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Monte Carlo benchmark on coincidence summing corrections
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Angular Correlation Effects in Gamma-Ray spectrometry – Coincidence summing corrections

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Outline

1. Introduction
2. Gamma-gamma angular correlations
3. Implementation in coincidence summing software
4. Results
5. Conclusions

Introduction

Recent interest for the sum peak method in the metrology of specific nuclides

- Sum peak method – absolute method for the assessment of activity
- Initially proposed by Brinkman (IJARI 14 (1963) 153)
- Modified sum peak method (Ogata et al, NIMA 775 (2015) 34; Aso et al., ARI 134 (2018) 147) – to avoid the need for the total count rate
- Other applications – correlation method (Suvaila et al, ARI 87 (2014) 384)

– Angular correlations important contribution to the sum peak count rate:

  - Kim et al, ARI 58 (2003) 227
  - Nemes et al, NIMA 918 (2019) 37
  - Vidmar et al, ARI 67 (2009) 160
  - Ilie et al., Rom. Rep. Phys. 71 (2019) 211
  - Hager and Krane, NIMA 976 (2020) 164239

Effect of angular correlations on the measurement of environmental samples:

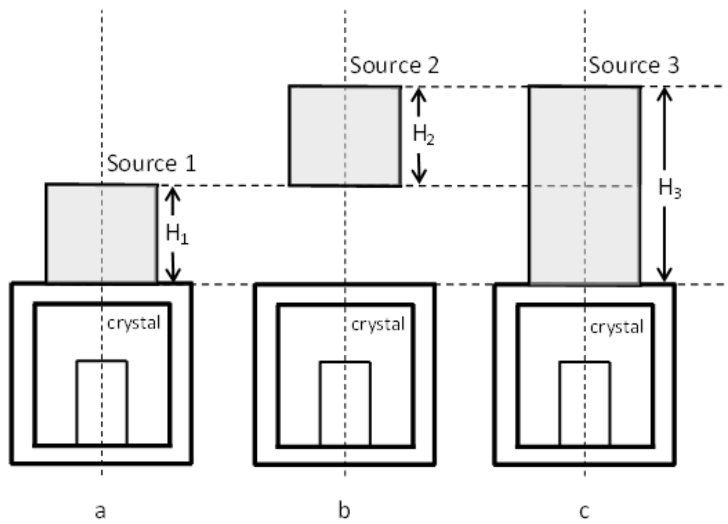
  - Roteta and Garcia-Torano, NIMA 369 (1996) 665

$\Rightarrow$ During the meeting of the GSWG (Salamanca, 2019) an action intended to test the methods applied for the evaluation of the angular correlation effects in the context of coincidence summing corrections was proposed

  $\Rightarrow$ It was postponed, maybe now it can start

Effect of angular correlations. Coincidence summing correction factors FC neglecting angular correlations with respect to the values obtained with angular correlation included (Sima et al., ARI 155 (2020) 108921)

		FC(ISOT)/FC(AC)-1			(1-FC(ISOT))/(1-FC(AC))-1		
Nuclide	E(keV)	Geom a	Geom b	Geom c	Geom a	Geom b	Geom c
Co-60	1173	-0.004	-0.003	-0.004	0.032	0.073	0.037
Co-60	1332	-0.004	-0.003	-0.004	0.032	0.073	0.037
Cs-134	604.7	-0.006	-0.005	-0.006	0.032	0.080	0.038
Cs-134	795.9	-0.007	-0.005	-0.006	0.036	0.084	0.042
Co-60	2506	0.037	0.076	0.042			
Cs-134	1401	0.036	0.075	0.041			



Detector: Crystal: R=3 cm, L=6 cm , RH=0.5 cm, LH=4 cm, Crystal to endcap: DCE=0.5 cm.

Sources: H_1=H_2=2.5 cm, H_3=5cm, R=2 cm; matrix: air (vacuum)

O. Sima et al., ICRM Conference, Salamanca 2019

Gamma-Gamma Angular Correlations

- The emission of a gamma photon in a transition from a nuclear state ($J_i\pi_i$) to a lower energy state ($J_f\pi_f$) – electromagnetic process, due to the dynamics of the charges and currents in the nucleus

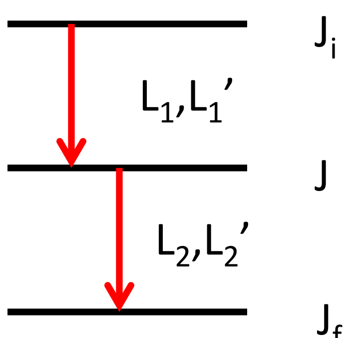
=> conservation of angular momentum => selection rules, emitted photon has a specific multipole (EL or ML), or a mixing of two multipoles (e.g. M1+E2)

$\Rightarrow$ In a transition $J_iM_i\pi_i$ -> $J_fM_f\pi_f$ – the probability of emission is not isotropic

- $\Rightarrow$ Depends on the orientation of the nuclear states (M_i, M_f) and on the multipole order
- $\Rightarrow$ In typical cases the emission from the ensemble of nuclei is isotropic due to the random orientation of nuclear states
- $\Rightarrow$ If a single gamma ray is measured (γ_1 or γ_2) – isotropy
- $\Rightarrow$ If γ_1 emitted by a nucleus and γ_2 emitted by another nucleus are measured, they are uncorrelated and each is isotropic

$\Rightarrow$ If γ_1 and γ_2 are emitted in the same decay act, they are correlated; the probability of detecting both by the same detector is no longer equal to the product of detecting probability of each of them

$\Rightarrow$ Angular correlation of γ_1 and γ_2

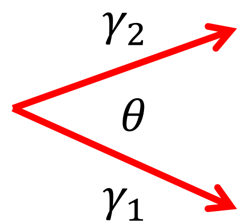


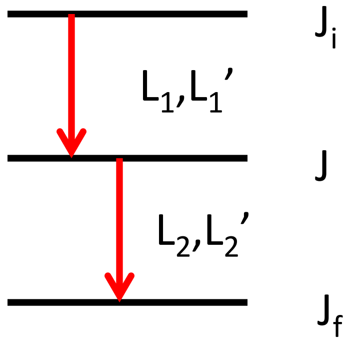
Angular correlations (AC) – well established in nuclear spectroscopy studies

- Assignment of spins by AC, measurement of magnetic dipole moment (PAC)

$$w(\theta) = 1 + \sum_{k=2,4,..} a_k \cdot P_k(\cos(\theta))$$

$$a_k = B_k(1) \cdot A_k(2)$$





Pure transitions (a single value of L in the transition):

$$B_k(1) = F_k(L_1, L_1, J, J_i) \qquad A_k(2) = F_k(L_2, L_2, J, J_f)$$

Mixed transitions (L and L'=L+1 contribute to the transition, δ = mixing ratio):

$$B_k(1) = \frac{(F_k(L_1, L_1, J, J_i) - 2\delta_1 F_k(L_1, L_1', J, J_i) + \delta_1^2 F_k(L_1', L_1', J, J_i))}{1 + \delta_1^2}$$

$$A_k(2) = \frac{\left(F_k\left(L_2, L_2, J, J_f\right) + 2\delta_2 F_k\left(L_2, L_2', J, J_f\right) + \delta_2^2 F_k\left(L_2', L_2', J, J_f\right)\right)}{1 + \delta_2^2}$$

F_k functions depend on the spins of the nuclear states and on the multipole order of the photon = > can be taken from specialized tables or computed using explicit formulas

Mixing ratio δ depends on the nuclear properties of the initial and final nuclear states

- Transition probability depends on δ^2
- Angular distribution of linearly polarized photons depends on δ
- γ-γ angular correlations depend on δ

$\Rightarrow$ Easier to obtain δ^2 from measurement than δ

Data bases: DDEP, ENSDF

- DDEP – values of δ^2 for transitions in specific nuclides

	Energy keV	$P_{\gamma+ce}$ $\times$ 100	Multipolarity
$\gamma_{4,3}$(Ba)	242,746 (6)	0,0262 (34)	(M1+E2)
$\gamma_{5,4}$(Ba)	326,585 (6)	0,0177 (11)	(M1+E2)
$\gamma_{4,2}$(Ba)	475,368 (5)	1,496 (7)	M1+97(92)%E2
$\gamma_{2,1}$(Ba)	563,2457 (36)	8,402 (15)	E2
$\gamma_{5,3}$(Ba)	569,331 (6)	15,512 (21)	M1+7,27(4)%E2
$\gamma_{1,0}$(Ba)	604,7223 (19)	98,21 (8)	E2
$\gamma_{3,1}$(Ba)	795,8677 (44)	85,73 (9)	E2
$\gamma_{5,2}$(Ba)	801,953 (5)	8,720 (16)	E2
$\gamma_{1,0}$(Xe)	847,041 (23)	0,0003 (1)	E2
$\gamma_{4,1}$(Ba)	1038,6137 (44)	0,9930 (33)	M1+33,5(19)%E2

Example: the 1038 keV transition of Cs-134 is a mixed magnetic dipole (M1) and electric quadrupole (E2) with the square of the mixing ratio δ^2=33.5(19)%

Data from LNE-LNHB/CEA Tables de Radionucleides, M. M. Bé, 2012

Web site: http://www.nucleide.org/DDEP_WG/DDEPdata.htm

- ENSDF (Evaluated Nuclear Structure Data File):

E_γ [†]	I_γ [‡&]	E_i(level)	J^π_i	E_f	J^π_f	Mult. [§]	δ
232.6 [#]	<0.0011	1400.591	4^+	1167.970	2^+	[E2]	
242.738 *8*	0.0272 *30*	1643.335	3^+	1400.591	4^+	(M1+E2)	
326.589 *13*	0.0162 *10*	1969.923	4^+	1643.335	3^+	[M1+E2]	
475.365 *2*	1.477 *7*	1643.335	3^+	1167.970	2^+	M1+E2	−6.0 *35*
563.246 *5*	8.338 *14*	1167.970	2^+	604.7230	2^+	M1+E2	−7.4 *9*
569.331 *3*	15.373 *17*	1969.923	4^+	1400.591	4^+	M1+E2	+0.28 *3*
604.721 *2*	97.62 *11*	604.7230	2^+	0.0	0^+	E2	
795.864 *4*	85.46 *6*	1400.591	4^+	604.7230	2^+	E2	
801.953 *4*	8.688 *16*	1969.923	4^+	1167.970	2^+	E2	
1038.610 *7*	0.990 *3*	1643.335	3^+	604.7230	2^+	M1+E2	+0.76 *+10−18*

Example: for the 1038 keV transition δ=+0.76 +10-18
Data source: ENSDF web page:
https://www.nndc.bnl.gov/ensdf/

Attention: if the sign of δ is not explicitly given in ENSDF file, it means that the sign is not known => only δ^2 is in fact known

⇒ The values of the F_k functions evaluated with good accuracy, the spins being generally known for transitions of interest for activity evaluation by gamma spectrometry

⇒ The accuracy of the $B_k(1)$ and $A_k(2)$ coefficients is lower than of F_k in the case of mixed transitions, due to uncertainties in the values of δ

Examples of the coefficients a_k for the cascade J_i (L_1) J (L_2) J_f:

Cascade	Coefficients		
4 (2) 2 (2) 0	a_2=0.10204	a_4=0.00907	a_6=0
2 (1) 1 (1) 0	a_2=0.05	a_4=0	a_6=0
0 (2) 2 (2) 0	a_2=0.35714	a_4=1.14285	a_6=0
2 (2) 0 (2) 2	a_2=0	a_4=0	a_6=0
0 (3) 3 (3) 0	a_2=0.75000	a_4=0.04545	a_6=1.70454

For the transition 1/2 (2) 5/2 (1+2) 7/2 with δ_1=0 and different values for δ_2 the a_k coefficients are:

δ_2=0	a_2= - 0.07143	a_4=0	a_6=0
δ_2=1	a_2= - 0. 49360	a_4= - 0. 03628	a_6=0
δ_2= -1	a_2= 0. 24870	a_4= - 0. 03628	a_6=0

Attention: the definition and sign convention in the formulas above are in accord with ENSDF definition for δ. Alternative definition have been also used, implying different equations for A_k, B_k and a_k.

Effect of angular correlation on detection probability

⇒ The probability of simultaneous detection of the (γ_1, γ_2) photons in a single detector is:

- point source:
  - Both photons completely absorbed in the detector:

$$\frac{1}{(4\pi)^2}\iint \varepsilon(E_1,\overrightarrow{\Omega_1})\cdot\varepsilon(E_2,\overrightarrow{\Omega_2})\cdot w(\overrightarrow{\Omega_1}\cdot\overrightarrow{\Omega_2})\, d\Omega_1\, d\Omega_2$$

  - First photon completely absorbed, the second depositing any energy:

$$\frac{1}{(4\pi)^2}\iint \varepsilon(E_1,\overrightarrow{\Omega_1})\cdot\eta(E_2,\overrightarrow{\Omega_2})\cdot w(\overrightarrow{\Omega_1}\cdot\overrightarrow{\Omega_2})\, \mathrm{d}\Omega_1\mathrm{d}\Omega_2$$

- volume source:

$$\frac{1}{(4\pi)^2 V}\iiint \varepsilon(E_1,\vec{r},\overrightarrow{\Omega_1})\cdot\varepsilon(E_2,\vec{r},\overrightarrow{\Omega_2})\cdot w(\overrightarrow{\Omega_1}\cdot\overrightarrow{\Omega_2})\, dv\, d\Omega_1\mathrm{d}\Omega_2$$

$$\frac{1}{(4\pi)^2 V}\iiint \varepsilon(E_1,\vec{r},\overrightarrow{\Omega_1})\cdot\eta(E_2,\vec{r},\overrightarrow{\Omega_2})\cdot w(\overrightarrow{\Omega_1}\cdot\overrightarrow{\Omega_2})\, dv\, d\Omega_1\mathrm{d}\Omega_2$$

⇒ The angular correlations effects depend not only on the angular correlation function, but also on the efficiencies dependence on the emission point and on the photons propagation direction (ε - peak, η - total efficiency)

⇒ Difficult to test the quality of angular correlation effects on the basis of the values of the coincidence summing correction factors

Implementation in coincidence summing software

Implementation of angular correlations in the software for the evaluation of coincidence summing corrections – case of linked γ_1-γ_2 pair of photons:

1. Evaluate the coefficients F_k, A_k, B_k and a_k
2. Evaluate $w(\theta)$
3. Random selection of the correlated directions of the two photons:

   - Generate θ according to $w(\theta)$; rejection method is normally sufficiently efficient
   - Generate φ uniformly distributed; (θ, φ) correspond to the direction of the second photon in a reference frame with OZ as the direction of the first photon
   - Generate isotropically the direction of the first photon
   - Find the direction of the second photon in the fixed reference frame, by rotation according to the direction of the first photon

   Or,

   - Generate the directions of the two photons uncorrelated and apply a corresponding weight function

Note: More complex expression for $w(\theta)$ for unlinked γ_1-γ_2 photons
More involved simulations for triple angular correlations

Results

- Computations of FC with and without angular correlation included for the following nuclides and peaks (pure sum peaks in red):
  - Ba-133: 160 keV, 383 keV, 437 keV
  - Co-60: 1173 keV, 2505 keV
  - Cs-134: 1038 keV, 1167 keV, 1365 keV, 1400 keV
  - Rh-106: 1133.7 keV
- P type detector with several geometries:
- Point sources at 12 distances from the end cap (0.5 to 10 cm)
- Water source with radius 4.5 cm and height from 0.5 to 4 cm

- Well type detector and cylindrical water source inside the well
- In the case of the well type detector measurements the discrepancy between the values of FC with and without angular correlations negligible (below 1%)
  => high solid angle, the integrated angular correlations effects over almost 4 π solid angle similar to integration of the isotropic distribution

Computations done with GESPECOR:

- Provision for including the angular correlations from GESPECOR version 1
- Analytical calculation of joint emission probabilities => decay data files
- Default simulation: angular correlation neglected
- Inclusion of angular correlation: user input of a2, a4, a6 in the decay data file

Example: 1332 keV peak of ^{60}Co

- Default decay data file:

Peak energy (keV): 1332.492 gamma yield: 0.9998E+00

Number of secondary correlated radiations: 1

1 1173.23 0.99863E+00 0.00000 0.00000 0.00000

0

Number of cases with multiple correlated gammas: 0

Number of sum peak combinations: 0

- File including angular correlations:

Peak energy (keV): 1332.492 gamma yield: 0.9998E+00

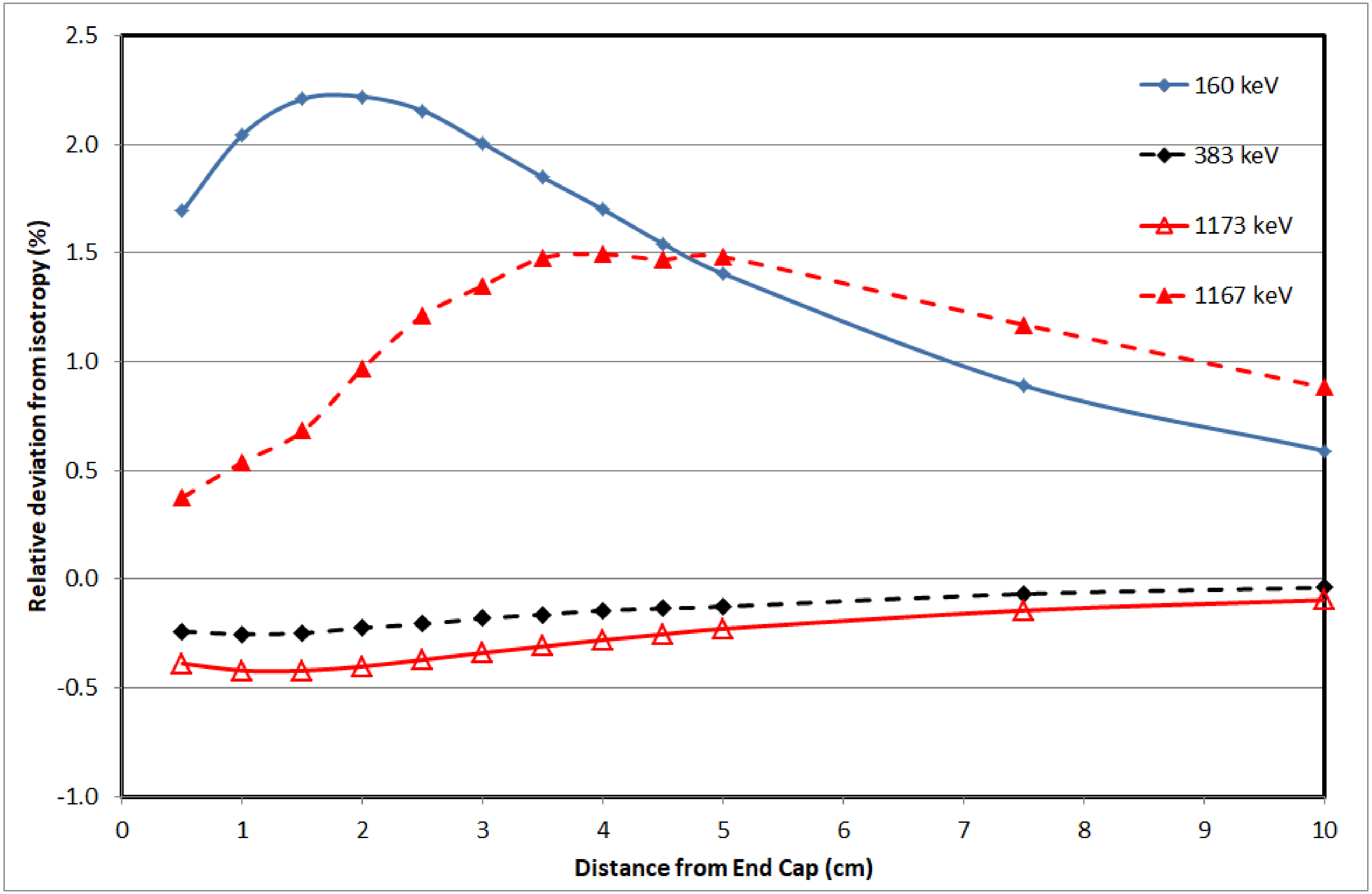
Number of secondary correlated radiations: 1

1 1173.23 0.99863E+00 0.10204 0.00907 0.00000

0

Number of cases with multiple correlated gammas: 0

Number of sum peak combinations: 0



Relative difference FC(AC) vs FC(ISOT) for point sources at several distances

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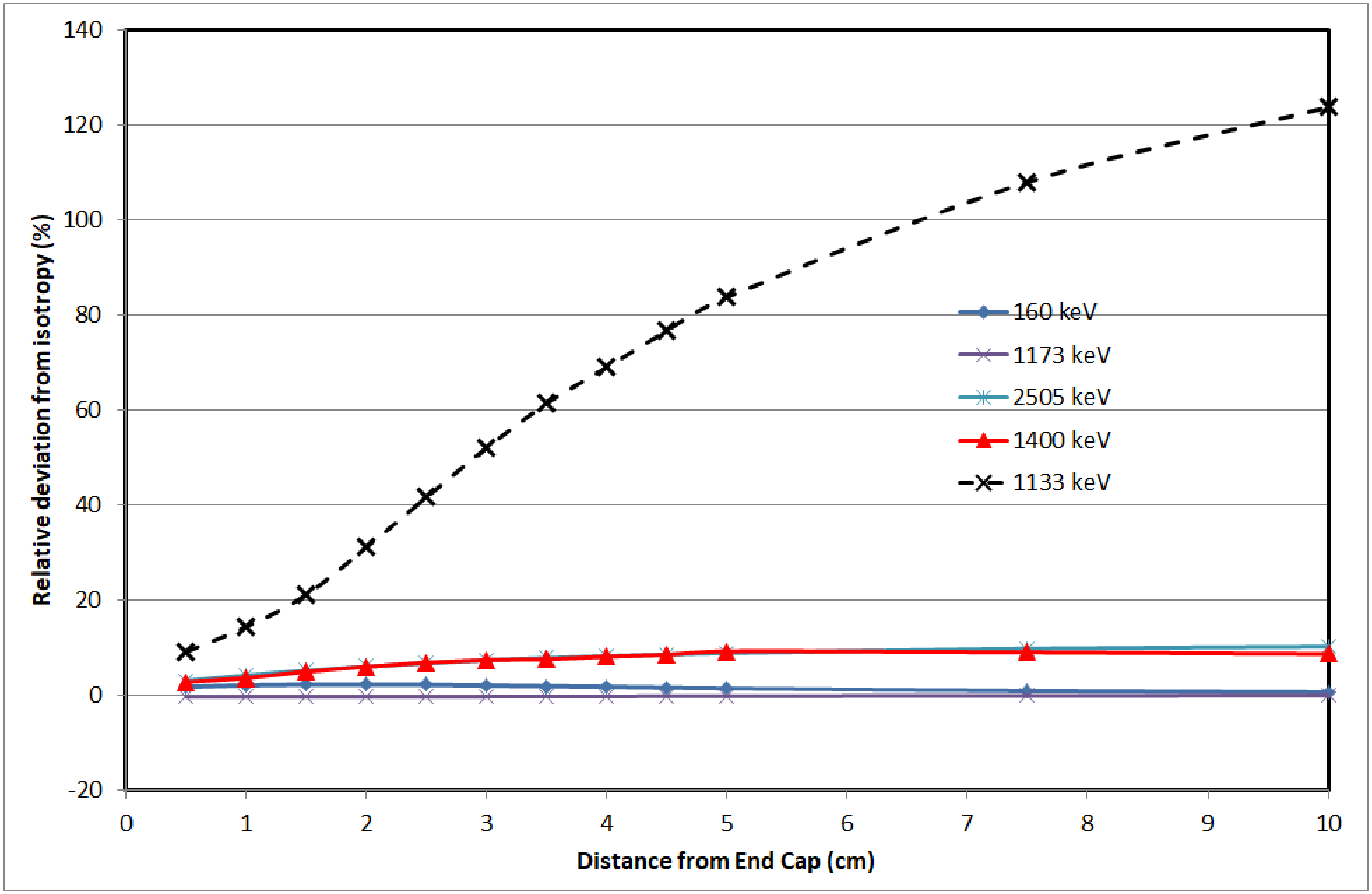
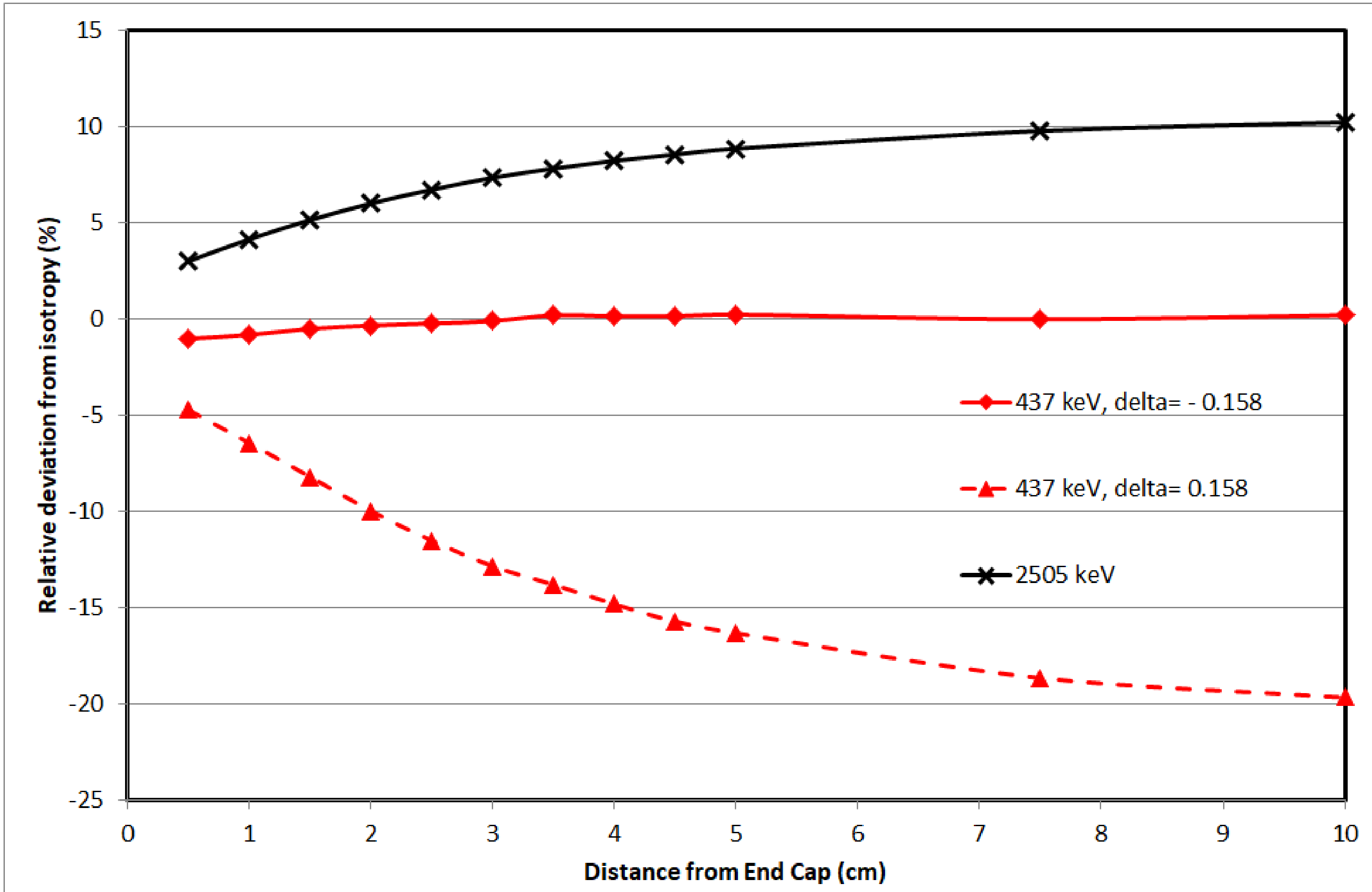
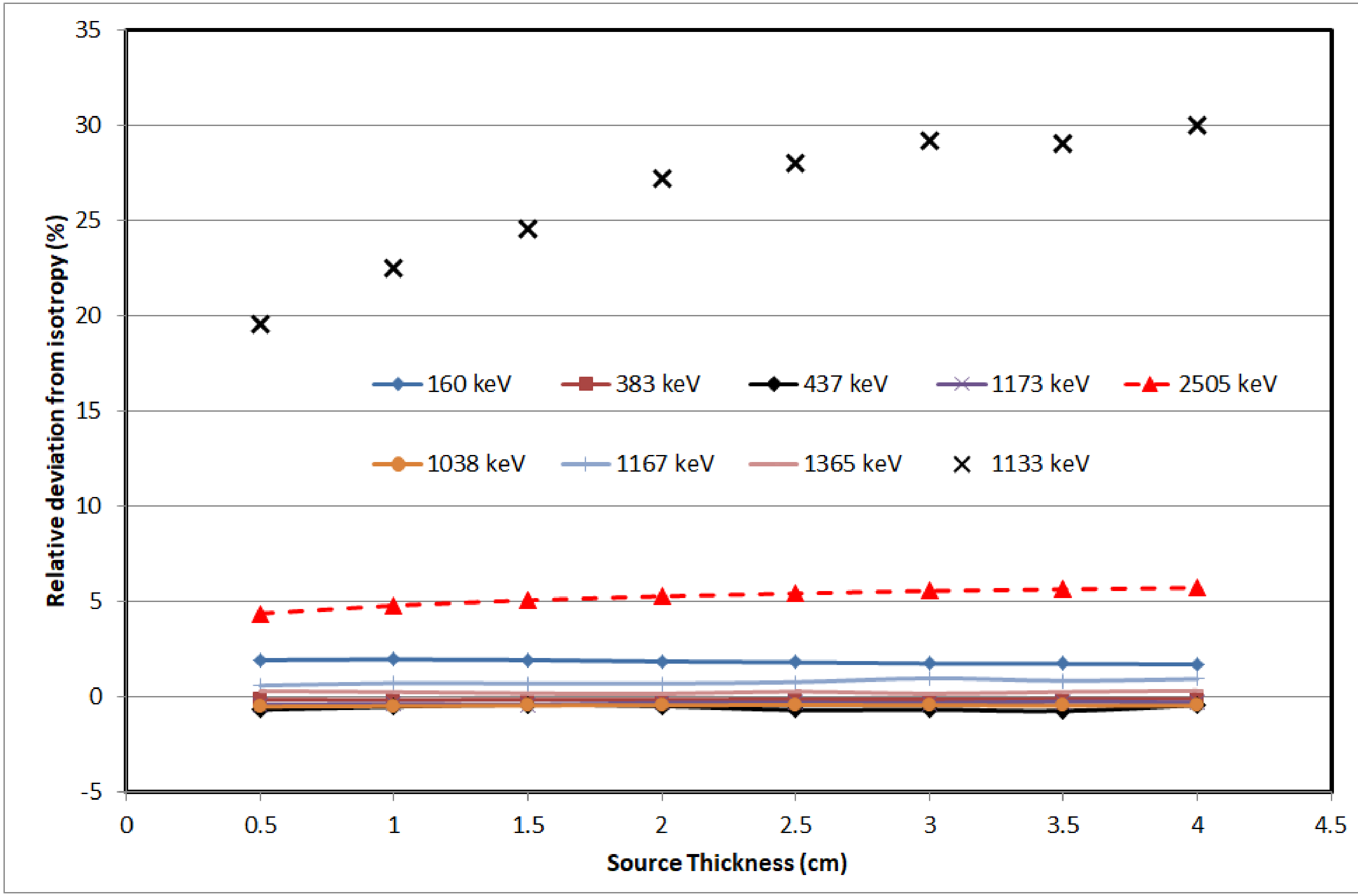


Relative difference FC(AC) vs FC(ISOT) for point sources at several distances
Very high difference for pure sum peaks, especially for 1133.7 keV of Rh-106 (spin sequence 0-2-0)



Relative difference FC(AC) vs FC(ISOT) for point sources – pure sum peaks.
For 437 keV, two values for delta(81keV): -0.158 (sign based on Comments in ENSDF) and 0.158 => high sensitivity to the sign of delta!!



Relative difference FC(AC) vs FC(ISOT) for a cylindrical water sources with several filling heights. Very high difference for the peak of 1133.7 keV of Rh-106

Conclusions

Angular correlations may have a non-negligible contribution to coincidence summing corrections

- significant contributions especially in the case of pure sum peaks
- Important corrections in the application of the sum peak method of activity evaluation

Angular correlations can be implemented in the software for the computation of coincidence summing corrections

An action of the GS working group is proposed to test the capability of different methods to deal with angular correlations