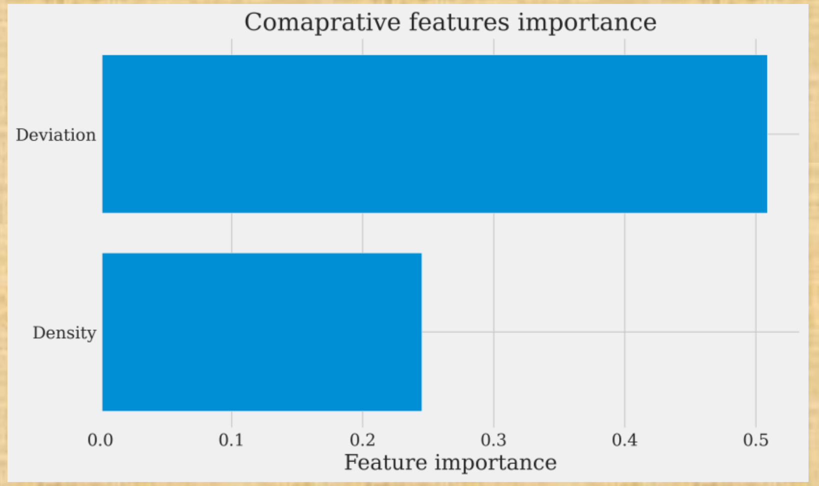
Set6, 1 week data using detectors of 100 μm resolution; Bins 120

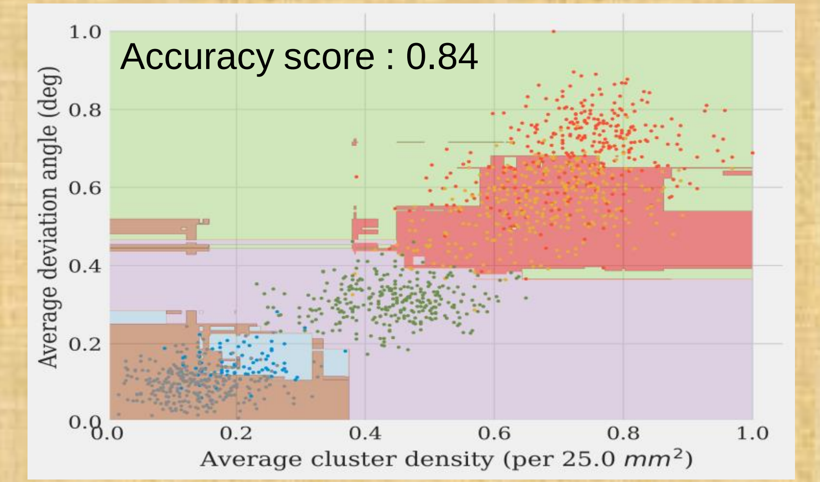
Typical RF results











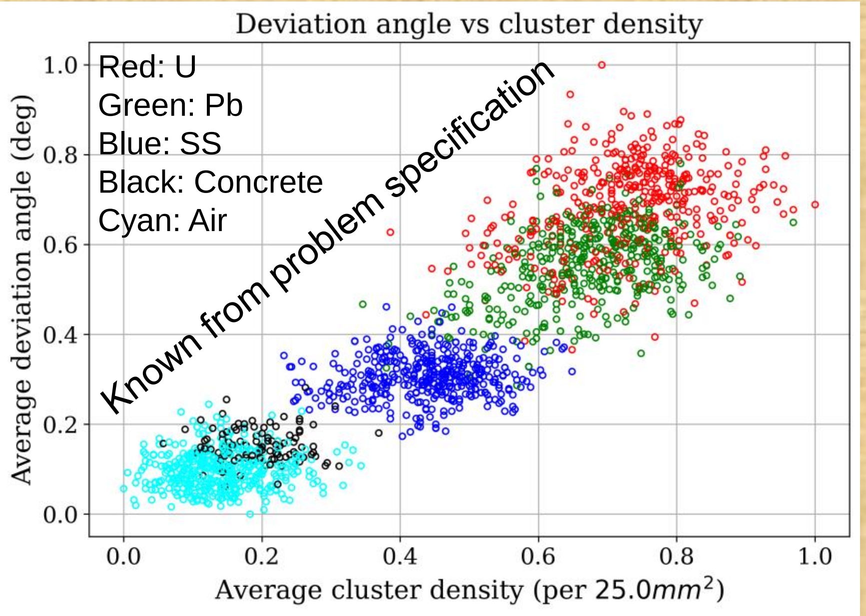
Typical XGB results

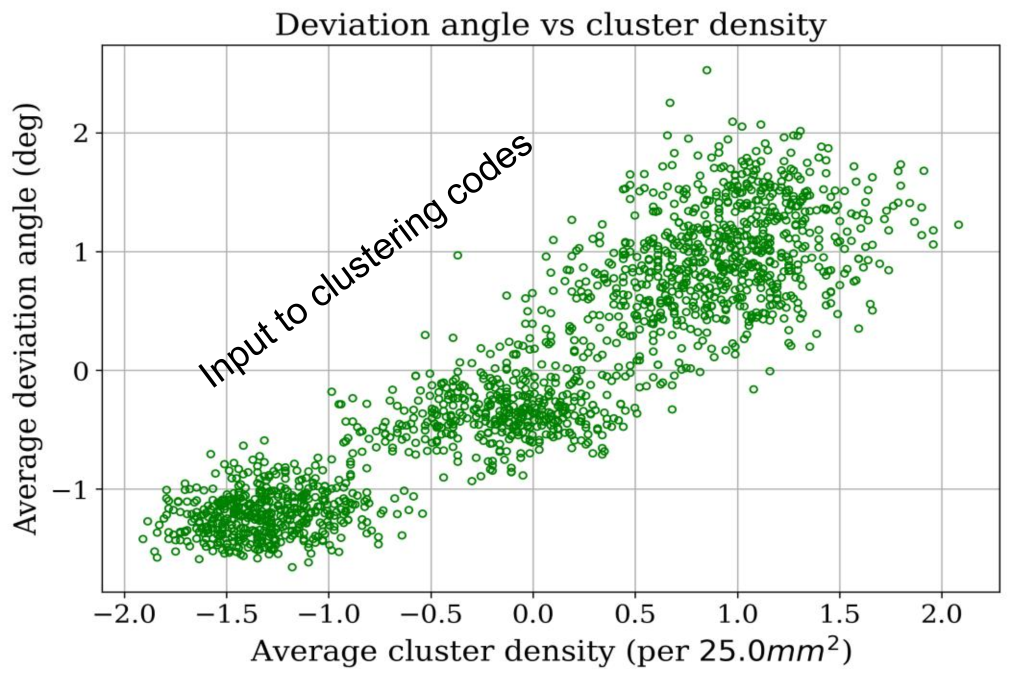
Comparison of storage configurations

- Set 1 has all circular cylinders. This is difficult to be represented by rectangular binning. This leads to relatively poor prediction accuracy – around 75% for an exposure of one week.
- Since the internal cylinders are smaller in size, making them rectangular (set 3) immediately improves the prediction accuracy to close to 80%.
- With set 6, where all cylinders are rectangular, model accuracy is 85% on an average.
- It is expected that monitoring will be easier stored materials are rectangular in shape.
- Arbitrary shapes can be handled with body-conforming binning mesh, but this will lead to significant complications

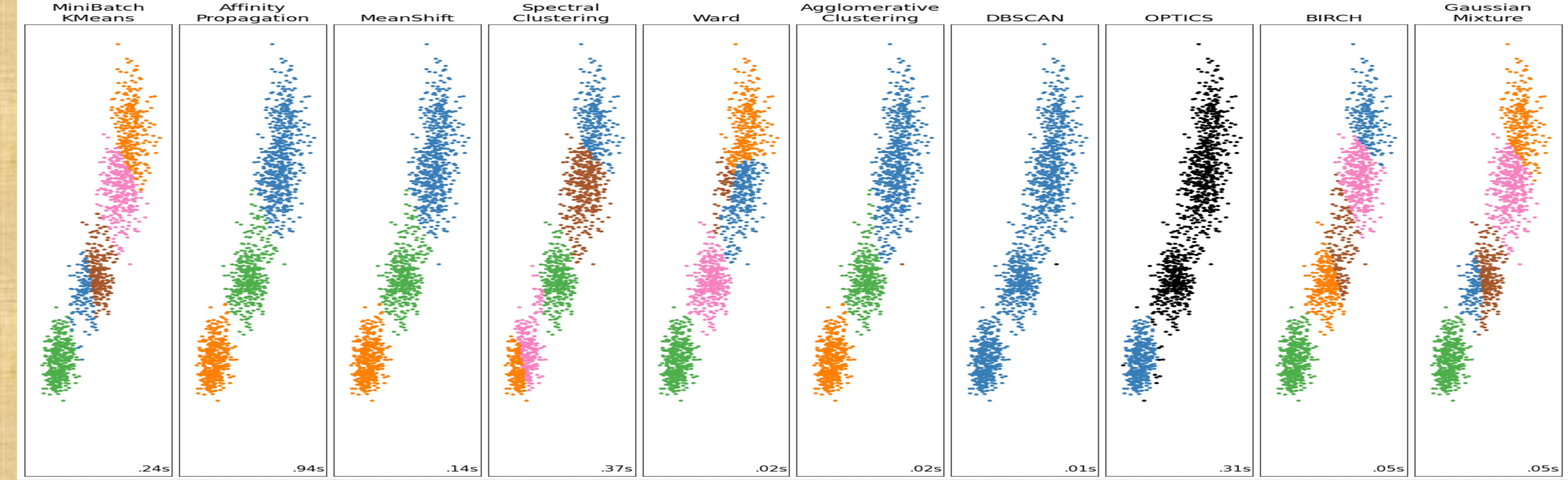
Clustering of unknown materials: Unsupervised ML

- The challenge is to find clusters of different materials.
- The features, cluster density and scattering angle, are extracted from the PoCA points, as before.
- Clustering algorithms used: Minibatch-KMeans, Affinity Propagation, Mean Shift, Spectral Clustering, Ward, Agglomerative Clustering, DBSCAN, OPTICS, BIRCH, Gaussian Mixture.
- Input PoCA points are from set 6 for a detector position resolution 100μm. Geant4 simulation carried out for a data acquisition time of 1 week.





Results for clustering algorithms



- Five clusters are clearly identified by the following algorithms:
  1) MiniBatch KMeans, 2) Spectral clustering, 3) Ward, 4) BIRCH and 5) Gaussian Mixture.
- Among these, Ward yields unreliable clusters, when compared with known input clusters.
- BIRCH and Gaussian Mixture methods are also computationally very efficient.

Conclusion

1. Cosmic ray muon imaging can be a reliable technique to monitor nuclear waste.
2. The PoCA algorithm is found to work well. However, there are scopes of improvement.
3. While the technique allows continuous monitoring, an exposure of 1 week is found to be sufficient to discriminate materials relevant for nuclear waste management.
4. Detector resolution of around 0.5 mm is sufficient to carry out the job.
5. Binning of 120 (for the geometry considered here) proves to be sufficient.
6. The PRM operations help the subsequent analysis procedures by removing noise, identifying shapes and thresholds corresponding to different materials.
7. Geometry of nuclear wate storage can play an important part in its subsequent monitoring. According to the present study, set 6, comprising of all rectangular shapes, is found to be more promising.
8. Few reliable classification techniques such as KNN, SVM, RF and XGBoost have been identified using which materials relevant for nuclear waste monitoring can be discriminated with more than 80% accuracy.
9. Few successful clustering techniques have also been identified, such as MiniBatch-Kmeans, Spectral Clustering, Birch, and Gaussian Mixtures. As a result, it is possible to identify the clusters in an unsupervised manner.

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