

## ***Supplementary Material***

We present in this Supplementary Material the full results of our investigation on the performance and the sensitivity to statistical fluctuations of the MLEM and Tikhonov algorithms, evaluated on simulated beta spectra. Twelve radionuclides with various spectrum shapes, energy ranges and counting statistics were selected, matching the experimental limitations of our  $4\pi$  silicon-based detection system.

Each simulated spectrum was determined from a theoretical spectrum convoluted with the response matrix of our apparatus and adding Poisson noise. The same input statistics, ranging from  $10^5$  to  $8.5 \cdot 10^8$ , was considered in each total beta spectrum. However, because of the different theoretical spectral shapes, of the random Poisson noise and of the detection energy threshold of 15 keV applied, this results in different counting statistics in the simulated spectrum for each radionuclide.

For each of the twelve isotopes, we provide in the following a figure that compares the theoretical, true spectrum with the unfolded simulated spectra obtained with the MLEM and Tikhonov algorithms. The same counting statistics of  $10^7$  was considered. Each figure is followed by a table that presents the four root mean square errors (RMSE) described in section 3.1 from the unfolding together with the regularization parameters, over 24 increasing counting statistics. Each RMSE is normalized by the number of counts in the simulated spectrum and expressed in per cent mille (pcm). From inspection of the tables, it is important to note that both algorithms exhibit a similar behavior with the counting statistics: the accuracy of the unfolding process increases with the number of counts in the input spectra. This trend was expected as Poisson noise becomes larger for low counting statistics, increasing the difficulty for both algorithms to regularize the spectra.

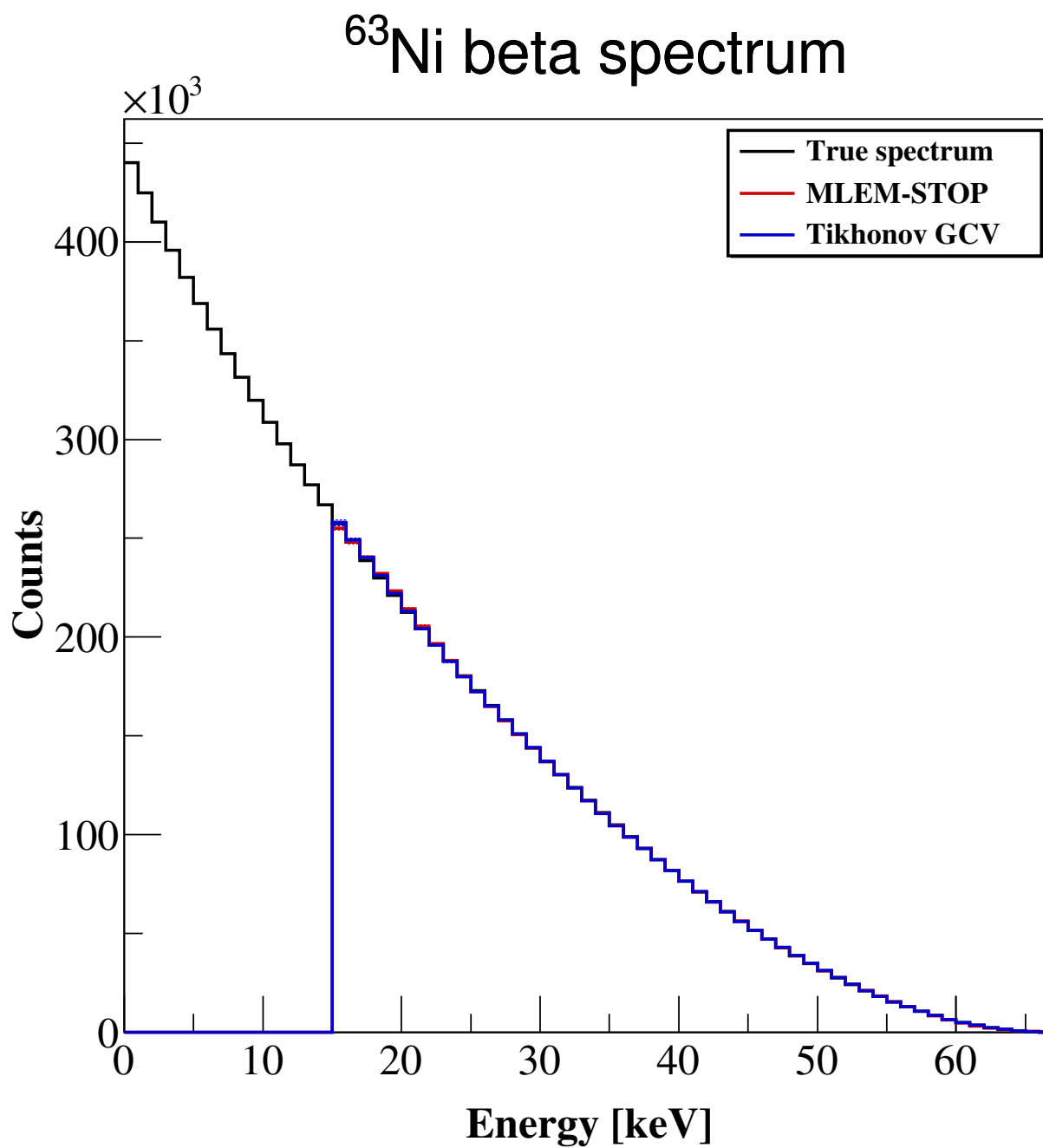


Figure S1: Comparison between the theoretical, true beta spectrum of  $^{63}\text{Ni}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	RMSE <sup>MLEM</sup> <sub>min</sub> [reg. param.]	RMSE <sup>MLEM</sup> <sub>stop</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>min</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>gcv</sub> [reg. param.]
<sup>63</sup> Ni	3.2e+04	46.4 [6]	58.0 [4]	45.8 [4.0e-03]	46.2 [5.2e-03]
	8.0e+04	38.3 [9]	49.8 [5]	32.7 [5.2e-04]	32.9 [6.7e-04]
	1.3e+05	34.7 [11]	43.4 [6]	28.4 [2.4e-04]	28.7 [3.1e-04]
	1.8e+05	32.0 [12]	44.3 [6]	25.0 [1.1e-04]	25.1 [1.5e-04]
	2.2e+05	30.5 [13]	39.1 [7]	23.6 [8.8e-05]	23.6 [8.8e-05]
	2.7e+05	29.2 [15]	39.4 [7]	22.2 [5.3e-05]	22.2 [5.3e-05]
	3.2e+05	28.1 [16]	39.7 [7]	20.9 [4.1e-05]	20.9 [4.1e-05]
	8.0e+05	23.8 [22]	30.5 [10]	16.6 [1.1e-05]	17.1 [6.8e-06]
	1.3e+06	22.0 [26]	28.7 [11]	15.1 [5.3e-06]	15.6 [3.2e-06]
	1.8e+06	21.0 [30]	25.8 [13]	14.2 [3.2e-06]	14.7 [1.9e-06]
	2.2e+06	20.3 [33]	24.7 [14]	13.7 [2.5e-06]	14.4 [1.1e-06]
	2.7e+06	19.7 [37]	23.8 [15]	13.3 [1.9e-06]	13.9 [8.9e-07]
	3.2e+06	18.8 [244]	23.0 [16]	13.0 [1.5e-06]	13.4 [6.9e-07]
	8.0e+06	14.4 [333]	19.2 [24]	11.5 [3.2e-07]	12.0 [1.2e-07]
	1.3e+07	12.8 [379]	17.9 [29]	10.9 [1.5e-07]	11.1 [5.4e-08]
	1.8e+07	11.8 [413]	17.1 [33]	10.3 [7.0e-09]	10.6 [3.2e-08]
	2.2e+07	11.2 [442]	16.5 [37]	9.8 [4.2e-09]	10.2 [1.9e-08]
	2.7e+07	10.8 [468]	16.1 [41]	9.5 [3.3e-09]	9.8 [1.2e-08]
	3.2e+07	10.4 [492]	15.7 [44]	9.2 [2.5e-09]	9.6 [9.0e-09]
	8.0e+07	8.7 [705]	13.7 [69]	8.1 [7.0e-10]	8.1 [1.2e-09]
	1.3e+08	8.0 [872]	12.9 [86]	7.6 [3.3e-10]	7.7 [5.5e-10]
	1.8e+08	7.6 [997]	12.3 [101]	7.4 [2.0e-10]	7.4 [2.5e-10]
	2.2e+08	7.2 [1094]	11.8 [114]	7.2 [1.2e-10]	7.2 [2.0e-10]
	2.7e+08	7.0 [1176]	11.5 [126]	7.0 [9.2e-11]	7.1 [1.2e-10]

Table S1: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for <sup>63</sup>Ni beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

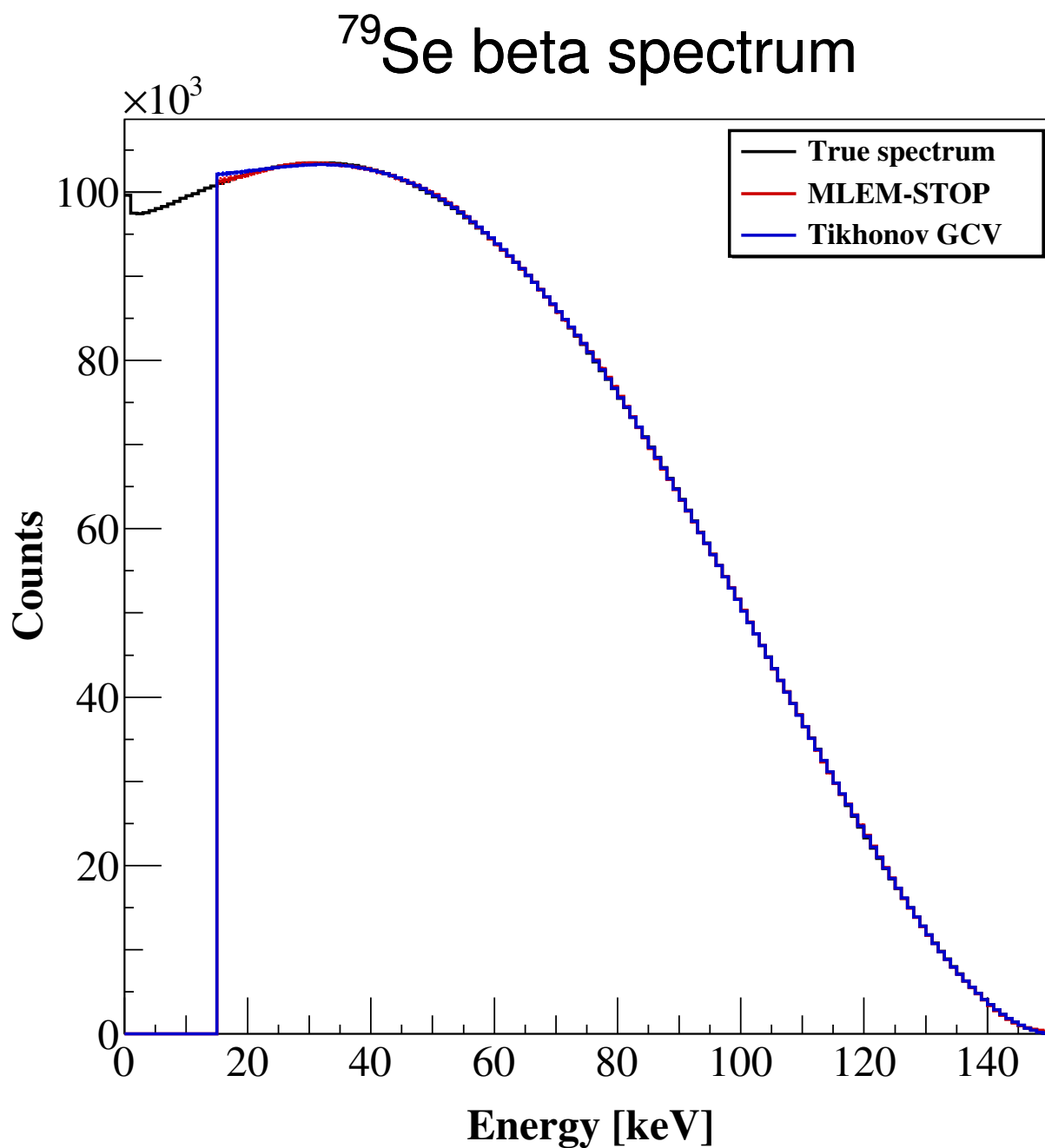


Figure S2: Comparison between the theoretical, true beta spectrum of  $^{79}\text{Se}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{79}\text{Se}$	7.1e+04	12.7 [3]	12.7 [3]	7.8 [1.8e+00]	7.9 [2.4e+00]
	1.8e+05	8.9 [3]	10.2 [4]	6.1 [2.4e-01]	6.4 [5.1e-01]
	2.9e+05	7.3 [3]	8.3 [4]	5.8 [1.1e-01]	6.0 [2.4e-01]
	3.9e+05	6.1 [3]	6.8 [4]	5.3 [6.6e-02]	5.6 [1.4e-01]
	5.0e+05	5.7 [3]	6.2 [4]	5.0 [4.0e-02]	5.4 [1.1e-01]
	6.1e+05	5.0 [3]	5.4 [4]	4.8 [2.4e-02]	5.0 [6.6e-02]
	7.1e+05	4.5 [3]	4.7 [4]	4.7 [1.9e-02]	5.2 [6.6e-02]
	1.8e+06	3.3 [4]	3.3 [4]	3.5 [3.1e-03]	3.9 [1.1e-02]
	2.9e+06	2.6 [4]	2.6 [4]	3.0 [1.1e-03]	3.3 [4.0e-03]
	3.9e+06	2.3 [4]	2.3 [4]	2.7 [5.2e-04]	3.0 [2.4e-03]
	5.0e+06	2.1 [4]	2.2 [5]	2.4 [2.4e-04]	2.8 [1.4e-03]
	6.1e+06	1.8 [5]	1.8 [5]	2.1 [1.1e-04]	2.8 [1.4e-03]
	7.1e+06	1.7 [5]	1.7 [5]	2.0 [6.8e-05]	2.5 [8.7e-04]
	1.8e+07	1.2 [5]	1.2 [5]	1.3 [1.5e-05]	1.7 [1.5e-04]
	2.9e+07	1.1 [6]	1.1 [5]	1.2 [8.8e-06]	1.4 [5.3e-05]
	3.9e+07	1.0 [6]	1.0 [6]	1.0 [4.1e-06]	1.3 [3.2e-05]
	5.0e+07	0.9 [7]	0.9 [6]	0.9 [2.5e-06]	1.2 [2.4e-05]
	6.1e+07	0.9 [7]	0.9 [6]	0.9 [2.5e-06]	1.1 [1.5e-05]
	7.1e+07	0.8 [7]	0.8 [6]	0.8 [1.5e-06]	1.0 [1.1e-05]
	1.8e+08	0.7 [9]	0.7 [7]	0.6 [4.1e-07]	0.7 [1.9e-06]
	2.9e+08	0.6 [19]	0.6 [8]	0.5 [1.9e-07]	0.6 [8.9e-07]
	3.9e+08	0.6 [22]	0.6 [9]	0.4 [1.2e-07]	0.5 [5.3e-07]
	5.0e+08	0.5 [24]	0.6 [9]	0.4 [9.0e-08]	0.4 [3.2e-07]
	6.1e+08	0.5 [26]	0.6 [10]	0.4 [5.4e-08]	0.4 [2.5e-07]

Table S2: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{79}\text{Se}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

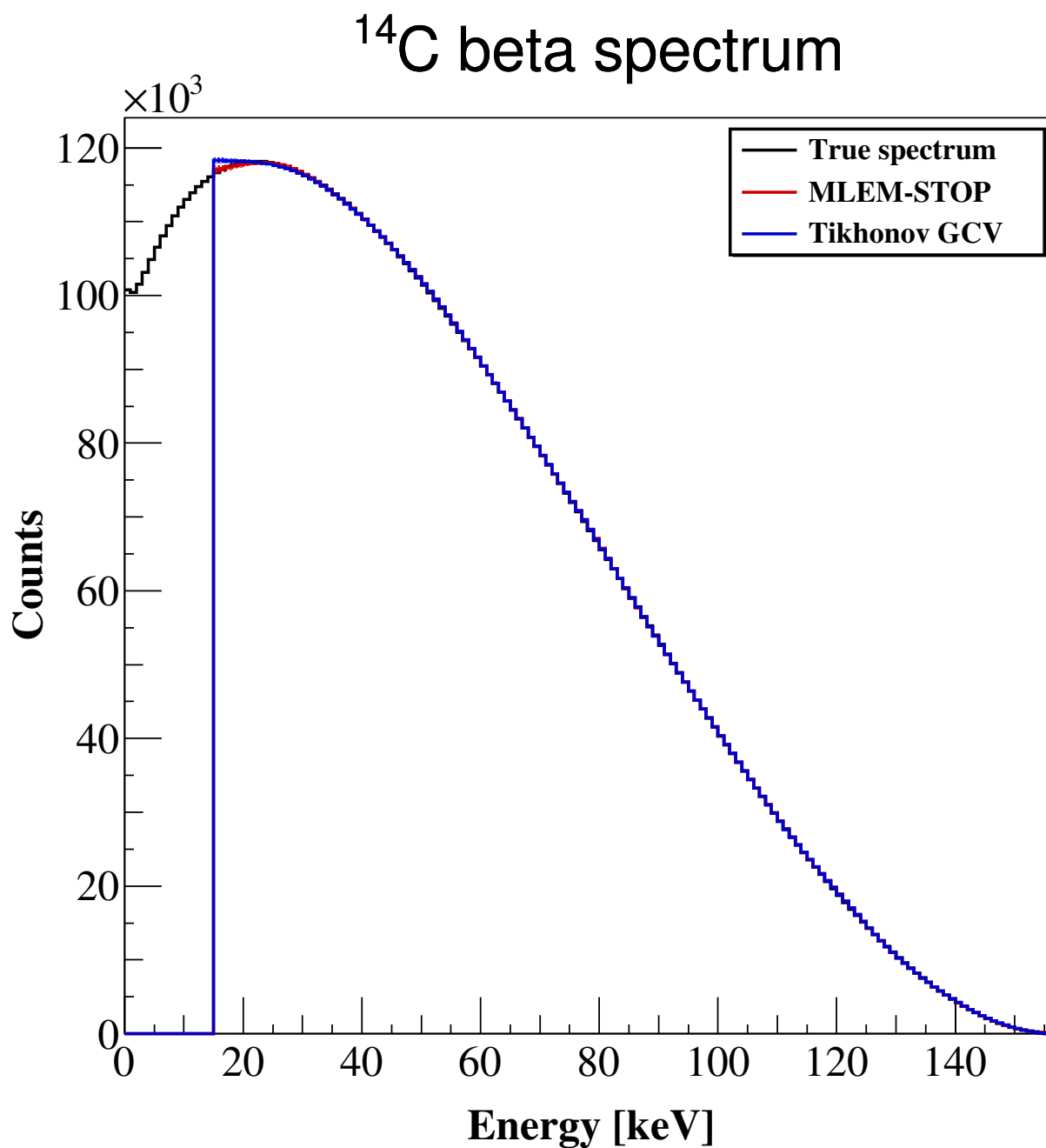


Figure S3: Comparison between the theoretical, true beta spectrum of  $^{14}\text{C}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	RMSE <sup>MLEM</sup> <sub>min</sub> [reg. param.]	RMSE <sup>MLEM</sup> <sub>stop</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>min</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>gcv</sub> [reg. param.]
<sup>14</sup> C	6.9e+04	13.0 [3]	13.0 [3]	10.6 [1.4e+00]	11.4 [5.1e+00]
	1.7e+05	8.9 [3]	10.0 [4]	9.0 [1.4e-01]	10.1 [8.5e-01]
	2.8e+05	6.9 [3]	7.5 [4]	8.3 [5.1e-02]	9.3 [3.1e-01]
	3.8e+05	6.3 [3]	6.6 [4]	7.7 [1.9e-02]	9.0 [1.8e-01]
	4.8e+05	5.2 [4]	5.2 [4]	7.1 [4.0e-03]	8.6 [1.1e-01]
	5.9e+05	5.2 [4]	5.2 [4]	6.8 [5.2e-03]	8.1 [6.6e-02]
	6.9e+05	4.8 [4]	4.8 [4]	6.4 [3.1e-03]	7.6 [4.0e-02]
	1.7e+06	2.8 [4]	2.8 [4]	4.2 [1.9e-04]	5.8 [5.2e-03]
	2.8e+06	2.6 [4]	2.7 [5]	3.6 [8.8e-05]	4.5 [1.1e-03]
	3.8e+06	2.3 [5]	2.3 [5]	3.2 [5.3e-05]	4.1 [6.7e-04]
	4.8e+06	2.1 [5]	2.1 [5]	2.9 [3.2e-05]	3.7 [4.0e-04]
	5.9e+06	1.7 [5]	1.7 [5]	2.5 [2.4e-05]	3.5 [3.1e-04]
	6.9e+06	1.6 [5]	1.6 [5]	2.4 [1.9e-05]	3.2 [1.9e-04]
	1.7e+07	1.3 [6]	1.3 [6]	1.7 [4.1e-06]	2.2 [3.2e-05]
	2.8e+07	1.1 [6]	1.1 [6]	1.4 [1.9e-06]	1.8 [1.1e-05]
	3.8e+07	1.1 [7]	1.1 [6]	1.3 [1.1e-06]	1.6 [6.8e-06]
	4.8e+07	1.0 [7]	1.0 [6]	1.2 [8.9e-07]	1.4 [4.1e-06]
	5.9e+07	0.9 [8]	1.0 [6]	1.1 [6.9e-07]	1.3 [2.5e-06]
	6.9e+07	0.9 [8]	0.9 [7]	1.0 [5.3e-07]	1.2 [1.9e-06]
	1.7e+08	0.7 [20]	0.8 [7]	0.8 [1.2e-07]	0.8 [3.2e-07]
	2.8e+08	0.6 [25]	0.8 [8]	0.6 [5.4e-08]	0.7 [1.2e-07]
	3.8e+08	0.6 [31]	0.7 [8]	0.6 [3.2e-08]	0.6 [5.4e-08]
	4.8e+08	0.5 [35]	0.7 [9]	0.5 [1.9e-08]	0.6 [3.2e-08]
	5.9e+08	0.5 [40]	0.7 [9]	0.5 [1.5e-08]	0.5 [2.5e-08]

Table S3: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for <sup>14</sup>C beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

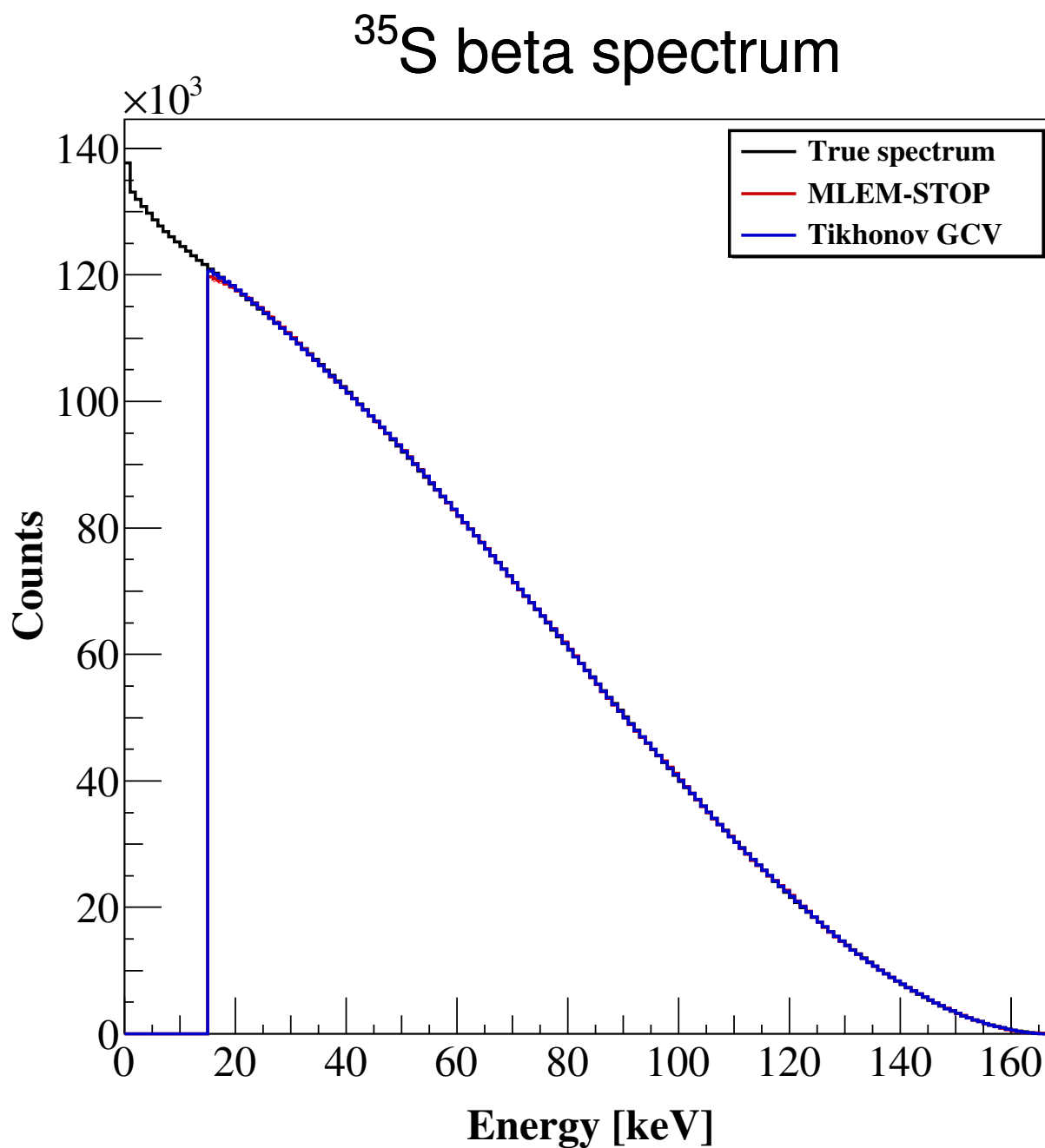


Figure S4: Comparison between the theoretical, true beta spectrum of  $^{35}\text{S}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.



Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{35}\text{S}$	6.6e+04	11.3 [3]	12.6 [4]	4.6 [6.6e-01]	8.3 [3.9e+00]
	1.7e+05	8.7 [4]	8.7 [4]	3.5 [1.8e-01]	3.8 [1.1e-01]
	2.7e+05	6.6 [4]	6.6 [4]	3.3 [8.6e-02]	3.4 [6.6e-02]
	3.6e+05	6.5 [4]	6.5 [4]	2.8 [6.6e-02]	3.6 [2.4e-02]
	4.6e+05	5.9 [4]	5.9 [4]	2.6 [4.0e-02]	3.2 [1.9e-02]
	5.6e+05	4.9 [5]	5.1 [4]	2.5 [3.1e-02]	2.7 [1.9e-02]
	6.6e+05	4.6 [5]	5.0 [4]	2.4 [2.4e-02]	2.6 [1.4e-02]
	1.7e+06	3.7 [6]	3.8 [5]	1.8 [6.7e-03]	2.1 [3.1e-03]
	2.6e+06	3.1 [7]	3.6 [5]	1.6 [3.1e-03]	1.8 [1.4e-03]
	3.6e+06	2.8 [7]	2.9 [6]	1.5 [1.9e-03]	1.7 [8.7e-04]
	4.6e+06	2.3 [8]	2.6 [6]	1.4 [1.1e-03]	1.4 [8.7e-04]
	5.6e+06	2.2 [9]	2.6 [6]	1.3 [8.7e-04]	1.3 [6.7e-04]
	6.6e+06	2.2 [9]	2.7 [6]	1.3 [6.7e-04]	1.4 [4.0e-04]
	1.7e+07	1.6 [12]	2.2 [7]	1.1 [1.1e-04]	1.1 [1.1e-04]
	2.6e+07	1.4 [14]	1.8 [8]	0.9 [4.1e-05]	0.9 [5.3e-05]
	3.6e+07	1.2 [16]	1.6 [9]	0.8 [1.5e-05]	0.9 [3.2e-05]
	4.6e+07	1.0 [18]	1.5 [9]	0.7 [6.8e-06]	0.8 [1.9e-05]
	5.6e+07	1.1 [18]	1.6 [9]	0.7 [5.3e-06]	0.8 [1.5e-05]
	6.6e+07	1.0 [19]	1.4 [10]	0.7 [4.1e-06]	0.7 [8.8e-06]
	1.7e+08	0.8 [24]	1.1 [12]	0.5 [6.9e-07]	0.5 [1.5e-06]
	2.6e+08	0.7 [28]	0.9 [14]	0.4 [3.2e-07]	0.4 [5.3e-07]
	3.6e+08	0.6 [31]	0.8 [16]	0.4 [1.9e-07]	0.4 [2.5e-07]
	4.6e+08	0.6 [33]	0.8 [17]	0.4 [1.2e-07]	0.4 [1.5e-07]
	5.6e+08	0.6 [35]	0.7 [18]	0.3 [9.0e-08]	0.3 [1.2e-07]

Table S4: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{35}\text{S}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

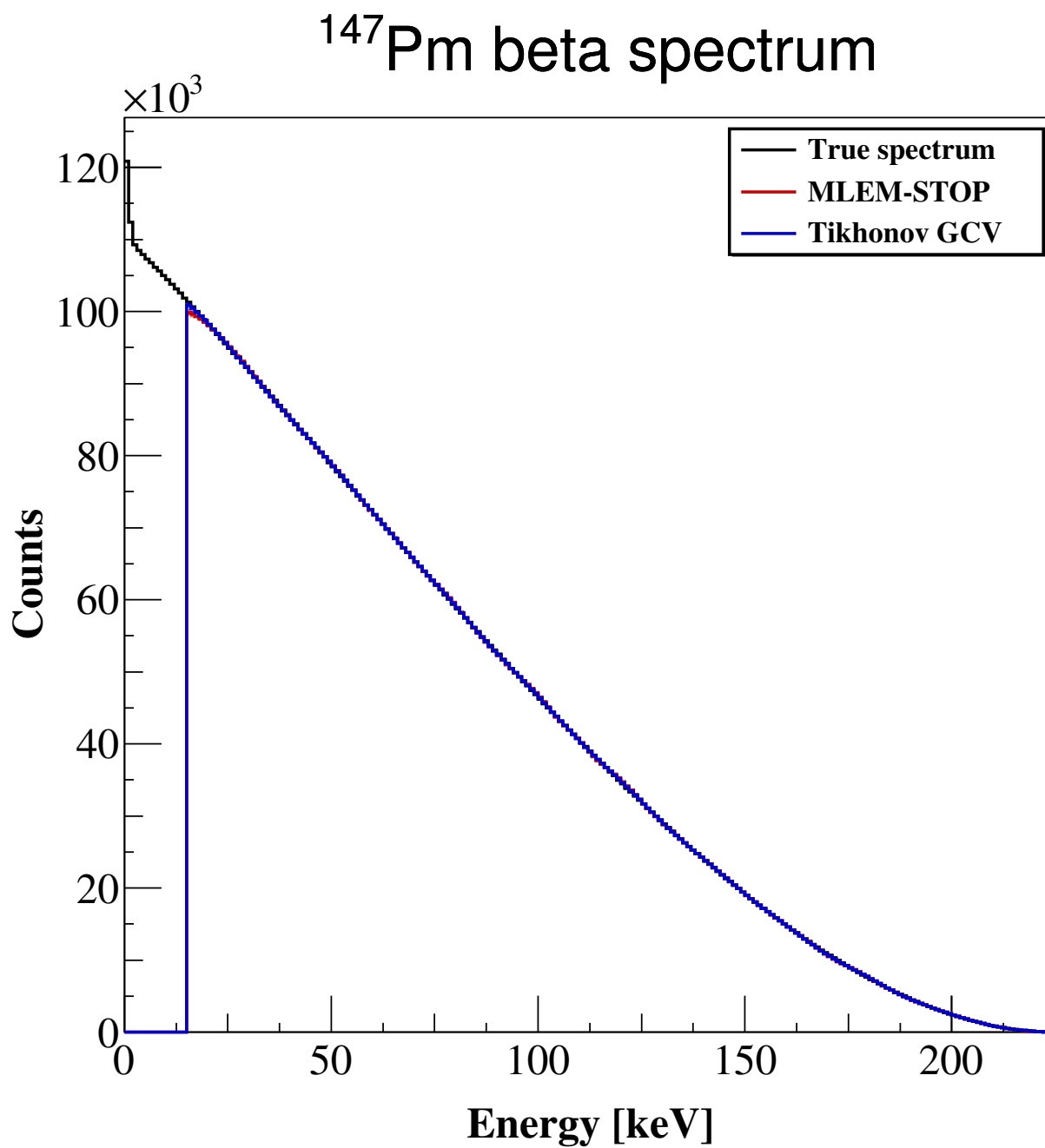


Figure S5: Comparison between the theoretical, true beta spectrum of  $^{147}\text{Pm}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{147}\text{Pm}$	7.0e+04	11.0 [3]	11.5 [4]	5.5 [8.5e-01]	6.3 [1.8e+00]
	1.7e+05	7.4 [4]	7.4 [4]	3.5 [1.8e-01]	3.6 [1.4e-01]
	2.8e+05	6.1 [4]	6.1 [4]	3.2 [8.6e-02]	3.3 [6.6e-02]
	3.8e+05	5.8 [5]	5.8 [4]	3.0 [6.6e-02]	3.1 [4.0e-02]
	4.9e+05	5.2 [5]	5.5 [4]	2.7 [4.0e-02]	2.9 [2.4e-02]
	5.9e+05	4.8 [5]	4.8 [5]	2.5 [3.1e-02]	2.7 [1.9e-02]
	7.0e+05	4.5 [5]	4.5 [5]	2.4 [2.4e-02]	2.6 [1.4e-02]
	1.7e+06	3.2 [6]	3.5 [5]	1.8 [5.2e-03]	1.9 [3.1e-03]
	2.8e+06	2.7 [7]	2.8 [6]	1.6 [2.4e-03]	1.6 [1.9e-03]
	3.8e+06	2.6 [8]	2.8 [6]	1.5 [1.4e-03]	1.6 [8.7e-04]
	4.9e+06	2.2 [8]	2.5 [6]	1.4 [8.7e-04]	1.4 [6.7e-04]
	5.9e+06	2.2 [9]	2.6 [6]	1.3 [8.7e-04]	1.4 [4.0e-04]
	7.0e+06	2.1 [9]	2.6 [6]	1.3 [5.2e-04]	1.3 [4.0e-04]
	1.7e+07	1.5 [12]	1.8 [8]	1.0 [1.1e-04]	1.0 [8.8e-05]
	2.8e+07	1.3 [14]	1.7 [8]	0.9 [3.2e-05]	0.9 [4.1e-05]
	3.8e+07	1.1 [16]	1.4 [9]	0.8 [1.5e-05]	0.8 [2.4e-05]
	4.9e+07	1.0 [18]	1.2 [10]	0.7 [6.8e-06]	0.8 [1.9e-05]
	5.9e+07	0.9 [19]	1.2 [10]	0.7 [5.3e-06]	0.7 [1.1e-05]
	7.0e+07	0.9 [19]	1.2 [10]	0.7 [4.1e-06]	0.7 [8.8e-06]
	1.7e+08	0.7 [24]	0.9 [13]	0.5 [6.9e-07]	0.5 [1.5e-06]
	2.8e+08	0.6 [28]	0.8 [14]	0.5 [3.2e-07]	0.5 [5.3e-07]
	3.8e+08	0.6 [31]	0.7 [16]	0.4 [1.9e-07]	0.4 [3.2e-07]
	4.9e+08	0.6 [33]	0.7 [17]	0.4 [1.2e-07]	0.4 [1.9e-07]
	5.9e+08	0.5 [35]	0.7 [18]	0.4 [9.0e-08]	0.4 [1.2e-07]

Table S5: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{147}\text{Pm}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

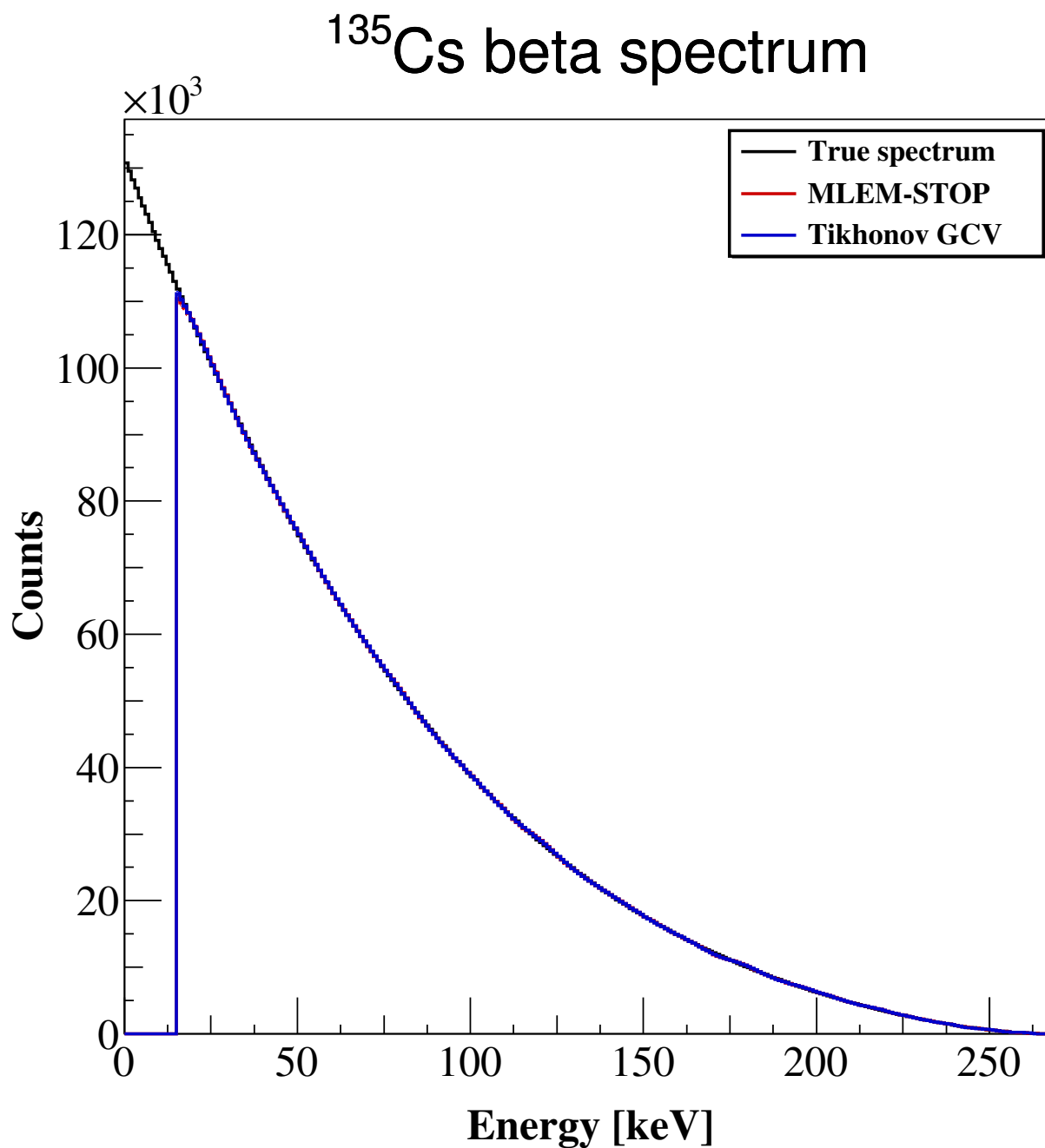


Figure S6: Comparison between the theoretical, true beta spectrum of  $^{135}\text{Cs}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{135}\text{Cs}$	6.7e+04	12.5 [4]	12.5 [4]	6.5 [4.0e-01]	8.7 [1.1e-01]
	1.7e+05	8.2 [4]	8.2 [4]	4.4 [8.6e-02]	5.2 [4.0e-02]
	2.7e+05	6.8 [5]	6.8 [5]	4.0 [4.0e-02]	4.6 [1.9e-02]
	3.7e+05	6.3 [5]	6.3 [5]	3.7 [2.4e-02]	4.3 [1.1e-02]
	4.7e+05	5.3 [6]	5.4 [5]	3.5 [1.4e-02]	3.6 [1.1e-02]
	5.7e+05	4.9 [6]	5.1 [5]	3.3 [1.1e-02]	3.4 [8.6e-03]
	6.7e+05	5.0 [6]	5.3 [5]	3.2 [8.6e-03]	3.5 [5.2e-03]
	1.7e+06	3.4 [8]	3.7 [6]	2.5 [1.9e-03]	2.5 [1.4e-03]
	2.7e+06	2.8 [9]	3.5 [6]	2.2 [6.7e-04]	2.2 [6.7e-04]
	3.7e+06	2.5 [11]	2.9 [7]	2.0 [3.1e-04]	2.0 [4.0e-04]
	4.7e+06	2.5 [11]	3.0 [7]	2.0 [2.4e-04]	2.0 [2.4e-04]
	5.7e+06	2.2 [12]	2.9 [7]	1.8 [1.1e-04]	1.8 [1.9e-04]
	6.7e+06	2.1 [13]	2.5 [8]	1.7 [8.8e-05]	1.7 [1.5e-04]
	1.7e+07	1.5 [17]	2.1 [9]	1.2 [1.5e-05]	1.3 [2.4e-05]
	2.7e+07	1.4 [19]	1.8 [10]	1.1 [5.3e-06]	1.1 [1.1e-05]
	3.7e+07	1.3 [21]	1.7 [11]	1.0 [3.2e-06]	1.0 [5.3e-06]
	4.7e+07	1.2 [22]	1.5 [12]	0.9 [2.5e-06]	0.9 [3.2e-06]
	5.7e+07	1.1 [23]	1.5 [12]	0.9 [1.5e-06]	0.9 [2.5e-06]
	6.7e+07	1.1 [25]	1.4 [13]	0.8 [1.1e-06]	0.8 [1.5e-06]
	1.7e+08	0.9 [33]	1.1 [17]	0.7 [2.5e-07]	0.7 [2.5e-07]
	2.7e+08	0.8 [38]	1.0 [19]	0.6 [1.2e-07]	0.6 [9.0e-08]
	3.7e+08	0.8 [43]	0.9 [22]	0.6 [6.9e-08]	0.6 [5.4e-08]
	4.7e+08	0.7 [47]	0.8 [24]	0.5 [5.4e-08]	0.6 [3.2e-08]
	5.7e+08	0.7 [51]	0.8 [25]	0.5 [4.2e-08]	0.5 [2.5e-08]

Table S6: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{135}\text{Cs}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

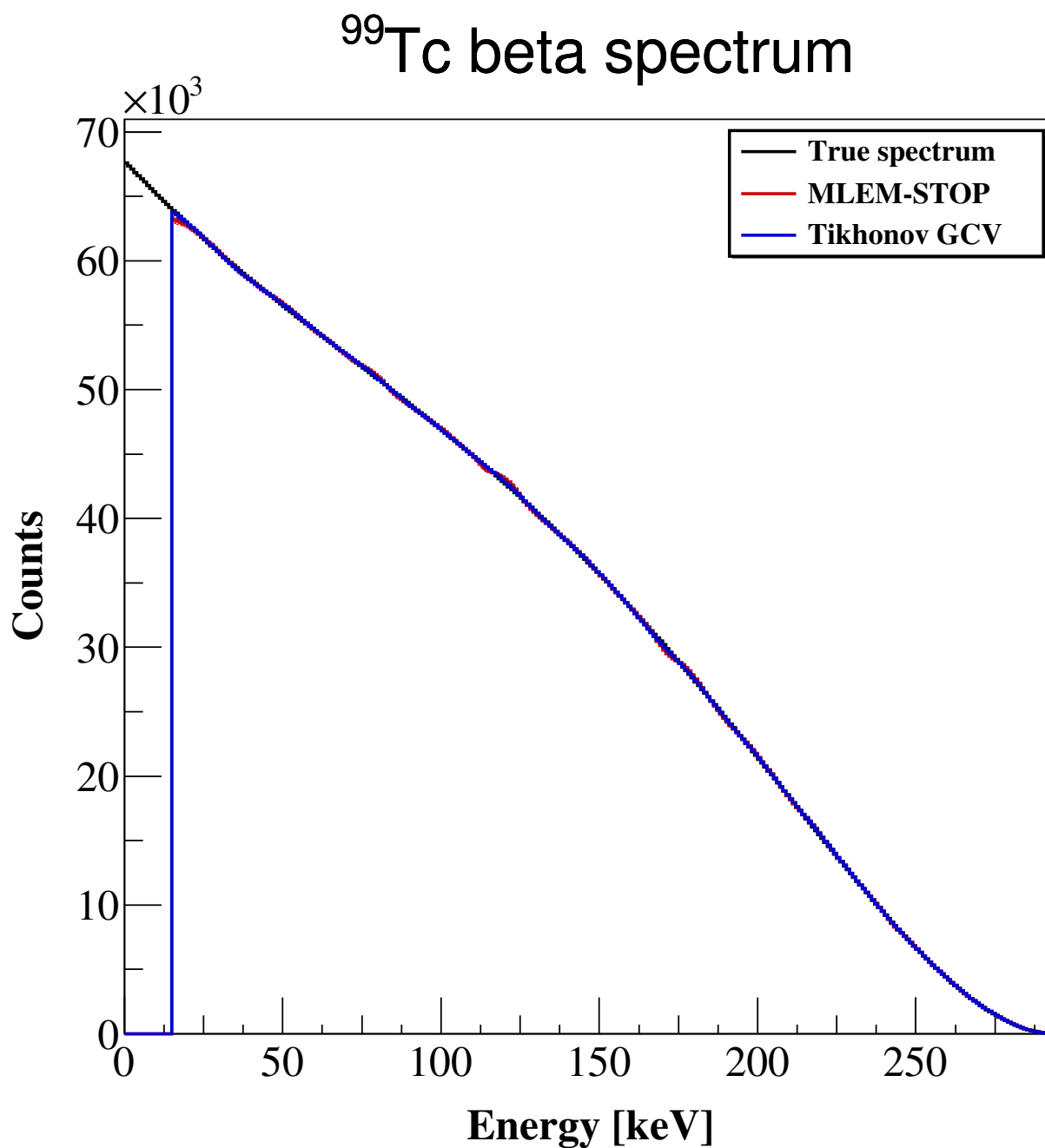


Figure S7: Comparison between the theoretical, true beta spectrum of  $^{99}\text{Tc}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{99}\text{Tc}$	7.7e+04	11.5 [3]	12.6 [4]	5.1 [3.0e+00]	6.4 [5.0e+01]
	1.9e+05	8.3 [3]	8.4 [4]	3.4 [6.6e-01]	4.3 [6.5e+00]
	3.1e+05	6.6 [4]	6.6 [4]	2.6 [3.1e-01]	3.7 [3.0e+00]
	4.2e+05	5.8 [4]	5.8 [4]	2.2 [1.8e-01]	2.9 [1.1e+00]
	5.4e+05	5.1 [4]	5.1 [4]	1.9 [1.4e-01]	2.3 [4.0e-01]
	6.5e+05	4.4 [4]	4.4 [4]	1.8 [1.1e-01]	1.9 [1.8e-01]
	7.7e+05	4.2 [4]	4.2 [4]	1.8 [8.6e-02]	1.8 [8.6e-02]
	1.9e+06	3.0 [5]	3.0 [5]	1.2 [1.9e-02]	1.8 [3.1e-03]
	3.1e+06	2.4 [6]	2.4 [5]	1.0 [8.6e-03]	1.2 [4.0e-03]
	4.2e+06	2.1 [6]	2.2 [5]	0.9 [5.2e-03]	1.0 [2.4e-03]
	5.4e+06	1.9 [6]	1.9 [6]	0.9 [4.0e-03]	0.9 [1.9e-03]
	6.5e+06	1.7 [7]	1.8 [6]	0.8 [3.1e-03]	0.9 [1.4e-03]
	7.7e+06	1.6 [7]	1.7 [6]	0.7 [2.4e-03]	0.8 [1.1e-03]
	1.9e+07	1.2 [9]	1.2 [7]	0.6 [5.2e-04]	0.7 [1.9e-04]
	3.1e+07	1.0 [10]	1.1 [7]	0.5 [2.4e-04]	0.5 [1.1e-04]
	4.2e+07	0.9 [11]	0.9 [8]	0.5 [1.5e-04]	0.5 [6.8e-05]
	5.4e+07	0.8 [11]	0.9 [8]	0.4 [8.8e-05]	0.5 [5.3e-05]
	6.5e+07	0.7 [12]	0.9 [8]	0.4 [6.8e-05]	0.4 [3.2e-05]
	7.7e+07	0.7 [13]	0.8 [8]	0.4 [5.3e-05]	0.4 [3.2e-05]
	1.9e+08	0.5 [17]	0.6 [10]	0.3 [8.8e-06]	0.3 [6.8e-06]
	3.1e+08	0.4 [19]	0.5 [11]	0.3 [2.5e-06]	0.3 [3.2e-06]
	4.2e+08	0.4 [20]	0.5 [12]	0.3 [1.1e-06]	0.3 [1.9e-06]
	5.4e+08	0.3 [22]	0.4 [13]	0.2 [6.9e-07]	0.2 [1.1e-06]
	6.5e+08	0.3 [23]	0.4 [13]	0.2 [5.3e-07]	0.2 [8.9e-07]

Table S7: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{99}\text{Tc}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

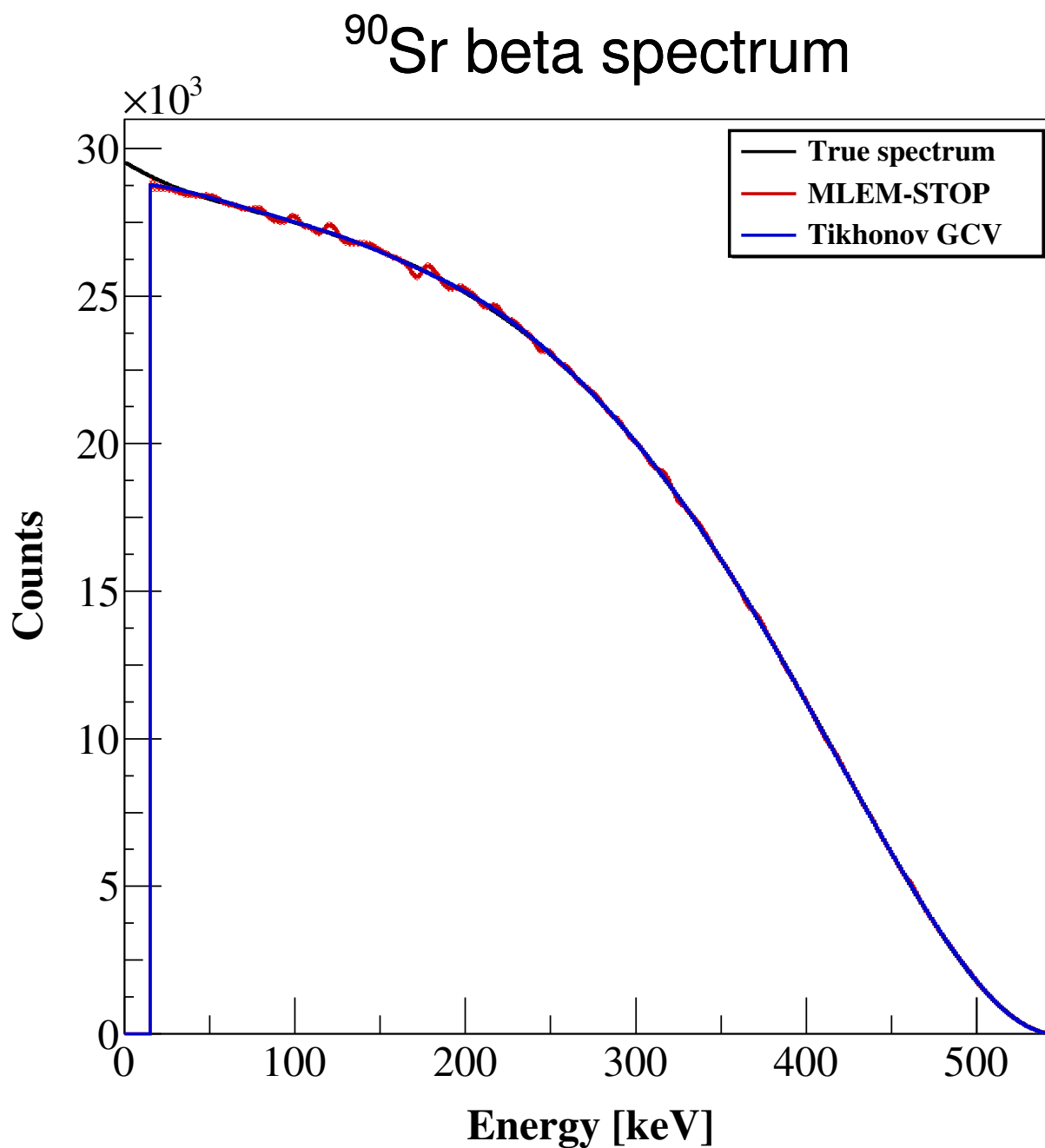


Figure S8: Comparison between the theoretical, true beta spectrum of  $^{90}\text{Sr}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.



Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{90}\text{Sr}$	8.4e+04	8.3 [3]	8.3 [3]	1.5 [1.4e+03]	1.6 [2.3e+03]
	2.1e+05	5.2 [3]	5.5 [4]	1.1 [3.9e+02]	1.2 [6.5e+02]
	3.3e+05	4.4 [3]	4.6 [4]	1.0 [1.8e+02]	1.1 [3.9e+02]
	4.6e+05	3.8 [4]	3.8 [4]	1.0 [1.4e+02]	1.0 [2.3e+02]
	5.8e+05	3.4 [4]	3.4 [4]	0.9 [5.0e+01]	1.0 [1.8e+02]
	7.1e+05	3.1 [4]	3.1 [4]	0.8 [3.0e+01]	0.8 [1.1e+02]
	8.3e+05	2.8 [4]	2.8 [4]	0.8 [1.8e+01]	0.9 [8.4e+01]
	2.1e+06	2.0 [4]	2.0 [5]	0.6 [1.8e+00]	0.7 [1.8e+01]
	3.3e+06	1.5 [5]	1.5 [5]	0.5 [6.6e-01]	0.6 [5.1e+00]
	4.6e+06	1.3 [5]	1.3 [5]	0.5 [2.4e-01]	0.5 [2.4e+00]
	5.8e+06	1.2 [5]	1.2 [5]	0.4 [1.1e-01]	0.5 [1.4e+00]
	7.1e+06	1.2 [5]	1.2 [5]	0.4 [5.1e-02]	0.5 [8.5e-01]
	8.3e+06	1.0 [6]	1.1 [5]	0.4 [5.1e-02]	0.4 [6.6e-01]
	2.1e+07	0.7 [6]	0.7 [6]	0.3 [8.6e-03]	0.3 [8.6e-02]
	3.3e+07	0.6 [7]	0.6 [6]	0.2 [4.0e-03]	0.3 [3.1e-02]
	4.6e+07	0.5 [7]	0.5 [7]	0.2 [2.4e-03]	0.2 [1.1e-02]
	5.8e+07	0.4 [8]	0.4 [7]	0.2 [1.9e-03]	0.2 [5.2e-03]
	7.1e+07	0.4 [8]	0.4 [7]	0.2 [1.1e-03]	0.2 [3.1e-03]
	8.3e+07	0.4 [8]	0.4 [7]	0.1 [8.7e-04]	0.1 [1.9e-03]
	2.1e+08	0.3 [9]	0.3 [8]	0.1 [1.9e-04]	0.1 [2.4e-04]
	3.3e+08	0.2 [10]	0.2 [8]	0.1 [8.8e-05]	0.1 [6.8e-05]
	4.6e+08	0.2 [11]	0.2 [9]	0.1 [5.3e-05]	0.1 [4.1e-05]
	5.8e+08	0.2 [11]	0.2 [9]	0.1 [4.1e-05]	0.1 [2.4e-05]
	7.1e+08	0.2 [12]	0.2 [9]	0.1 [3.2e-05]	0.1 [1.9e-05]

Table S8: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{90}\text{Sr}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

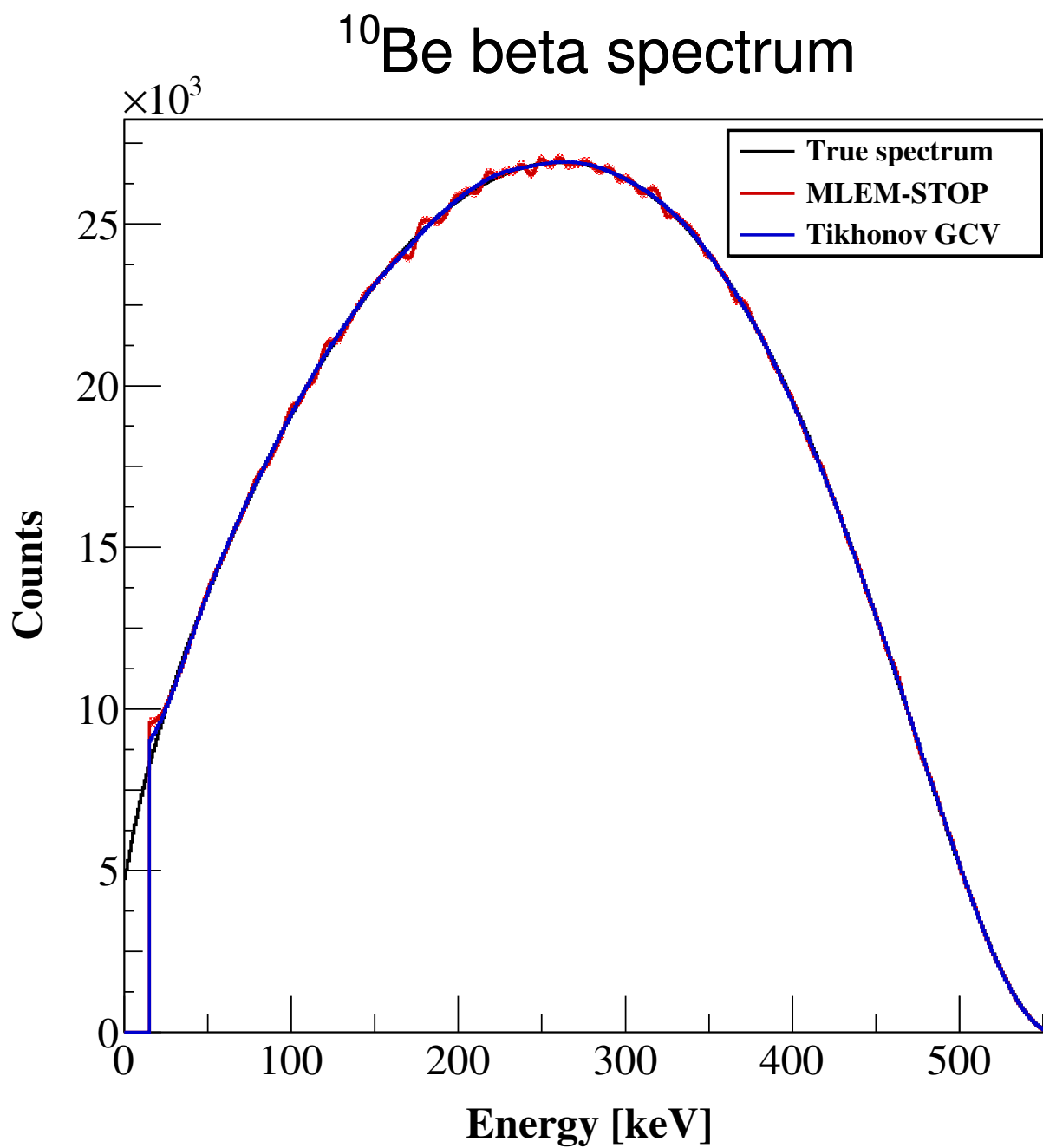


Figure S9: Comparison between the theoretical, true beta spectrum of  $^{10}\text{Be}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{10}\text{Be}$	8.7e+04	8.6 [3]	9.0 [4]	2.9 [8.4e+01]	2.9 [1.1e+02]
	2.2e+05	6.2 [4]	6.2 [4]	2.0 [1.1e+01]	2.1 [1.8e+01]
	3.5e+05	5.2 [5]	5.2 [5]	1.8 [3.9e+00]	1.8 [8.5e+00]
	4.8e+05	4.7 [5]	4.7 [5]	1.5 [2.4e+00]	1.6 [5.1e+00]
	6.1e+05	4.2 [6]	4.2 [5]	1.4 [1.4e+00]	1.4 [3.0e+00]
	7.4e+05	3.9 [6]	4.0 [5]	1.3 [6.6e-01]	1.4 [2.4e+00]
	8.7e+05	3.7 [6]	3.8 [5]	1.2 [6.6e-01]	1.3 [1.8e+00]
	2.2e+06	2.6 [8]	2.7 [7]	1.0 [8.6e-02]	1.1 [4.0e-01]
	3.5e+06	2.2 [10]	2.3 [7]	0.8 [3.1e-02]	0.9 [1.4e-01]
	4.8e+06	1.9 [11]	2.0 [8]	0.8 [1.9e-02]	0.8 [8.6e-02]
	6.1e+06	1.8 [11]	1.9 [8]	0.7 [1.1e-02]	0.8 [5.1e-02]
	7.4e+06	1.6 [12]	1.8 [8]	0.6 [6.7e-03]	0.7 [4.0e-02]
	8.7e+06	1.5 [13]	1.6 [9]	0.6 [5.2e-03]	0.7 [3.1e-02]
	2.2e+07	1.1 [16]	1.2 [10]	0.4 [8.7e-04]	0.5 [4.0e-03]
	3.5e+07	0.9 [18]	1.0 [11]	0.4 [3.1e-04]	0.4 [1.4e-03]
	4.8e+07	0.8 [20]	0.9 [12]	0.3 [1.9e-04]	0.4 [6.7e-04]
	6.1e+07	0.7 [21]	0.8 [13]	0.3 [1.1e-04]	0.3 [4.0e-04]
	7.4e+07	0.6 [22]	0.7 [14]	0.3 [8.8e-05]	0.3 [3.1e-04]
	8.7e+07	0.6 [23]	0.7 [14]	0.3 [6.8e-05]	0.3 [1.5e-04]
	2.2e+08	0.4 [30]	0.5 [17]	0.2 [1.1e-05]	0.2 [3.2e-05]
	3.5e+08	0.4 [33]	0.4 [19]	0.2 [5.3e-06]	0.2 [1.1e-05]
	4.8e+08	0.3 [36]	0.4 [21]	0.2 [2.5e-06]	0.2 [6.8e-06]
	6.1e+08	0.3 [38]	0.3 [22]	0.1 [1.9e-06]	0.1 [4.1e-06]
	7.4e+08	0.3 [40]	0.3 [23]	0.1 [1.1e-06]	0.1 [3.2e-06]

Table S9: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{10}\text{Be}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

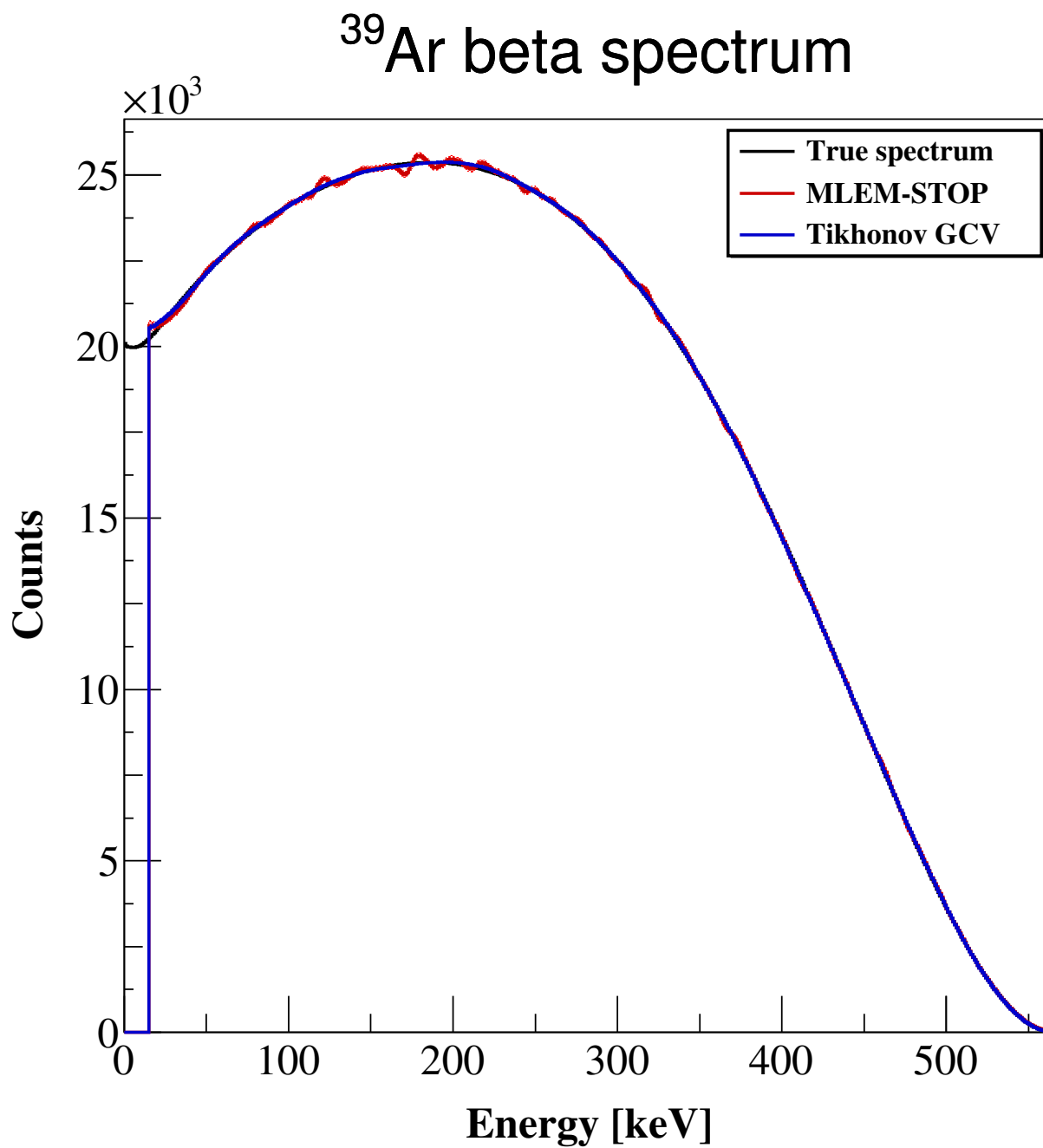


Figure S10: Comparison between the theoretical, true beta spectrum of  $^{39}\text{Ar}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{39}\text{Ar}$	8.5e+04	6.6 [2]	7.1 [3]	2.1 [2.3e+02]	2.1 [3.0e+02]
	2.1e+05	4.5 [3]	4.5 [3]	1.3 [3.0e+01]	1.4 [8.4e+01]
	3.4e+05	3.7 [3]	4.1 [4]	1.2 [1.8e+01]	1.3 [3.0e+01]
	4.7e+05	3.3 [3]	3.7 [4]	1.0 [8.5e+00]	1.1 [1.8e+01]
	5.9e+05	2.8 [3]	3.1 [4]	1.0 [5.1e+00]	1.0 [1.1e+01]
	7.2e+05	2.7 [3]	3.0 [4]	0.9 [5.1e+00]	0.9 [8.5e+00]
	8.5e+05	2.5 [3]	2.8 [4]	0.9 [3.9e+00]	0.9 [6.5e+00]
	2.1e+06	1.7 [3]	1.8 [4]	0.6 [6.6e-01]	0.6 [1.1e+00]
	3.4e+06	1.4 [3]	1.4 [4]	0.5 [2.4e-01]	0.6 [5.1e-01]
	4.7e+06	1.2 [4]	1.2 [4]	0.5 [1.4e-01]	0.5 [3.1e-01]
	5.9e+06	1.1 [4]	1.1 [4]	0.5 [8.6e-02]	0.5 [1.8e-01]
	7.2e+06	1.0 [4]	1.0 [4]	0.4 [6.6e-02]	0.4 [1.4e-01]
	8.5e+06	0.9 [4]	0.9 [4]	0.4 [5.1e-02]	0.4 [1.1e-01]
	2.1e+07	0.7 [4]	0.7 [5]	0.3 [8.6e-03]	0.3 [2.4e-02]
	3.4e+07	0.6 [4]	0.6 [5]	0.3 [3.1e-03]	0.3 [1.1e-02]
	4.7e+07	0.5 [5]	0.5 [5]	0.2 [1.9e-03]	0.3 [5.2e-03]
	5.9e+07	0.4 [5]	0.4 [5]	0.2 [1.1e-03]	0.2 [4.0e-03]
	7.2e+07	0.4 [5]	0.4 [5]	0.2 [8.7e-04]	0.2 [2.4e-03]
	8.5e+07	0.4 [6]	0.4 [5]	0.2 [5.2e-04]	0.2 [1.9e-03]
	2.1e+08	0.3 [8]	0.3 [6]	0.1 [1.1e-04]	0.1 [3.1e-04]
	3.4e+08	0.2 [10]	0.3 [6]	0.1 [4.1e-05]	0.1 [1.1e-04]
	4.7e+08	0.2 [11]	0.2 [6]	0.1 [2.4e-05]	0.1 [6.8e-05]
	5.9e+08	0.2 [12]	0.2 [7]	0.1 [1.5e-05]	0.1 [4.1e-05]
	7.2e+08	0.2 [13]	0.2 [7]	0.1 [1.1e-05]	0.1 [2.4e-05]

Table S10: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{39}\text{Ar}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

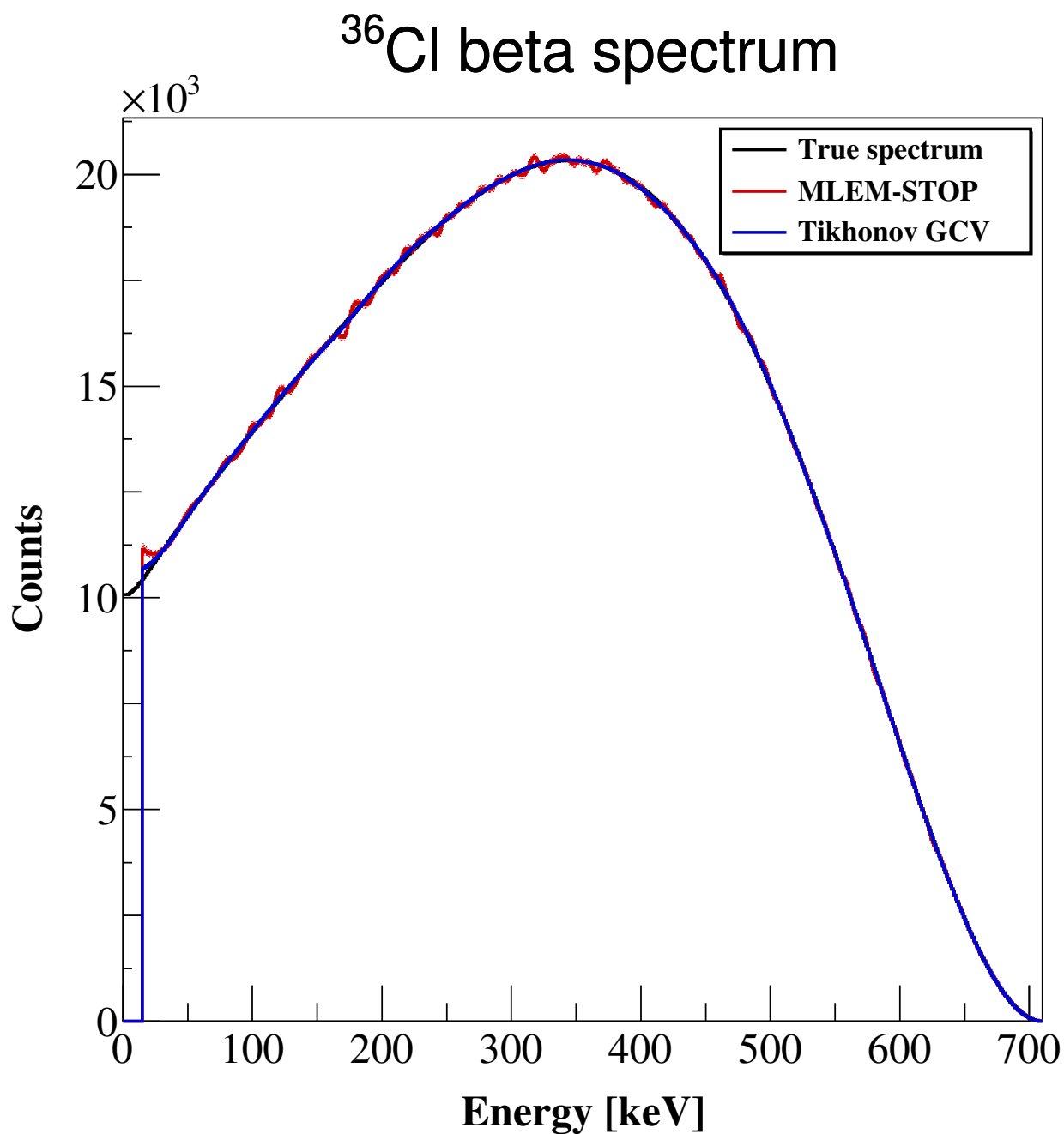


Figure S11: Comparison between the theoretical, true beta spectrum of  $^{36}\text{Cl}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.

Beta source	Counting statistics	$\text{RMSE}_{\min}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\text{stop}}^{\text{MLEM}}$ [reg. param.]	$\text{RMSE}_{\min}^{\text{Tik}}$ [reg. param.]	$\text{RMSE}_{\text{gcv}}^{\text{Tik}}$ [reg. param.]
$^{36}\text{Cl}$	8.7e+04	6.4 [2]	6.4 [3]	2.1 [6.5e+02]	2.1 [6.5e+02]
	2.2e+05	4.3 [3]	4.6 [4]	1.3 [1.1e+02]	1.3 [1.4e+02]
	3.5e+05	3.6 [3]	3.7 [4]	1.1 [5.0e+01]	1.1 [6.5e+01]
	4.8e+05	3.2 [4]	3.2 [4]	1.0 [3.0e+01]	1.0 [3.9e+01]
	6.1e+05	3.0 [4]	3.0 [4]	0.9 [1.4e+01]	0.9 [3.0e+01]
	7.4e+05	2.7 [4]	2.7 [4]	0.8 [1.1e+01]	0.8 [1.8e+01]
	8.7e+05	2.6 [4]	2.6 [4]	0.8 [8.5e+00]	0.8 [1.4e+01]
	2.2e+06	1.8 [5]	1.8 [5]	0.6 [1.4e+00]	0.6 [3.0e+00]
	3.5e+06	1.6 [6]	1.6 [5]	0.6 [6.6e-01]	0.6 [1.4e+00]
	4.8e+06	1.4 [6]	1.4 [6]	0.5 [4.0e-01]	0.5 [6.6e-01]
	6.1e+06	1.3 [7]	1.3 [6]	0.5 [1.8e-01]	0.5 [5.1e-01]
	7.4e+06	1.2 [7]	1.2 [6]	0.5 [1.4e-01]	0.5 [4.0e-01]
	8.6e+06	1.1 [7]	1.1 [6]	0.4 [1.1e-01]	0.5 [2.4e-01]
	2.2e+07	0.8 [9]	0.8 [7]	0.4 [1.9e-02]	0.4 [5.1e-02]
	3.5e+07	0.7 [10]	0.7 [8]	0.4 [6.7e-03]	0.4 [2.4e-02]
	4.8e+07	0.6 [11]	0.6 [8]	0.4 [4.0e-03]	0.4 [1.4e-02]
	6.1e+07	0.6 [12]	0.6 [9]	0.3 [2.4e-03]	0.4 [8.6e-03]
	7.4e+07	0.5 [12]	0.6 [9]	0.3 [1.4e-03]	0.4 [6.7e-03]
	8.6e+07	0.5 [13]	0.5 [9]	0.3 [1.1e-03]	0.3 [4.0e-03]
	2.2e+08	0.4 [16]	0.4 [11]	0.3 [1.9e-04]	0.3 [8.7e-04]
	3.5e+08	0.4 [18]	0.4 [12]	0.3 [6.8e-05]	0.3 [3.1e-04]
	4.8e+08	0.4 [19]	0.4 [13]	0.3 [4.1e-05]	0.3 [1.9e-04]
	6.1e+08	0.3 [20]	0.4 [13]	0.3 [2.4e-05]	0.3 [1.1e-04]
	7.3e+08	0.3 [21]	0.4 [13]	0.3 [1.9e-05]	0.3 [6.8e-05]

Table S11: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for  $^{36}\text{Cl}$  beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.

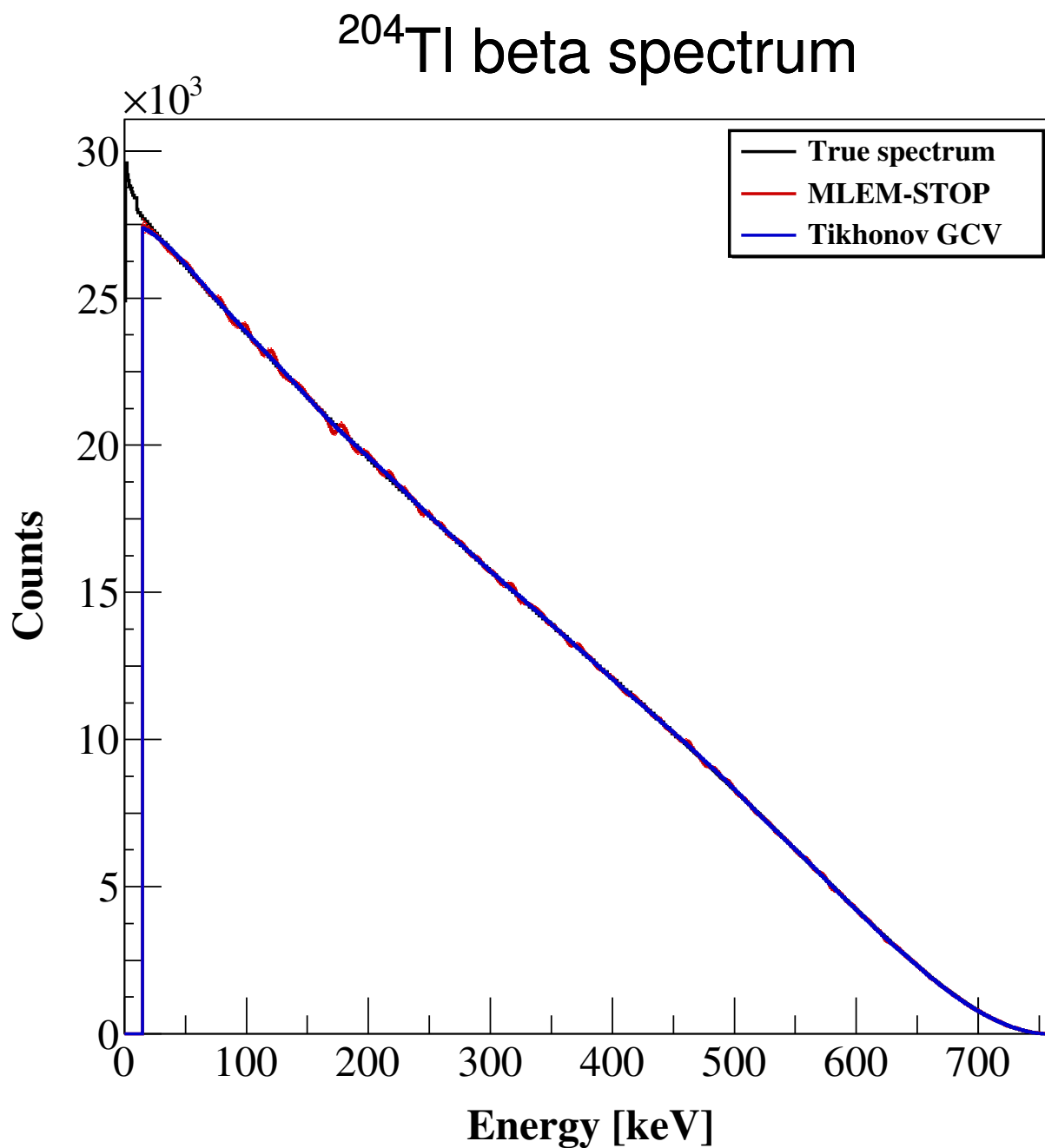


Figure S12: Comparison between the theoretical, true beta spectrum of  $^{204}\text{Tl}$  decay (black) with the MLEM (red) and Tikhonov (blue) unfolded spectra obtained with the MLEM-STOP and the GCV criteria, respectively.



Beta source	Counting statistics	RMSE <sup>MLEM</sup> <sub>min</sub> [reg. param.]	RMSE <sup>MLEM</sup> <sub>stop</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>min</sub> [reg. param.]	RMSE <sup>Tik</sup> <sub>gcv</sub> [reg. param.]
<sup>204</sup> Tl	8.2e+04	6.6 [3]	6.9 [4]	2.2 [6.5e+02]	2.2 [1.1e+03]
	2.1e+05	4.8 [4]	4.8 [4]	1.5 [1.1e+02]	1.6 [3.0e+02]
	3.3e+05	3.6 [4]	3.6 [4]	1.1 [3.0e+01]	1.2 [1.1e+02]
	4.5e+05	3.3 [4]	3.3 [4]	1.1 [1.4e+01]	1.3 [6.5e+01]
	5.7e+05	2.9 [4]	3.0 [5]	1.0 [1.1e+01]	1.1 [3.9e+01]
	7.0e+05	2.7 [4]	2.7 [5]	1.0 [6.5e+00]	1.1 [3.0e+01]
	8.2e+05	2.5 [5]	2.5 [5]	0.9 [5.1e+00]	1.0 [1.8e+01]
	2.0e+06	1.7 [5]	1.7 [5]	0.7 [4.0e-01]	0.8 [3.0e+00]
	3.3e+06	1.4 [6]	1.4 [6]	0.6 [1.4e-01]	0.7 [1.4e+00]
	4.5e+06	1.2 [6]	1.2 [6]	0.5 [6.6e-02]	0.6 [6.6e-01]
	5.7e+06	1.1 [6]	1.1 [6]	0.5 [5.1e-02]	0.6 [5.1e-01]
	7.0e+06	1.0 [6]	1.0 [6]	0.5 [3.1e-02]	0.5 [3.1e-01]
	8.2e+06	1.0 [6]	1.0 [6]	0.4 [2.4e-02]	0.5 [2.4e-01]
	2.0e+07	0.7 [7]	0.7 [7]	0.4 [5.2e-03]	0.4 [2.4e-02]
	3.3e+07	0.6 [8]	0.6 [7]	0.3 [2.4e-03]	0.3 [6.7e-03]
	4.5e+07	0.5 [8]	0.5 [7]	0.3 [1.4e-03]	0.3 [3.1e-03]
	5.7e+07	0.5 [9]	0.5 [8]	0.3 [1.1e-03]	0.3 [1.9e-03]
	7.0e+07	0.4 [9]	0.5 [8]	0.3 [8.7e-04]	0.3 [1.1e-03]
	8.2e+07	0.4 [9]	0.4 [8]	0.3 [6.7e-04]	0.3 [8.7e-04]
	2.0e+08	0.4 [11]	0.4 [9]	0.3 [1.5e-04]	0.3 [1.1e-04]
	3.3e+08	0.3 [12]	0.3 [9]	0.3 [5.3e-05]	0.3 [5.3e-05]
	4.5e+08	0.3 [13]	0.3 [10]	0.3 [3.2e-05]	0.3 [3.2e-05]
	5.7e+08	0.3 [13]	0.3 [10]	0.3 [1.9e-05]	0.3 [1.9e-05]
	7.0e+08	0.3 [14]	0.3 [10]	0.3 [1.5e-05]	0.3 [1.1e-05]

Table S12: Comparison of the four RMSE described in section 3.1 obtained with MLEM and Tikhonov methods on simulated data for <sup>204</sup>Tl beta spectrum. RMSE are normalized by the counting statistics and expressed in pcm. Numbers in brackets are the regularization parameter in each case.