

Shedding light on the origin of ^{204}Pb , the heaviest s-only isotope in the solar system: Supplemental Material

A. Casanovas-Hoste,^{1,2,3} C. Domingo-Pardo,² J. Lerendegui-Marco,⁴ C. Guerrero,⁴ A. Tarifeño-Saldivia,² M. Krτίčka,⁵ M. Pignatari,^{6,7,8,9} F. Calviño,¹ D. Schumann,¹⁰ S. Heintz,¹⁰ R. Dressler,¹⁰ U. Köster,¹¹ O. Aberle,³ J. Andrzejewski,¹² L. Audouin,¹³ V. Bécarea,¹⁴ M. Bacak,¹⁵ J. Balibrea-Correa,¹⁴ M. Barbagallo,¹⁶ S. Barros,¹⁷ F. Bečvář,⁵ C. Beinrucker,¹⁸ E. Berthoumieux,¹⁹ J. Billowes,²⁰ D. Bosnar,²¹ M. Brugger,³ M. Caamaño,²² M. Calviani,³ D. Cano-Ott,¹⁴ R. Cardella,³ D. M. Castelluccio,^{23,24} F. Cerutti,³ Y. H. Chen,¹³ E. Chiaveri,³ N. Colonna,¹⁶ G. Cortés,¹ M. A. Cortés-Giraldo,⁴ L. Cosentino,²⁵ L. A. Damone,^{16,26} M. Diakaki,¹⁹ E. Dupont,¹⁹ I. Durán,²² B. Fernández-Domínguez,²² A. Ferrari,³ P. Ferreira,¹⁷ P. Finocchiaro,²⁵ V. Furman,²⁷ K. Göbel,¹⁸ A. R. García,¹⁴ A. Gawlik-Ramiega,¹² T. Glodariu [†],²⁸ I. F. Gonçalves,¹⁷ E. González-Romero,¹⁴ A. Goverdovski,²⁹ E. Griesmayer,¹⁵ F. Gunsing,^{19,3} H. Harada,³⁰ T. Heftrich,¹⁸ J. Heyse,³¹ D. G. Jenkins,³² E. Jericha,¹⁵ F. Käppeler [†],³³ Y. Kadi,³ T. Katabuchi,³⁴ P. Kavargin,¹⁵ V. Ketlerov,²⁹ V. Khryachkov,²⁹ A. Kimura,³⁰ N. Kivel,¹⁰ M. Kokkoris,³⁵ E. Leal-Cidoncha,²² C. Lederer-Woods,³⁶ H. Leeb,¹⁵ S. Lo Meo,^{23,24} S. J. Lonsdale,³⁶ R. Losito,³ D. Macina,³ J. Marganec,¹² T. Martínez,¹⁴ C. Massimi,^{24,37} P. Mastinu,³⁸ M. Mastromarco,¹⁶ F. Matteucci,^{39,40} E. A. Maugeri,¹⁰ E. Mendoza,¹⁴ A. Mengoni,²³ P. M. Milazzo,³⁹ F. Mingrone,²⁴ M. Mirea [†],²⁸ S. Montesano,³ A. Musumarra,^{25,41} R. Nolte,⁴² A. Oprea,²⁸ N. Patronis,⁴³ A. Pavlik,⁴⁴ J. Perkowski,¹² I. Porras,^{3,45} J. Praena,^{4,45} J. M. Quesada,⁴ K. Rajeev,⁴⁶ T. Rauscher,^{47,48} R. Reifarth,¹⁸ A. Riego-Perez,⁴⁹ Y. Romanets,¹⁷ P. C. Rout,⁴⁶ C. Rubbia,³ J. A. Ryan,²⁰ M. Sabaté-Gilarte,^{3,4} A. Saxena,⁴⁶ P. Schillebeeckx,³¹ S. Schmidt,¹⁸ P. Sedyshev,²⁷ A. G. Smith,²⁰ A. Stamatopoulos,³⁵ G. Tagliente,¹⁶ J. L. Tain,² L. Tassan-Got,¹³ A. Tsinganis,³⁵ S. Valenta,⁵ G. Vannini,^{24,37} V. Variale,¹⁶ P. Vaz,¹⁷ A. Ventura,²⁴ V. Vlachoudis,³ R. Vlastou,³⁵ A. Wallner,⁵⁰ S. Warren,²⁰ M. Weigand,¹⁸ C. Weiss,^{3,15} C. Wolf,¹⁸ P. J. Woods,³⁶ T. Wright,²⁰ and P. Žugec^{21,3}

(The n_TOF Collaboration)

¹ *Institut de Tècniques Energètiques (INTE) - Universitat Politècnica de Catalunya, Spain*

² *Instituto de Física Corpuscular, CSIC - Universidad de Valencia, Spain*

³ *European Organization for Nuclear Research (CERN), Switzerland*

⁴ *Universidad de Sevilla, Spain*

⁵ *Charles University, Prague, Czech Republic*

⁶ *Konkoly Observatory, Research Centre for Astronomy and Earth Sciences (CSFK),*

ELKH, H-1121, Budapest, Konkoly Thege M. út 15-17., Hungary

⁷ *CSFK, MTA Centre of Excellence, Budapest, Konkoly Thege Miklós út 15-17., H-1121, Hungary*

⁸ *E. A. Milne Centre for Astrophysics, University of Hull, United Kingdom*

⁹ *NuGrid Collaboration, <http://nugridstars.org>*

¹⁰ *Paul Scherrer Institut (PSI), Villigen, Switzerland*

¹¹ *Institut Laue-Langevin (ILL), Grenoble, France*

¹² *University of Lodz, Poland*

¹³ *Institut de Physique Nucléaire, CNRS-IN2P3, Univ. Paris-Sud,*

Université Paris-Saclay, F-91406 Orsay Cedex, France

¹⁴ *Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT), Spain*

¹⁵ *TU Wien, Atominstytut, Stadionallee 2, 1020 Wien, Austria*

¹⁶ *Istituto Nazionale di Fisica Nucleare, Sezione di Bari, Italy*

¹⁷ *Instituto Superior Técnico, Lisbon, Portugal*

¹⁸ *Goethe University Frankfurt, Germany*

¹⁹ *CEA Irfu, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France*

²⁰ *University of Manchester, United Kingdom*

²¹ *Department of Physics, Faculty of Science, University of Zagreb, Zagreb, Croatia*

²² *University of Santiago de Compostela, Spain*

²³ *Agenzia nazionale per le nuove tecnologie (ENEA), Bologna, Italy*

²⁴ *Istituto Nazionale di Fisica Nucleare, Sezione di Bologna, Italy*

²⁵ *INFN Laboratori Nazionali del Sud, Catania, Italy*

²⁶ *Dipartimento Interateneo di Fisica, Università degli Studi di Bari, Italy*

²⁷ *Affiliated with an institute (or an international laboratory) covered by a cooperation agreement with CERN.*

²⁸ *Horia Hulubei National Institute of Physics and Nuclear Engineering, Romania*

²⁹ *Institute of Physics and Power Engineering (IPPE), Obninsk, Russia*

³⁰ *Japan Atomic Energy Agency (JAEA), Tokai-Mura, Japan*

³¹ *European Commission, Joint Research Centre (JRC), Geel, Retieseweg 111, B-2440 Geel, Belgium*

³² *University of York, United Kingdom*

³³ *Karlsruhe Institute of Technology, Campus North, IKP, 76021 Karlsruhe, Germany*

- ³⁴ *Tokyo Institute of Technology, Japan*
³⁵ *National Technical University of Athens, Greece*
³⁶ *School of Physics and Astronomy, University of Edinburgh, United Kingdom*
³⁷ *Dipartimento di Fisica e Astronomia, Università di Bologna, Italy*
³⁸ *Istituto Nazionale di Fisica Nucleare, Sezione di Legnaro, Italy*
³⁹ *Istituto Nazionale di Fisica Nucleare, Sezione di Trieste, Italy*
⁴⁰ *Dipartimento di Astronomia, Università di Trieste, Italy*
⁴¹ *Dipartimento di Fisica e Astronomia, Università di Catania, Italy*
⁴² *Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany*
⁴³ *University of Ioannina, Greece*
⁴⁴ *University of Vienna, Faculty of Physics, Vienna, Austria*
⁴⁵ *University of Granada, Spain*
⁴⁶ *Bhabha Atomic Research Centre (BARC), India*
⁴⁷ *Centre for Astrophysics Research, University of Hertfordshire, United Kingdom*
⁴⁸ *Department of Physics, University of Basel, Switzerland*
⁴⁹ *Universitat Politècnica de Catalunya, Spain*
⁵⁰ *Australian National University, Canberra, Australia*

Summary of systematic uncertainties

In the main text a 12% total systematic uncertainty (σ_{sys}) in the experimental determination of the $^{204}\text{Tl}(n, \gamma)$ yield is stated. The main sources of uncertainty are reported in Table I. Specific details on the determination of other uncertainties not directly quoted from a reference can be found in [1].

The uncertainty in the ^{204}Tl enrichment was evaluated with the aid of an R-matrix analysis. First, the concentration of the daughter ^{204}Pb was adjusted to the strongest resonance of ^{204}Pb and the corresponding resonance parameters from Ref. [2]. The uncertainty was then obtained by performing a sensitivity test around the adjusted value. Both the adjusted concentration and its uncertainty are in agreement with an independent estimate based on the knowledge of the neutron flux at ILL and the effective irradiation time.

TABLE I. Main individual contributions to the systematic uncertainty in the determination of the $^{204}\text{Tl}(n, \gamma)$ yield.

Source	σ_{sys} (%)
^{204}Tl sample normalization	7
^{203}Tl resonances area (A_r)	6
^{204}Tl enrichment	5
Detector calibration & gain drift	3
Pulse Height Weighting Technique	2 [3]
Neutron flux in the 0.1 – 10 keV E_n range	2 [4]
Background subtraction	2
Threshold correction factors, f_{th}	1.5
Total	12

Semi-empirical procedure for the determination of the MACS at neutron energies relevant for nucleosynthesis studies

In p.4 of the main text, the procedure to extend the MACS of $^{204}\text{Tl}(n, \gamma)$ up to the range of energies relevant for nucleosynthesis studies is briefly described. A more detailed description of the method follows here. First, a threshold in resonance kernel K_r (with $K_r = gJ \frac{\Gamma_n \Gamma_\gamma}{\Gamma_n + \Gamma_\gamma}$) for the observation of resonances above the background level was experimentally determined. This was done by reducing artificially the kernel of an existing resonance until it fell under the background, or equivalently, by increasing the kernel of an hypothetical resonance up to the point that it would be unequivocally detected. Secondly, an initial set of average resonance parameters was derived from our experimental data and evaluations [5].

Specifically, based on data for neighboring nuclei the neutron strength functions for s - and p -wave resonances is expected to be $S_0 \approx 1.1 \times 10^{-4}$ and $S_1 \approx 0.3 \times 10^{-4}$. Moreover, the number of observed resonances above the threshold allowed to determine the s -wave resonance spacing to be $D_0 \approx 300$ eV. This set of parameters were fine-tuned; in order to reproduce the experimentally-observed fraction of the MACS for $kT = 1 - 5$ keV, as well as to match the A_r values for the measured ^{204}Tl resonances, it was required to set an average radiation width $\bar{\Gamma}_\gamma$ similar to that recommended by Mughabghab [5] of 750(150) meV.

It is worth noting that at low kT energies the observable MACS is calculated to be an important fraction of the total MACS, namely, 0.93(2) at 1 keV, 0.75(4) at 2 keV, 0.60(5) at 3 keV, 0.48(7) at 4 keV and 0.40(8) at 5 keV. These observable fractions of the MACS were determined by generating several thousands of random resonance sequences using the previously mentioned set of average parameters and assuming the expected fluctuation of all involved quantities. In practice, we checked the impact of changes around the aforementioned esti-

mated values of $\pm 20\%$ in D_0 , of $\pm 50\%$ in S_0 and S_1 , and up to a factor of two in Γ_γ . The observable MACS fraction was found to be similar in all tested cases, with the main uncertainty contribution arising from the randomness of the resonance positions. As an example, the relative contribution to the total uncertainty due to the randomness of the resonance sequence is 21% and 25% at 8 keV and 30 keV, respectively. On the other hand, the relative contribution from the uncertainty in the statistical parameters is 10% and 15%, respectively. Finally, combining the predicted observable fraction with the directly measured contribution at each kT energy, the total MACS was determined in the full thermal neutron energy between 5 and 100 keV, including its uncertainty.

Quality of R-Matrix fits

Figure 1 shows an example of the R-Matrix fit with SAMMY of both $^{204}\text{Tl}(n, \gamma)$ and $^{203}\text{Tl}(n, \gamma)$ resonances around neutron energy $E_n = 2$ keV in the ^{204}Tl -enriched sample (red solid line), compared to a fit of only the $^{203}\text{Tl}(n, \gamma)$ resonance data. The residuals of the fits are shown in the bottom panels. This particular example illustrates how the unique combination of high flux and very high resolution in E_n of the n_TOF EAR1 neutron beam was crucial in the determination of new $^{204}\text{Tl}(n, \gamma)$ resonances.

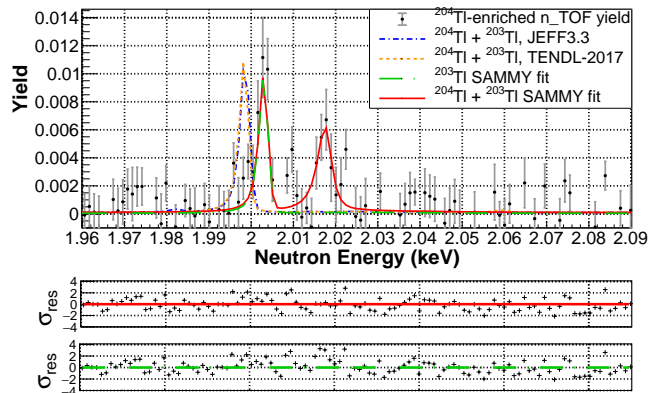


FIG. 1. In the top panel, fit of the $^{203}\text{Tl}(n, \gamma)$ and $^{204}\text{Tl}(n, \gamma)$ resonances in the sample (solid red line) compared to a fit of only the ^{203}Tl resonances (dashed green line). Evaluations for both nuclei are also shown for comparison. The bottom panels show the residuals of the fits, thereby illustrating the quality of the R-matrix analysis.

n_TOF total MACS values

The values of the $^{204}\text{Tl}(n, \gamma)$ MACS from this work are reported in Table II. These are the values depicted in Figure 2 of the main manuscript and employed for the nucleosynthesis calculations.

TABLE II. The detailed values of the new n_TOF total MACS and the corresponding uncertainty in the range of kT energies from 5 keV to 100 keV are reported in the table below. The values of Bao et al. (2000) as quoted in the KADoNiS v0.3 database are included for comparison.

kT (keV)	n_TOF total MACS (mb)	Bao et al. (mb)
5	801(203)	874
8	580(168)	-
10	497(147)	412
15	387(121)	317
20	326(108)	269
25	286(95)	238
30	260(90)	215
40	220(83)	183
50	198(74)	161
60	176(66)	145
80	157(62)	122
100	137(66)	106

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