

Cosmogenic activation: recent results

Vitaly A. Kudryavtsev
The University of Sheffield

Outline

- Definitions.
- Production cross-sections.
- Particle spectra.
- Codes.
- Production rates and decay rates: comparison with measurements.
- Summary.

Definitions and equations

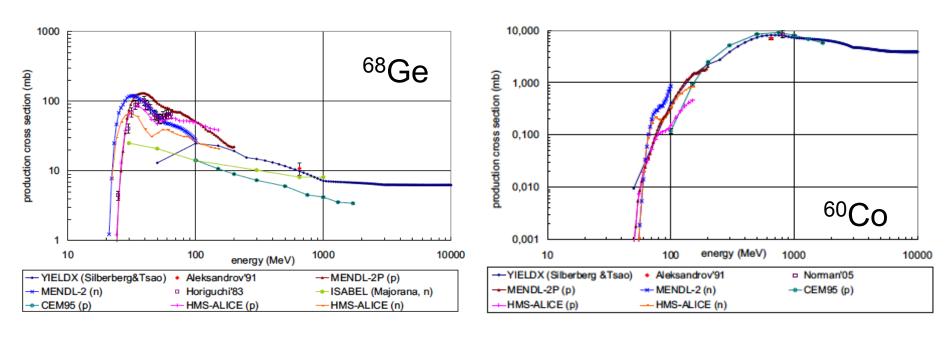
- Low-background experiments are sensitive to the radioactivity induced by activation of materials, primarily by cosmic rays at the surface.
- Decay rate of a radioactive isotope is proportional to the production rate. It also depends on the time of material exposure to the source of radiation, such as cosmic rays (t_{exp}) and on the time the isotopes were allowed to decay without being exposed to cosmic rays, sometimes called cooling time (t_{dec}) .

$$\frac{dN}{dt} = R \left(1 - \exp\left(-\frac{t_{\text{exp}}}{\tau}\right) \right) \exp\left(-\frac{t_{\text{dec}}}{\tau}\right)$$

■ Production rate of radioactive isotopes at the surface of the Earth is determined by the flux of cosmic rays and the cross-section of the production of a particular isotope.

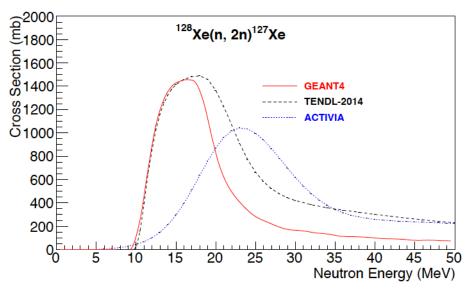
$$R = \int \sigma(E) \frac{dF}{dE} dE$$

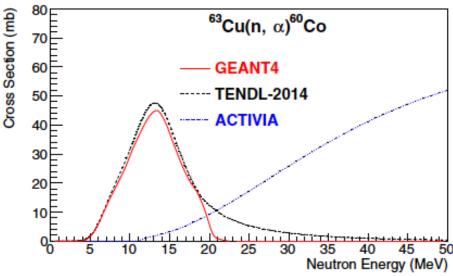
Production cross-sections



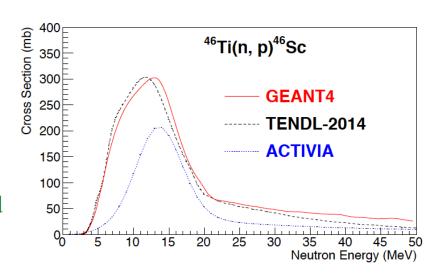
- Production cross-sections (excitation functions) of ⁶⁸Ge and ⁶⁰Co in natural Ge (Cebrian et al. Journal of Physics, Conference series, **39** (2006) 344; Cebrian et al. Astroparticle Physics, **33** (2010) 316; Review by Cebrian at LRT2013).
- Cross-sections:
 - o Silberberg, Tsao and Barghouty. Astrophys. J., **501** (1998) 911; YELDX.
 - o MENDL-2/2P libraries: https://www-nds.iaea.org/publications/iaea-nds/iaea-nds-0136 htm

Production cross-sections

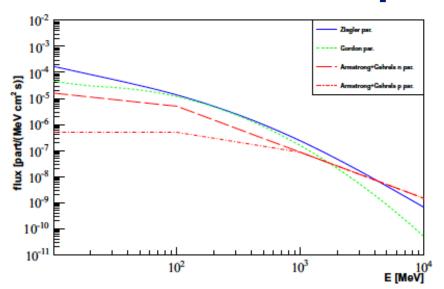


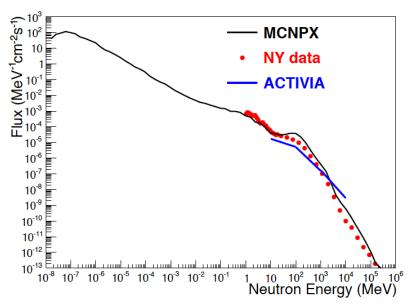


- Evaluated Nuclear Data File ENDF (used in GEANT4, G4NDL).
- Experimental Nuclear Reaction Data, EXFOR.
- TENDL (-2015) output from TALYS.
- From Zhang et al. Astroparticle Physics, **84** (2016) 62.
- ACTIVIA (Silberberg and Tsao): J. J. Back and Y. A. Ramachers, Nucl. Instr. and Meth. A586 (2008) 286.

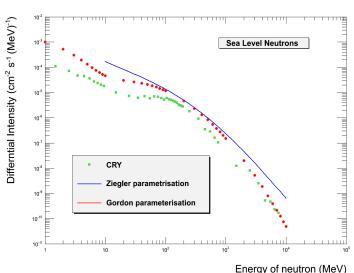


Neutron spectra at sea level

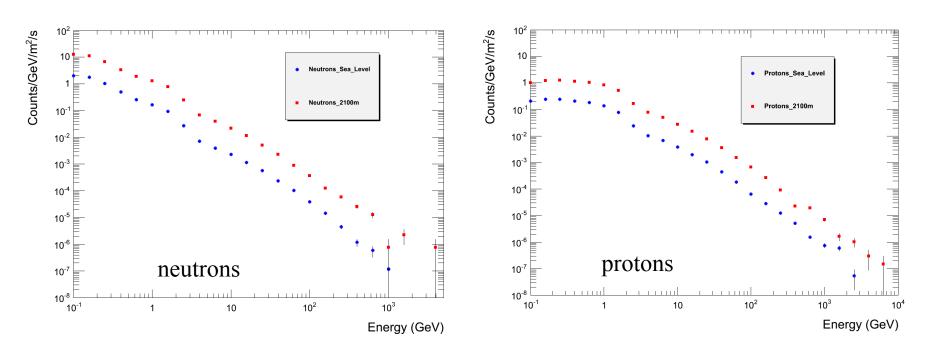




- Parameterisations: Ziegler, IBM J. Res. Develop. 42, 117–140 (1998) and Gordon et al., IEEE Transactions on Nuclear Science 51 3427 (2004).
- Normalised CRY: http://nuclear.llnl.gov/simulation/ main.html (CRY accounts for primary protons only).
- Top left graph from: V. Lozza and J. Petzoldt. Astroparticle Physics, 61 (2015) 62.
- Top right graph from: C. Zhang et al. Astroparticle Physics, **84** (2016) 62.

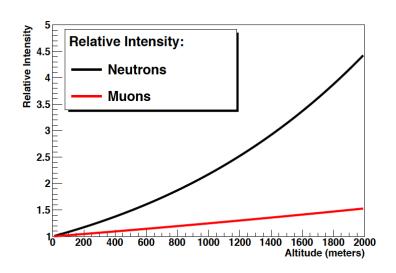


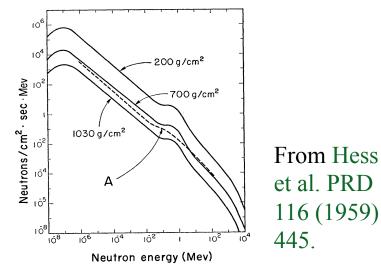
Neutron and proton spectra at different altitudes



- Spectra from CRY. T. Blackwell, PhD thesis (University of Sheffield, 2015).
- CRY accounts for primary protons only (not nuclei), hence underestimate the flux of hadrons by $\sim 70\%$.

Neutron spectra at different altitudes





Correction for altitude (from Ziegler)

$$I_2 = I_1 \exp\left(\frac{A_1 - A_2}{L}\right),\tag{1}$$

where I_1 is the cascade flux at some altitude (pressure) A_1 , and I_2 is the flux at altitude A_2 , both altitudes being expressed in g/cm^2 . To convert terrestrial altitudes to atmospheric pressure, g/cm^2 , we use

$$A = 1033 - (0.03648H) + (4.26 \times 10^{-7}H^2), \tag{2}$$

where H is in feet and A is in g/cm^2 (this assumes an average barometric pressure and a temperature of 0° C). In the lower altitudes, typical absorption lengths are as follows:

Electrons:
$$L_c = 100 \text{ g/cm}^2$$
,

Protons and pions:
$$L_p = 110 \text{ g/cm}^2$$
,

Neutrons:
$$L_n = 148 \text{ g/cm}^2$$
,

Muons:
$$L_{\mu} = 520 \text{ g/cm}^2$$
. (3)

Relative neutron flux calculation: http://seutest.com/cgi-bin/FluxCalculator.cgi

Codes

- COSMO: Martoff and Lewin, Computer Physics Comm. 72 (1992) 96.
- ACTIVIA: Back and Ramachers, Nucl. Instrum. Meth. A 586 (2008)
 286; http://universityofwarwick.github.io/ACTIVIA/
- Also multipurpose codes: GEANT4, FLUKA etc.
- Large variations between the codes.
- A comprehensive review by Cebrian at LRT2013 (LNGS, 2013).
 Also Zhang et al. Astroparticle Physics, 84 (2016) 62.
- COSMO and ACTIVIA use fast neutrons only (E > a few MeV). This approach is quite realistic for activation at the surface of the Earth (both codes deal with neutrons at the surface of the Earth) but ignores thermal neutron capture.

Production rate: Ge

Table 1. Production rates (in kg⁻¹d⁻¹) in natural germanium obtained in this work, considering HMS-ALICE and YIELDX below and above 150 MeV respectively, and in previous estimates. Some results from reference [12] are based on Monte Carlo calculations (MC) while others come from measurements (exp).

	HMS-ALICE+YIELDX	Ref. [10]	Ref. [11]	Ref. [12] (MC)	Ref. [12] (exp)
$^{68}\mathrm{Ge}$	77+12=89	58.4	26.5	29.6	30 ± 7
60 Co	0.3+4.5=4.8	6.6	4.8		
$^{65}\mathrm{Zn}$	36+41=77	79.0	30.0	34.4	38 ± 6
$^{58}\mathrm{Co}$	0.5 + 13 = 14	16.1	4.4	5.3	3.5 ± 0.9
57 Co	0.3 + 9.4 = 9.7	10.2	0.5	4.4	2.9 ± 0.4
$^{54}{ m Mn}$	0.01 + 7.2 = 7.2	9.1		2.7	3.3 ± 0.8
$^{63}\mathrm{Ni}$	1.7 + 3.5 = 5.2	4.6			
55 Fe	0.06+7.9=7.9	8.4			

S. Cebrian. Talk at LRT2013 at LNGS.

Production rate: copper

Isotope, Half life	n(<4 MeV)	n(>4 MeV) (kg ⁻¹ d ⁻¹)	Muon	Proton GEANT4	Total (kg ⁻¹ d ⁻¹)	ACTIVIA1/2 (kg ⁻¹ d ⁻¹)		nang et al. stroparticle
²² Na, 2.6 yr	0	0.012	0.0027	0.002	0.014	0.31/0.19	Pł	nysics, 84
$^{26}_{13}Al$, 7.2 × 10 ⁵ yr	0	0.016	0.0027	0.002	0.021	0.23/0.14		016) 62.
³² Si, 150 yr	0	0.063	0.0027	0.002	0.068	0.13/0.092	(2	010) 02.
$^{40}_{19}K$, 1.3 \times 10 9 yr	0	0.48	0.022	0.04	0.54	1.82/1.75		
⁴⁷ ₂₀ Ca, 4.5 d	0	0.12	0.0	0.01	0.13	0.026/0.036		
⁴⁶ ₂₁ Sc, 83.8 d	0	1.05	0.024	0.12	1.19	3.13/4.09	4.6±1.6	
⁴⁷ ₂₁ Sc, 3.3 d	0	0.98	0.011	0.10	1.09	0.62/0.86		
⁴⁴ ₂₂ Ti, 63 yr	0	1.72	0.05	0.26	2.02	0.16/0.19		Laubenstein
$^{50}_{23}V$, 1.4 \times 10 ¹⁷ yr	0	3.32	0.03	0.26	3.60	4.43/7.43		and Heusser.
⁵¹ ₂₄ Cr, 27.7 d	0	15.20	0.12	1.16	16.48	10.00/18.08		
⁵⁴ ₂₅ Mn, 312.3 d	0	11.68	0.08	0.55	12.31	14.32/30.00	19 ± 2	Applied
⁵⁵ ₂₆ Fe, 2.7 yr	0	53.66	0.25	2.43	56.33	19.32/42.79		Radioactive
⁵⁹ Fe, 44.5 d	0	8.56	0.04	0.18	8.77	4.24/10.49	39±10	Isotopes, 67
$^{60}_{26}Fe$, 1.5 × 10 ⁶ yr	0.00	4.90	0.03	0.10	5.03	0.80/1.98		(2009) 750.
⁵⁶ Co, 77.3 d	0	9.71	0.08	0.54	10.32	8.74/20.13	20 ± 3	
⁵⁷ ₂₇ Co, 271.8 d	0	64.33	0.26	2.55	67.15	32.44/77.45	156 ± 35	
58Co, 70.9 d	0	55.52	0.17	1.57	57.26	56.61/138.06	143 ± 8	
⁶⁰ Co, 5.3 yr	0.02	63.12	0.24	1.25	64.63	26.28/66.12	181±16	
⁶⁵ Zn, 244.3 d	0	1.80	0.02	0.22	2.04	19.58/62.78		

Activation of copper: codes against data

Activated	Geant4	ACTIVIA1/2	Cosmo	TALYS [20]	Baudis et al. data [19]
Isotope	$(\mu \mathrm{Bq/kg})$				
$^{46}\mathrm{Sc}$	12.4	36.2/47.4	17.0	-	27^{+11}_{-9}
$^{54}\mathrm{Mn}$	136.0	165.7/347.2	156	188	154^{+35}_{-34}
⁵⁹ Fe	99.5	49.1/121.5	50	-	47^{+16}_{-14}
⁵⁶ Co	113.2	101.2/233	81	-	108^{+14}_{-16}
⁵⁷ Co	747.7	375.5/896.4	350	650	519_{-95}^{100}
⁵⁸ Co	644.4	655.2/1597.8	632	-	798^{+62}_{-58}
⁶⁰ Co	713.2	295.7/744	297	537	340^{+82}_{-68}

• Data: Baudis et al., EPJ C 75 (2015) 485.

Production and decay rates: Xe

Table 6Decay rates of two xenon isotopes as calculated with Geant4 and ACTIVIA, and measured by LUX [18,43] after 90 days cooling down underground.

Activated isotope	Target	This work (µ Bq/kg) GEANT4	ACTIVIA1/2 $(\mu \mathrm{Bq/kg})$	LUX data[18] (μ Bq/kg)
¹²⁷ Xe	Xe	470	73/180	(490 ± 95)
¹³³ Xe	Xe	7.0×10^{-3}	$9.8\times 10^{-4}/2.4\times 10^{-3}$	$(25.0 \pm 5.0) \times 10^{-3}$

Table 7
Saturation activity at sea level assuming infinite exposure time.

Activated isotope	Target	This work $(\mu \mathrm{Bq/kg})$	ACTIVIA1/2 $(\mu \mathrm{Bq/kg})$	Baudis et al. data [19] $(\mu~{ m Bq/kg})$
¹¹³ Sn	Xe	67.6	45.7/61.3	<55
¹²⁵ Sb	Xe	16.8	0.2/0.01	590^{+260}_{-230}
¹²⁷ Xe	Xe	2670	413/1040	1870^{+290}_{-270}

Zhang et al.
Astroparticle
Physics, 84
(2016) 62.
Data from LUX:
Akerib et al.
Astroparticle
Physics 62
(2015) 33.

Data: Baudis et al., EPJ C 75 (2015) 485.

Measurements: Ge

Table 7

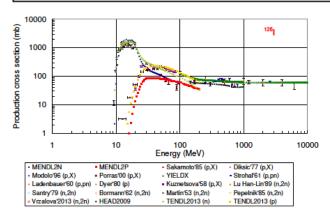
Rates of production (expressed in kg^{-1} . day^{-1}) of isotopes induced in natural germanium as measured in EDELWEISS-III germanium detectors, compared with previous estimates and measurements in Refs. [6,19,28,29,33] and [5]. Errors on the production rate include statistical and for 3H and 65 Zn systematic uncertainties, too. Systematic uncertainty is based on the minimization of the reduced χ^2 . Estimate in this work refers to ACTIVIA calculation, considering semi-empirical [21–25] (a) and MENDL-2P database [26] (b) cross sections. For 49 V, both calculations give the same result. An upper limit for 3H from IGEX data (E) is shown together with calculations [6] for all isotopes. (I) and (II) refer to GEANT4 and ACTIVIA calculations from [19]. The lower limit for 68 Ge at saturation value is listed. It is derived from the fit value of 81 \pm 6 at a 90% C.L. ACTIVIA calculations for 68 Ge are also given including a potential 10-h flight of Ge powder (a* and b*). The last two columns refer to estimates from model [30] and experimental data (Exp.) from Ref. [5].

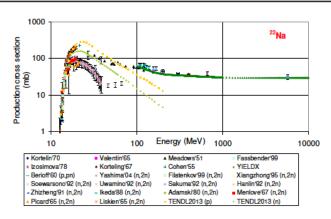
	This work		Cebrian [2	8]	Barabanov	Mei	Zhang	Klapdor	Avigno	one [5]
	Exp.	Calc.	(Ziegler)	(Gordon)	[33]	[6]	[19]	[29]	[30]	Exp.
³H	82± 21	46(a) 43.5(b)				27.7 <21(E)	48.3 (I) 52.4 (II)		210	
49 V	2.8 ± 0.6	1.9 (a,b)					()			
⁶⁵ Zn	106± 13	38.7(a) 65.8(b)	77	63		37.1		79	34.4	38± 6
55Fe	4.6± 0.7	3.5(a) 4.0(b)	8.0	6.0		8.6		8.4		
⁶⁸ Ge	>71	23.1(a) 36.2(a*) 45.0(b) 97.6(b*)	89	60	81.6	41.3		58.4	29.6	30± 7

Armengaud et al. Astroparticle Phys. 91 (2017) 51. Data from EDELWEISS-III.

Measurements: Nal

Isotope	Excitation function	Calculated rate		Experimental rate	Cal/Exp
	LE+HE	LE+HE	total		
¹²⁶ I	MENDL-2+YIELDX	250.0+47.0	297.0	283±36	1.1
^{125}I	TENDL-2013+HEAD-2009	230.2 + 12.1	242.3	220 ± 10	1.1
^{124}I	MENDL-2+HEAD-2009	113.6 + 22.3	135.9		
$^{127m}\mathrm{Te}$	TENDL-2013+extrapolation	6.9+0.2	7.1	10.2 ± 0.4	0.7
$^{125m}\mathrm{Te}$	TENDL-2013+HEAD-2009	38.4 + 3.5	41.9	28.2 ± 1.3	1.5
$^{123m}\mathrm{Te}$	TENDL-2013+HEAD-2009	29.5 + 3.7	33.2	31.6 ± 1.1	1.1
$^{123}\mathrm{Te}$	TENDL-2013+HEAD-2009	8.8+1.4	10.2		
$^{121m}\mathrm{Te}$	TENDL-2013+HEAD-2009	19.1 + 4.7	23.8	$23.5 {\pm} 0.8$	1.0
$^{121}\mathrm{Te}$	TENDL-2013+YIELDX	5.8+2.6	8.4	9.9 ± 3.7	0.8
²² Na	TENDL-2013+YIELDX	43.2 + 10.4	53.6	45.1 ± 1.9	1.2





Data from J. Amare et al. JCAP 02 (2015) 046.

Production of tritium

Target	n(<4 MeV)	$n(>4~{ m MeV})$	μ	Total	ACTIVIA1/2	TALYS	ANAIS-25
Target		$(kg^{-1}day^{-1})$ GEANT4		$(\mathrm{kg^{-1}day^{-1}})$	$(\mathrm{kg^{-1}day^{-1}})$	$(\mathrm{kg^{-1}day^{-1}})$	$(\mathrm{kg^{-1}day^{-1}})$
C_2H_6	0	279.1	0.374	279.5	-/-	-	-
Si	0	27.12	0.1668	27.29	56.54/108.74	-	-
Argon	0	84.49	0.4135	84.91	45.58/82.9	44.4	-
Ge	0	48.21	0.1127	48.32	34.13/52.39	27.7	82±21 (EDW3)
NaI	0	42.6	0.2629	42.87	31.9/36.19	31.1	40.6
Xenon	0	32.13	0.6348	32.76	31.14/35.63	16.0	-

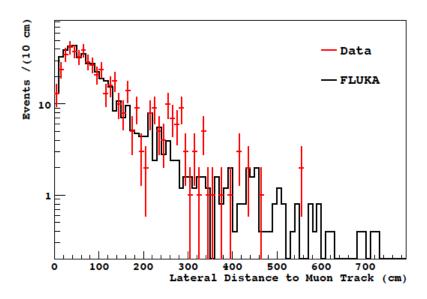
- ANAIS data: Amare et al., AIP Conf. Proc. 1672, 140001 (2015).
- Calculations with TALYS from: Mei et al. Astropart. Phys. 31 (2009) 417-420.
- Also recent measurements and calculations of tritium in EDELWEISS-III and SuperCDMS.

Other recent measurements

- R. Strauss et al. JCAP 06 (2015) 030. Background in CRESST-II including activation.
- B. S. Wang et al. PRC 92 (2015) 024620. Irradiation of Te with high-energy neutrons and measurements of activation.
- V. E. Guiseppe et al. Astroparticle Physics 64 (2015) 34. Measurements of activation of lead using fast neutrons at LANSCE.
- **...**

Activation underground: Borexino and Kamland

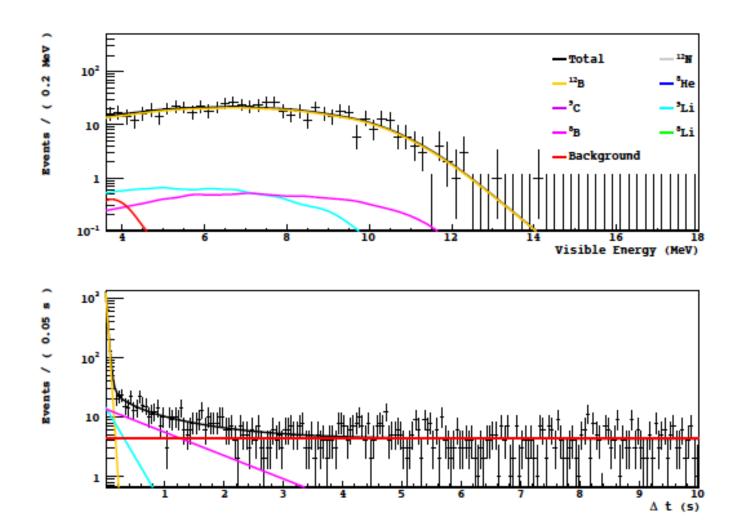
Cosmogenic	Lifetime	Q-Value	Decay	Cosmogenic	Lifetime	Q-Value	Decay
Isotope		[MeV]	Type	Isotope		[MeV]	Type
^{12}N	$15.9\mathrm{ms}$	17.3	β^-	$^6\mathrm{He}$	$1.16\mathrm{s}$	3.51	β^-
^{12}B	$29.1\mathrm{ms}$	13.4	β^+	$^{8}\mathrm{Li}$	$1.21\mathrm{s}$	16.0	β^-
8 He	$171.7\mathrm{ms}$	10.7	β^-	11 Be	$19.9\mathrm{s}$	11.5	β^-
⁹ C	$182.5\mathrm{ms}$	16.5	β^+	^{10}C	$27.8\mathrm{s}$	3.65	β^+
9 Li	$257.2\mathrm{ms}$	13.6	β^-	¹¹ C	$29.4 \mathrm{min}$	1.98	β^+
^{8}B	$1.11\mathrm{s}$	18.0	β^+				



Looking at a certain time windows and energy ranges after the muon trigger. Event should not be far from the muon track.

From Borexino Collaboration, JCAP 1308 (2013) 049, arXiv:1304.7381.

Borexino



Activation underground: results

	GEANT4	GEANT4	FLUKA	Borexino	KamLAND		
	Model III	Model IV					
		$-\langle E_{\mu} \rangle = 289$	$9 \pm 19 \mathrm{GeV}$ —		$\langle E_{\mu} \rangle = 260 \pm 8 GeV$		
Isotopes		Y	ield $[10^{-7} (\mu)]$	$g/cm^2)^{-1}$			
$^{12}{ m N}$	1.11 ± 0.13	3.0 ± 0.2	0.5 ± 0.2	< 1.1	1.8 ± 0.4		
$^{12}{ m B}$	30.1 ± 0.7	29.7 ± 0.7	28.8 ± 1.9	56 ± 3	42.9 ± 3.3		
$^8{ m He}$	< 0.04	0.18 ± 0.05	0.30 ± 0.15	< 1.5	0.7 ± 0.4		
$^9{f Li}$	0.6 ± 0.1	1.68 ± 0.16	3.1 ± 0.4	2.9 ± 0.3	2.2 ± 0.2		
$^8{ m B}$	0.52 ± 0.09	1.44 ± 0.15	6.6 ± 0.6	14 ± 6	8.4 ± 2.4		
$^6{ m He}$	18.5 ± 0.5	8.9 ± 0.4	17.3 ± 1.1	38 ± 15	not reported		
$^8{ m Li}$	27.7 ± 0.7	7.8 ± 0.4	28.8 ± 1.0	7 ± 7	12.2 ± 2.6		
${}^9{f C}$	0.16 ± 0.05	0.99 ± 0.13	0.91 ± 0.10	< 16	3.0 ± 1.2		
$^{11}{ m Be}$	0.24 ± 0.06	0.45 ± 0.09	0.59 ± 0.12	< 7.0	1.1 ± 0.2		
$^{10}{ m C}$	15.0 ± 0.5	41.1 ± 0.8	14.1 ± 0.7	18 ± 5	16.5 ± 1.9		
^{11}C	315 ± 2	415 ± 3	467 ± 23	886 ± 115	866 ± 153		
Neutrons	Yield $[10^{-4} (\mu \text{g/cm}^2)^{-1}]$						
	3.01 ± 0.05	2.99 ± 0.03	2.46 ± 0.12	3.10 ± 0.11	2.79 ± 0.31		

Summary

- Cosmogenic activation of materials becomes more or more important with the increase of detector sensitivity. We may not have full (and accurate) knowledge of this yet.
- Several codes exist to calculate production rates but they do not provide consistent results.
- Neutron spectra and excitation functions (cross-sections) are important for calculating production rates of radio-isotopes and they are not well known.
- Measurements should provide key figures, also for tuning the codes but require knowledge of the material (exposure) history.
- There is no consistent picture of what code is better: different codes tend to better match data from different targets.
- We all hope to achieve progress here for better assessing the requirements for future experiments.
- Activation by calibration sources?