Improvement of Quantification in Non-Destructive Characterization of Radioactive Waste Packages – 22176

Stefan Rummel *, Thomas Buecherl *, Christoph Lierse von Gostomski *
* TU Muenchen, ZTWB Radiochemie Muenchen RCM

ABSTRACT

For the characterization of radioactive waste packages, like 200l barrels, preferably non-destructive methods like segmented gamma scanning (SGS), transmission measurement or computer tomography are used. Segmented gamma scanning allows to identify gamma-emitting isotopes and their distribution in the vessel. To determine the corresponding activities additional information on the absorption of the waste matrix is needed. In addition, supplemental information on the waste like its chemical composition, materials and nuclide vectors are frequently available.

A unified method allowing to combine data from measurements and a-priory knowledge on each waste package giving a precise result as well as robust error estimates is currently not available. A method allowing to achieve this task is Bayesian statistics.

The application of the Bayesian approach requires to solve integrals (marginalize), whose dimensions are determined by the number of parameters. Depending on the size of the problem, various methods to accomplish this are described in literature. Common to all of them is the need of a model to predict the outcome of the measurement given the parameters of interest like the radioactive inventory. In case of large number of parameters, the Marcov-Chain-Monte-Carlo (MCMC) algorithm is the method of choice.

The talk will focus on the modelling of the SGS measurement process using information of the matrix derived from computer tomography. In a first step the CT-data is transferred into a 3d-voxel model. The loss of photons due to absorption and scattering on their path to the detector is calculated using an adapted Box-Intersect algorithm to get the individual contribution of each voxel. In addition, the solid angle and attenuation due to the collimator are incorporated using geometric considerations. The approach allows to assign a weight to each voxel allowing to predict the photon count rate given the activity in the voxel.

The validity of the procedure has been tested using various vessels with well-known matrix and calibration standards. The results of these comparisons will be presented.

The model predicts the count rate in each segment of the scan given the radioactive inventory in each voxel, one of the key elements to run the Marcov-Chain-Monte-Carlo algorithm. Besides the model connecting the observed quantities with the parameters of interest. Further considerations are necessary to model the activity. In a straightforward manner each voxel is allowed to carry an activity determined by prior knowledge. In a previous paper we demonstrated that this assumption delivers good performance for a subset of problems. However once the measurement don’t constrain the spatial distribution of the radioactive inventory properly convergence problems arise. To mitigate them the activity distribution is modeled using a compositional distribution consisting of point sources and various forms of homogeneous sources. This effectively reduces the dimensionality of the posterior distribution and improves the convergence of the algorithm.

INTRODUCTION

To evaluate the radioactive inventory of waste packages, preferably nondestructive methods are used. Segmented Gamma Scanning (SGS) is an established method to investigate the inventory of gamma emitting isotopes.

The primary motivation for this project is the optimal utilization of existing SGS measurement setups at nuclear facilities focusing on the analysis of those measurement data. As an intermediate step we assume that the matrix of the waste barrel is known e.g. a computed tomography has been performed. As a second step the information on the matrix will be derived using a simultaneous transmission source in combination with the SGS-setup.

In this publication, we focus on the hypothesis required on the activity distribution to analyses SGS measurement data within the framework presented in [Bücherl et al. 2021].

To analyze the data generated by SGS-measurements a number of assumptions on both the matrix and the distribution of the radioactive inventory are made. In its most simple from both activity and matrix are assumed to be homogeneously distributed. However most waste packages exhibit significant inhomogeneity's in both activity and matrix density leading to significant systematic deviation from the true value. In addition, most algorithm developed for this task result in a single best estimate of the activity given the measurement. Depending on the algorithm employed, a classical error propagation can be used to determine the uncertainty of the result, in case of complex optimization algorithms an estimate of uncertainty intervals might be even impossible.

The project described in this paper therefore tries to tackle some of the issues mentioned. In particular, we focus on the determination of uncertainty intervals allowing simple judgement of the results, incorporation of additional hypothesis on activity distributions, as well as modelling arbitrary matrices using data for computed tomography measurements.

A framework to combine various measurements, a-priory knowledge as well as statistical fluctuations due to counting processes is the Bayes-statistics. The approach is actively used in several projects, exemplarily [Carasco 2021] and [Laloy et al. 2021] can be mentioned. Both utilize Bayesian statistics, but differ in the modelling the matrix and assumptions on source distributions.

The mathematical details as well as first examples of the method described in this paper can be found in [Bücherl et al. 2021]. In this publication we give a short summary on the modelling of the measurement process, the methods involved to reconstruct the activity and introduce additional hypothesis on activity distributions and mixture models of those.

THE BAYES STATISTICS

The roots of the Bayes approach can be traced back to the Bayes-theorem formulated by Thomas Bayes in 1763. In the following equation the probability distribution of the activity A given the observation (here measurement Y^{obs} and an additional parameter env is described by:

$$p\left(A\middle|Y^{obs}, env\right) = \frac{p\left(Y^{obs}\middle|A, env\right)\cdot p(A|env)\cdot p(env)}{\int p\left(Y^{obs}\right)\, dY^{obs}} \qquad \text{(Equ. 1)}$$

The formula expands the basic Bayes-theorem by the parameter “evn” which can be used to specify additional a-priori knowledge, e.g. the chemical composition, of the waste package. Using this, the activity given the measurement data can be determined by predicting the probability of the measured data given the activity and the “env” variable $p(Y^{obs}|A,env)$. To tackle the first term established simulation methods and analytic approaches exist. Codes like MCNP, GEANT or Penelope can be used, alternatively the attenuation in matter can be analytically described using Beer-Lambert-law with linear attenuation coefficients form various available databases [Knoll 2010].

The last challenge to solve equation 1 is the denominator, which serves as normalization for the posterior distribution. Calculating the factor is hardly possible in most cases, therefore alternative methods are required. An elegant way to overcome this challenge is the Marcov-Chain-Montecarlo (MCMC) technique. It allows to construct a chain of random numbers distributed according to the posterior without the need of normalization. Since the MCMC algorithms use a quotient of the distribution the calculation of the cumbersome denominator is not necessary at all [Robert and Casella 2004].

The actual implementation of the MCMC algorithms is noticeable eased since high level implementations in various programming languages are available. The most prominent examples are Greta in R, STAN in C++ as well as PyMC3 in Python. In this project, the latter is used.

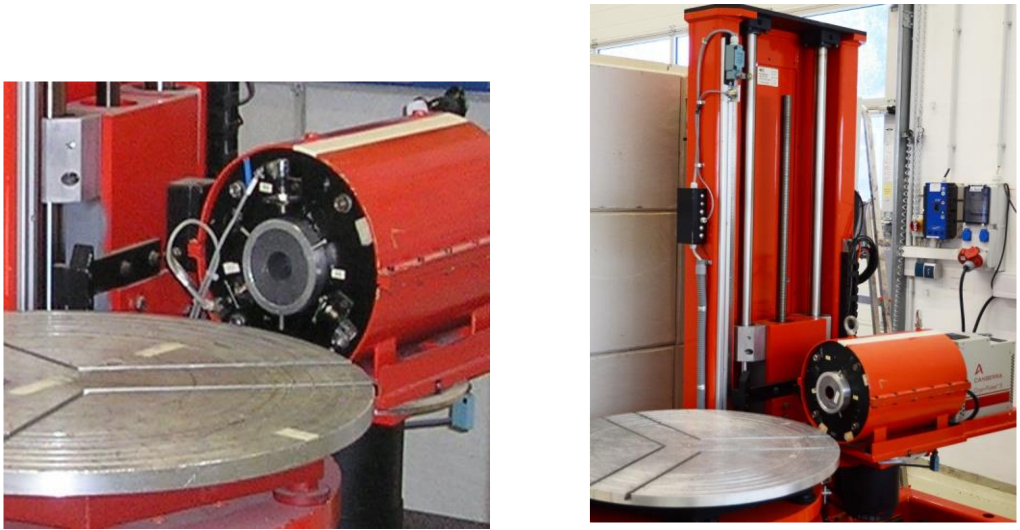

Figure 1 40mm circular lead collimator (left), SGS setup (right)

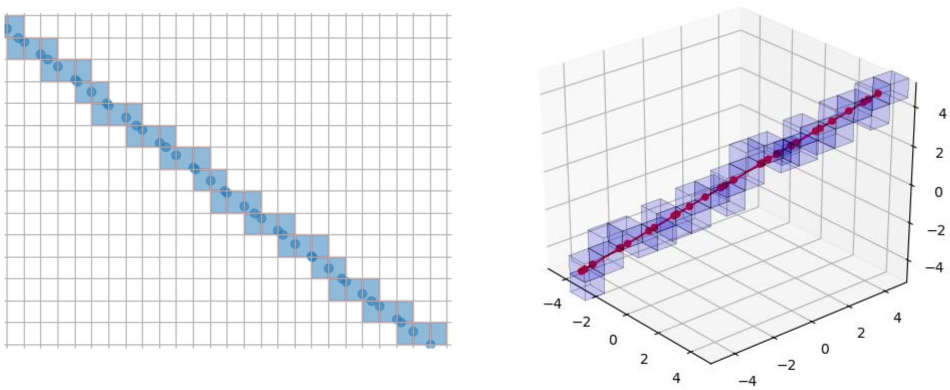

Figure 2: Voxel with track and intersections on the left 2d, right 3d

MODELLING THE MEASUREMENT PROCESS

As pointed out in the previous paragraph the modelling of the measurement process is the key for the Bayes-methodology. As the goal of this project is to develop a time efficient, real world algorithm the modelling of the measurement process uses the Beer-Lambert-law with linear attenuation derived from measurements or databases (e.g. XCOM from NIST). To describe the waste package it is subdivided in even sized "voxel" each voxel carries the information on the attenuation coefficient. The attenuation of radiation along a line is calculated by summing up the individual attenuation contributions and track length $\mu_{lmn,E} * L_{lmn}$ in each voxel. Figure 2 shows two examples of a track in the voxel model in 2d and 3d. To obtain the actual number of detected photons the detector- and collimator response, integration time and solid angle is added as factor. The response of the detector is modeled using an empirical obtained Full Energy Peak Efficiency i.e. the number of photons detected in the full energy peak over the photons impinging the detector. The solid angle and collimator function have been numerically evaluated using geometric model based on "constructive solid geometry". The implementation uses the ROOT geometry package. As indicated in formula 2 the collimator function is parametrized in energy and two space coordinates. Using this considerations the signal strength, i.e. the expectation value of the underlying Poisson distribution, in segment s from an activity in voxel ijk can be expressed as:

$$N_{ijk,s,E} = A_{ijk,E} * W_{ijk,s,E} = A_{ijk,E} * T * \Omega_{det} * \mathrm{Col}(x_{det,s}, x_{ijk}, E) * \varepsilon_E * e^{-\sum \mu_{lmn,E} * L_{lmn,s}} \qquad \text{(Equ. 2)}$$

Where A is the activity in voxel ijk, T the integration time, ε_E the full energy peak efficiency, the exponential function contains the absorption in the matrix. The factors in equation 2 can be interpreted as sensitivity or weight factor of an individual voxel. Figure 3 shows a plot of weight matrices for two angles of the SGS scan with the detector in the upper right corner (left) and on the right side (right) plot. The weight contains all information on the measurement process. Using this the total counts in each segment of the SGS measurement can be expressed as:

$$N_{s,E} = \sum_{klm} A_{klm} * W_{s,klm,E}\ with\ A_{klm} \ \text{the activity in each voxel} \quad \text{(Equ. 3)}$$

Equation 3 clearly shows the linear character of the problem, moreover the formulations allows a simple variation of activity configurations ranging from "point like" i.e one pixel carries activity to more complex ones.

To evaluate measurements with continuous scanning i.e. the barrel under investigation rotates permanently each segment is subdivided into a number of segment with finer angular resolution. Those segments are averaged to incorporate the effect of continuous rotation. For a typical measurement with 24 segments each is subdivided in 15 sub segments.

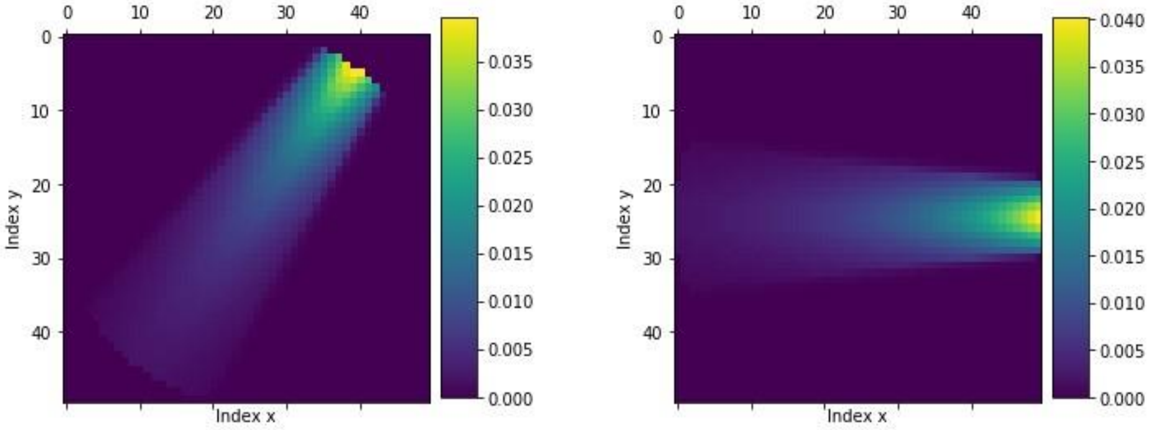

Figure 3: Weight matrices of a homogeneous matrix

Figure 3 shows two examples of a weight matrix of a homogeneous waste barrel.

VALIDATION OF THE MODEL

A number of validation measurements of the model have been performed using the SGS setup. Figure 4 shows the comparison between the measured collimator function with the simulation at a distance of 1m between detector and source.

A validation measurement using a source measurement can be found in [Bücherl et al. 2021].

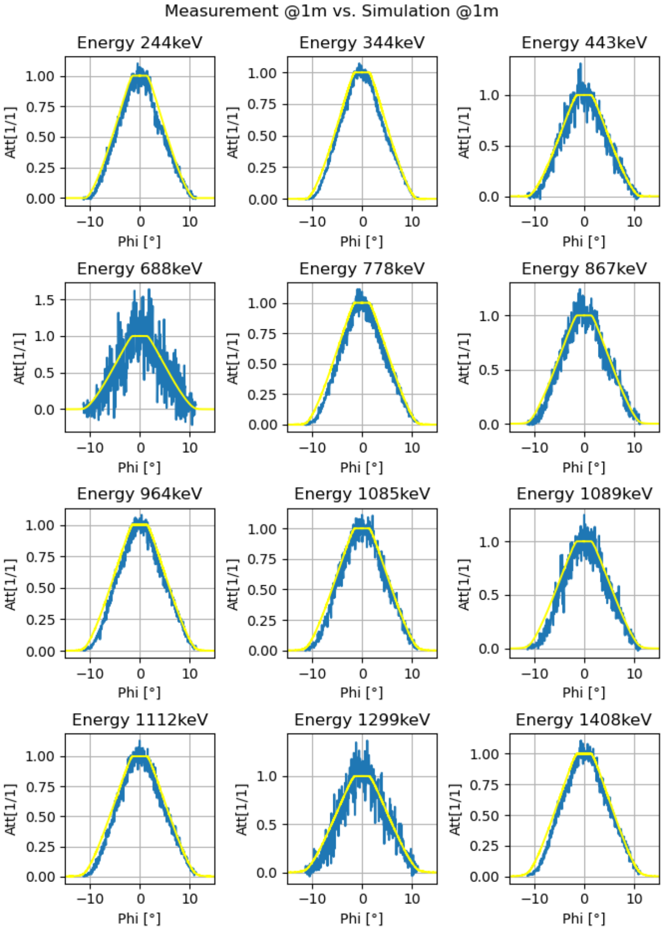

Figure 4 Validation of the collimator function for energies from 244keV to 1408keV. The noise is caused by the different signal strength of the individual Eu152 emissions.

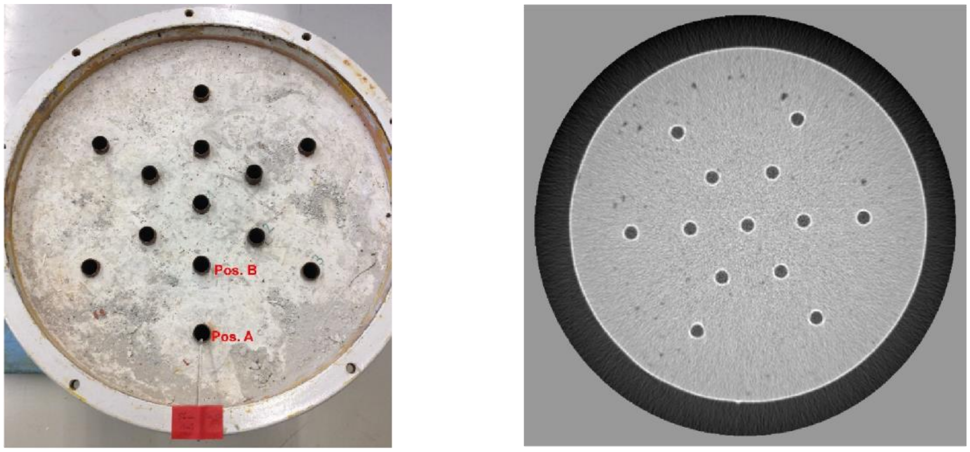

Figure 5 Left: Calibration barrel with cement matrix; Right: Computed tomography using Co60-Source;

INVESTIGATION OF SYSTEMATICS OF SOURCE HYPOTHESIS

To fully reconstruct the true activity distribution of a waste package methods like single photon emission spectroscopy (SPECT) must be employed. In comparison to SGS-measurements those scans require more degrees of freedom for the relative movement of detector and barrel under investigation leading to higher instrumentation effort and longer measurement times [Bertrand Pérot et al. 2018]. Therefore, it is accepted in the field of waste package characterization to limit the source distribution hypothesis to a small number of simple cases.

To investigate this aspect simulated measurement data has been generated by defining an activity distribution and calculating the signal strength according to equation 3. To incorporate the statistical nature of the counting process Poisson random number are generated using the signal strength as expectation value.

The geometry of the analyzed SGS-scans is fixed to 24 segments with continuous scanning. The underlying model used describes the calibration barrel depicted in figure 5. It consist of a cement matrix with a number of steel lined boreholes to insert sources. The matrix is homogeneous and has a comparatively high absorption coefficient of 0.23 cm^{-1}at 334keV.

The generated counts are feed into the Bayes-analysis. To evaluate the posterior distribution the Metropolis-Hastings-algorithm implemented in PyMC3 is used. In total twenty chain with 60k samples are calculated.

Example 1 – Centered source

In the first example, a point source having 10MRad activity is placed in the center of the barrel. The simulated data has an integration time of 200s for each segment, the energy considered is 334keV.

To analyze the synthetic data three hypothesis on the source distribution have been made. A point-like source i.e. one voxel “fires” and carries all the activity, a disk with arbitrary radius and position and finally a torus with arbitrary radius and position with a fixed radial extension of 2cm.

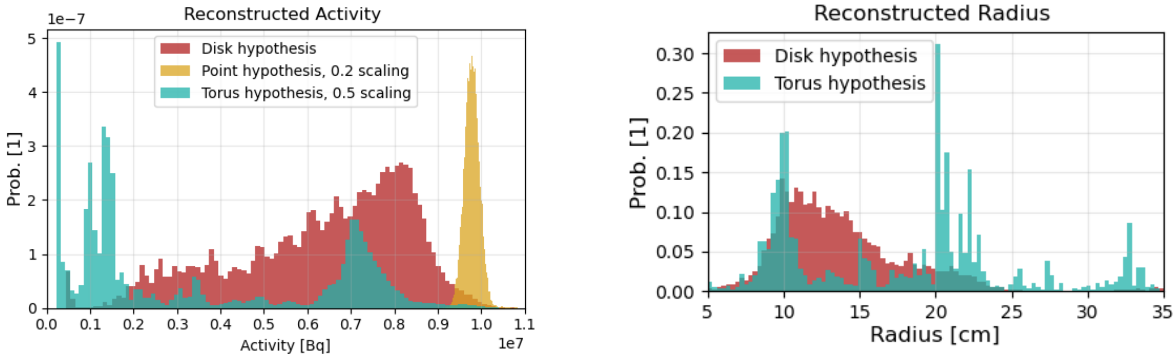

Figure 6: Source: point-like, 10MRad, centered; Left: reconstructed activity, to equalize the y range of the histogram scale factors applied to the individual contributions; Right: Reconstructed radius

Figure 7 shows the posterior distribution of the reconstructed activity and radius of the various assumptions. Only in case of the point hypothesis the activity of 10MBq is correctly reconstructed. The right figure shows the posterior distribution of the radius for the disk and the torus hypothesis. It is evident that it is not possible to constrain this parameter from the SGS-measurement using a single energy. In both cases, the radius varies between the limits of the flat prior distribution ranging from 5 to 35cm. It is worth mentioning that the upper limits the reconstructed activity of both torus and disc assumption coincide with the true activity. Those values are obtained for small radii i.e. when the distributions are close to a point source. On the other hand the torus distribution is able to explain the observed count rates with a total activity as low as 250kBq when placed close to the surface of the barrel. Even if the torus distribution is of limited practical relevance, it demonstrates the importance to investigate the systematic variations introduced by the source distribution hypothesis.

In particular, the distribution for the torus hypothesis show various areas with local maxima (at 10cm, 21cm) and a number of sporadic spikes. Those features are introduced by individual traces of the MCMC algorithm running in various local optima. The improvement of the convergence is still under investigation.

Example 2 – Off- centered disk

The second example consists of a homogeneous disc carrying 10MBq activity with its center placed at 10cm/10cm from the center of the barrel in both directions and having a radius of 10cm.

The reconstruction has been done using the point and the disc hypothesis. The results are shown in figure 7. Using the correct source distribution hypothesis it is possible to reconstruct the correct activity. The radius of the distribution is correctly obtained, too. The bimodal nature of the distribution can be attributed to the discrete nature of the voxel model. While the peak at 10MBq refers to correct position of the source at (10cm/10cm) the other peak belongs to the neighboring voxel closer to the center of the barrel. The point hypothesis leads to a significant higher activity of 34MBq.

While the various solutions in example 1 describe the underlying data correctly – all segments carry the same number of counts apart from the variations introduced by the counting statistics, example 2 allows to identify the correct solution by comparing the prediction of the posterior distribution to the data and therefore discarding the point hypothesis.

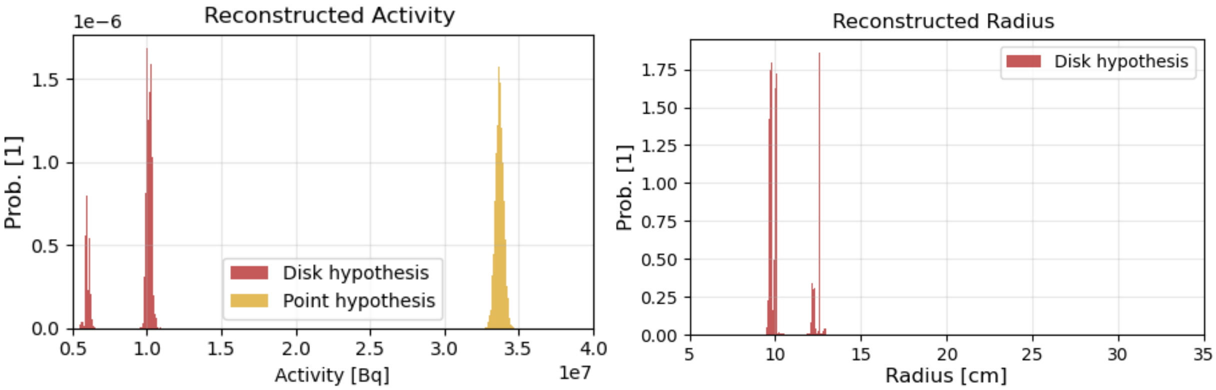

Figure 7. Source: 10cm disc, 10cm off-center; reconstructed activity left and reconstructed radius for example 2

OUTLOOK AND SUMMARY

We have developed a method to evaluate segmented gamma scans using Bayesian statistics. A key element is the modelling of the measurement process to predict the observed quantities i.e. count distribution given the activity. To do so the measured or known attenuation properties of a waste barrel are represented in a discrete voxel model. The weights / efficiencies for each voxel are calculated using a box-intersect algorithm to evaluate the attenuation between each voxel and detector position.

Since the usual SGS-measurement geometry does not allow a full reconstruction of the activity distribution it is necessary to introduce hypotheses on this distribution. Using the presented method one can easily introduce new hypothesis in the algorithm. Up to now, we have point sources, homogenous and torus distribution at arbitrary positions.

In this paper, focus has been put on the investigation of the systematic effects of this source hypothesis. Example 1 clearly demonstrates the importance even if the convergence of the algorithm still needs some attention.

For future work we will focus on the simultaneous evaluation of isotopes emitting multiple gamma energies or waste packages with well-defined nuclide vectors. This offers the possibility to mitigate some of the issues presented here as different energies allow to further constrain the source distribution due to different attenuation properties of the waste matrix.

In this work, the attenuation data of the waste barrel has been derived with a computed tomography using a Co60 source. Even if this allows a precise evaluation of the matrix inside the vessel it is not commonly practiced due to financial and time constraints. Therefore, it is planned to extend the SGS-measurement setup with a transmission source allowing to simultaneously evaluating the attenuation thought the center of the barrel.

REFERENCES

BERTRAND PÉROT, FANNY JALLU, CHRISTIAN PASSARD, OLIVIER GUETON, PIERRE-GUY ALLINEI, LAURENT LOUBET, NICOLAS ESTRE, ERIC SIMON, CÉDRIC CARASCO, CHRISTOPHE ROURE, LIONEL BOUCHER, HERVÉ LAMOTTE, JÉRÔME COMTE, MAÏTÉ BERTAUX, ABDALLAH LYOUSSI, PASCAL FICHET, AND FRÉDÉRICK CARREL. 2018. The characterization of radioactive waste: a critical review of techniques implemented or under development at CEA, France. *EPJ Nuclear Sci. Technol. 4*, 3. https://www.epj-n.org/articles/epjn/full_html/2018/01/epjn170038/epjn170038.html.

BÜCHERL, T., RUMMEL, S., AND KALTHOFF, O. 2021. A Bayesian method for the evaluation of segmented gamma scanning measurements – Description of the principle. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 165887. https://www.sciencedirect.com/science/article/pii/S0168900221008676.

CARASCO, C. 2021. Coupling gamma ray spectrometry and tomography in a Bayesian frame. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 990*, 164985. https://www.sciencedirect.com/science/article/pii/S0168900220313826.

KNOLL, G.F. 2010. *Radiation detection and measurement*. Wiley, Hoboken, NJ.

LALOY, E., ROGIERS, B., BIELEN, A., AND BODEN, S. 2021. *Bayesian inference of 1D activity profiles from segmented gamma scanning of a heterogeneous radioactive waste drum*.

ROBERT, C.P., AND CASELLA, G. 2004. *Monte Carlo statistical methods*. Springer texts in statistics. Springer, New York, NY.

ACKNOWLEDGEMENTS

We gratefully acknowledge the support of the German Federal Ministry of Education and Research (BMBF) under grant number 15S9411.