



HAL
open science

Which is the best ^9Be carrier for $^{10}\text{Be}/^9\text{Be}$ accelerator mass spectrometry?

Silke Merchel, Regis Braucher, Johannes Lachner, Georg Rugel

► To cite this version:

Silke Merchel, Regis Braucher, Johannes Lachner, Georg Rugel. Which is the best ^9Be carrier for $^{10}\text{Be}/^9\text{Be}$ accelerator mass spectrometry?. *MethodsX*, 2021, 8, pp.101486. 10.1016/j.mex.2021.101486 . hal-03320513

HAL Id: hal-03320513

<https://hal.science/hal-03320513v1>

Submitted on 17 Aug 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



ELSEVIER

Contents lists available at ScienceDirect

MethodsX

journal homepage: www.elsevier.com/locate/mex

Method Article

Which is the best ^9Be carrier for $^{10}\text{Be}/^9\text{Be}$ accelerator mass spectrometry?Silke Merchel^{a,b,*}, Régis Braucher^c, Johannes Lachner^{a,b}, Georg Rugel^a^a Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany^b Isotope Physics, University of Vienna, Austria^c CEREGE, CNRS, Collège de France, IRD, INRAE, Aix Marseille Univ., Plateau de l'Arbois, Aix-en-Provence 13545, France

A B S T R A C T

Commercial ^9Be solutions used for chemical preparation of samples for accelerator mass spectrometry contain the cosmogenic long-lived radionuclide ^{10}Be at elevated but different $^{10}\text{Be}/^9\text{Be}$ levels. Within a systematic study of recently produced solutions, comparison to published data and new data on customised solutions from minerals, we recommend - if no customised solution is available - the ^9Be solutions from Australian Chemical Reagents (ACR) or from LGC. They contain $^{10}\text{Be}/^9\text{Be}$ at the 3.4×10^{-15} level, which is still suitable for the majority of Earth science applications, compared to customised solutions at the 10^{-16} level for lowest-level studies. Commercial solutions from Scharlab having different lot numbers, i.e. an identification number assigned to a particular lot of material from a single manufacturer, vary in $^{10}\text{Be}/^9\text{Be}$ by up to a factor of nine. Hence, it seems an advisable strategy to buy a bigger quantity of a single production batch (such as 10×100 ml bottles of ^9Be at 1 g l^{-1}) and have them tested once at any AMS facility before first use.

- The best ^9Be carrier for low-level $^{10}\text{Be}/^9\text{Be}$ applications is a customised one from minerals like phenakite.
- The best ^9Be carriers for medium- and high-level $^{10}\text{Be}/^9\text{Be}$ applications are currently from Australian Chemical Reagents (ACR) or from LGC.
- As ^9Be carriers from Scharlab of different batches (LOT) contain $^{10}\text{Be}/^9\text{Be}$ at different levels, it is advisable to buy a bigger number of bottles of the same LOT of commercial carriers after being identified to have reasonably low isotope ratios.

© 2021 Published by Elsevier B.V.

This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>)

A R T I C L E I N F O

Method name: Best-suited ^9Be carrier

Keywords: AMS, Phenakite, Beryl, Carrier, Blank

Article history: Received 25 March 2021; Accepted 10 August 2021; Available online 11 August 2021

* Corresponding author at: Universität Wien, Fakultät für Physik, Isotopenphysik, Währinger Strasse 17, 1090 Wien
E-mail address: silke.merchel@univie.ac.at (S. Merchel).

Specifications table

Subject Area:	Earth and Planetary Sciences
More specific subject area:	Cosmogenic nuclide
Method name:	Best-suited ^9Be carrier
Name and reference of original method:	Best-suited ^9Be carrier (described in Merchel et al., 2008; doi:10.1016/j.nimb.2008.07.03)
Resource availability:	not applicable

Method details

Background and motivation

The technical development of accelerator mass spectrometry (AMS) within the last 30 years has led to at least one order of magnitude lower detection limits for cosmogenic radionuclides such as ^{10}Be [1]. The question if the better AMS technique has opened-up new applications in Earth and environmental sciences or if the technical improvement has been demanded by those users from interdisciplinary research remains unanswered. Very likely, it is a win-win-situation for both communities. However, as chemical preparation of samples is a mandatory step before AMS can be performed, most demanding requirements for lowest detection limits are also essential here.

A key issue for low ^{10}Be background from sample preparation is the use of a ^9Be carrier, usually in the form of acidic solution, which contains “no” intrinsic ^{10}Be . It has been noted as early as 1984 [2] that commercially available ^9Be compounds can contain $^{10}\text{Be}/^9\text{Be}$ at the 10^{-14} level making them not well-suitable for applications like surface exposure dating. As well-shielded – from cosmic rays – beryllium minerals like beryl were measured lower as 1.7×10^{-15} [2], experienced scientists have therefore produced their own ^9Be carrier solution from those minerals (e.g., [3–5]). Only this way samples with lowest ^{10}Be concentrations could have been successfully measured above background and led to exceptional research work such as the reconstruction of glacier changes by surface exposure or - combined with ^{26}Al data - burial dating (e.g., [6–11]). Two more examples are the ^{10}Be dating of a boulder from a medieval rock fall triggered by an earthquake [12] and the determination of stellar and thermal neutron capture cross sections of ^9Be [13]. In the past, there had been a few exceptions for commercial carriers being also acceptably low, e.g., one from the company MERCK used for more than twenty years at the now-closed AMS facility at Gif-sur-Yvette [14] and those from the company Scharlab being for a certain production time in the 10^{-16} and $1\text{--}2 \times 10^{-15}$ $^{10}\text{Be}/^9\text{Be}$ range, respectively [3]. As a newer (since 2006) MERCK solution and also all tested solutions from five other companies were measured to 8×10^{-14} to 4×10^{-14} (Fig. 1 in [3]), either customised carriers from minerals (beryl, phenakite) or Scharlab carriers have been predominantly used for projects in Earth and environmental sciences.

Besides the enormous effort including working with large amounts of carcinogenic Be powder and extremely toxic hydrofluoric acid, the drawback of customised carriers is the need for accurate ^9Be determination in the produced solution. Earlier work [3] has shown that the ^9Be concentration could be determined by three different methods – graphite-furnace atomic absorption spectrometry (GF-AAS), flame-AAS and gravimetry – as precise as 0.3%, which has to be added to the overall uncertainty of all ^{10}Be AMS data. Unexpectedly, later work [4] collected results from inductively coupled plasma mass spectrometry (ICP-MS) and optical emission spectrometry (ICP-OES) differing at the 8% level in a first attempt, resulting in an overall uncertainty of 0.5% for the ^9Be concentration only after remeasurements and additional laboratories being involved in a kind-of round-robin exercise. Hence, customised ^9Be carrier solutions might add an additional (higher) uncertainty or even a source of systematic bias to ^{10}Be AMS data, when single-lab (precise) data, but not several-lab accurate data are taken-into account.

To conclude, based on word-of-mouth recommendation several scientists have used the Scharlab ^9Be carrier for their applied research projects in the last decade. Unfortunately, the $^{10}\text{Be}/^9\text{Be}$ value of more recently produced Scharlab solutions have shown increased levels of $^{10}\text{Be}/^9\text{Be}$, i.e. from 1×10^{-15} for LOT number 14844801, over 5×10^{-15} for LOT number 15563401 to finally \geq

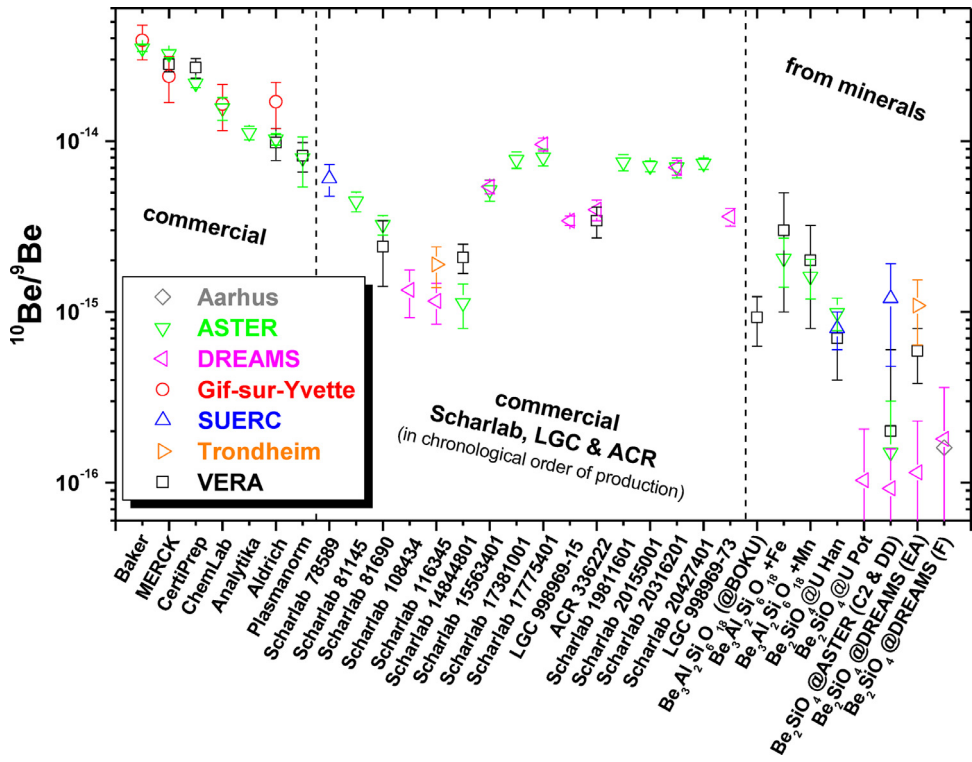


Fig. 1. $^{10}\text{Be}/^9\text{Be}$ results of different commercial and customised (from minerals) ^9Be carrier solutions measured at seven European AMS facilities. All results from Gif-sur-Yvette and SUERC, and results for Baker, MERCK, CertiPrep, ChemLab, Aldrich, Plasmanorm, and Scharlab with lower LOT numbers than 100,000 have been taken from Merchel et al. [3], thus production dates of solutions are 2007 and earlier. The commercial solution from Analytika is from 2019. Data from Aarhus (no uncertainty given) is from Heinemeier et al. [15]. Carriers made from phenakite, i.e. Be_2SiO_4 , “@U Pot”, “@U Han”, “@DREAMS (F)” have been produced at the university of Potsdam in 2016, the university of Hanover and at the DREAMS laboratory in 2014, respectively (all unpublished); carrier from beryll, i.e. $\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18}$ (@BOKU) has been produced at the University of Natural Resources and Life Sciences, Vienna. All other customised carriers have been described in Merchel et al. [3,4]. Values of commercial solutions (left) and customised from minerals (right) have been ordered in descending $^{10}\text{Be}/^9\text{Be}$ values, whereas “commercial Scharlab, LGC & ACR” has been ordered according to the production date of the solutions. Uncertainties depicted are on the 1 sigma-level and represent Poisson counting statistics as well as the scatter of multiple measurements on targets of the same material. Each datapoint corresponds to at least 1 event detected. If measurements on a single cathode at DREAMS yielded 0 events, the overall result was evaluated using the statistical approach of Feldman and Cousins [23].

7×10^{-15} for LOT numbers 17381001 and higher (Fig. 1), thus reaching blank levels, which are not any more acceptable for certain applications. Therefore, users of AMS facilities have been more and more interested in having their Scharlab or other commercial carriers (Analytika, Australian Chemical Reagents (ACR), LGC) tested before first real use. As AMS measurements of carrier solutions are cost- and time-intensive blocking precious AMS time for research projects, we would like to present here, the current status of $^{10}\text{Be}/^9\text{Be}$ determinations for different companies and LOT numbers. This way, we are hoping that we and other AMS facilities can spare some further tests and encourage the user and AMS community to spread word-of-mouth recommendations about their lowest commercial ^9Be carrier, but also to include important information about the used carrier in future scientific papers.

AMS details

New $^{10}\text{Be}/^9\text{Be}$ data added to the data set presented in [3], are published elsewhere [15] for the 1 MV AMS facility Aarhus while setting-up the facility, or have been investigated due to requests by

(external) users. This way the data collection (Fig. 1) combines unpublished data from the following AMS facilities:

- ASTER (Accélérateur pour les Sciences de la Terre, Environnement, Risques), Aix-en-Provence, France [16]
- DREAMS (DREsden AMS), Dresden, Germany [17,18]
- Trondheim, Norway [19]
- VERA (Vienna Environmental Research Accelerator), Vienna, Austria [20].

Compared to their earlier measurements [3], ASTER and VERA have changed their set-ups with respect to an ion-source with higher BeO^- currents (ASTER) and improved ^{10}Be detection using a passive absorber, consisting of a stack of silicon nitride foils, for isobar suppression (VERA). All values are traceable to the primary NIST SRM 4325 standard with the currently most accepted value of $(2.79 \pm 0.03) \times 10^{-11} \text{ }^{10}\text{Be}/^9\text{Be}$ [21] via in-house secondary standards. No further blank corrections e.g., by subtracting machine blank values have been applied.

Conclusion and recommendation

This study is a regionally-influenced snapshot of the last years and we would like to raise awareness that the "best manufacturer" can change over time. However, currently we can recommend the ^9Be carrier (LOT 998969-15; purchased in 2019) from LGC being as low as $(3.42 \pm 0.22) \times 10^{-15} \text{ }^{10}\text{Be}/^9\text{Be}$ or the ACR (LOT 336222; purchased in 2019) being as low as $(3.41 \pm 0.70) \times 10^{-15} \text{ }^{10}\text{Be}/^9\text{Be}$. Another more recent batch from LGC (LOT 998969-73; purchased in 2021) gave the same result within uncertainty, i.e. $(3.61 \pm 0.43) \times 10^{-15}$, as the LGC batch from two years earlier. There is no difference within 1-sigma uncertainty between most of the $^{10}\text{Be}/^9\text{Be}$ data from the same commercial carrier solutions measured at different AMS facilities showing the excellent comparability (Fig. 1). In reverse, measuring a commercial carrier solution at a single AMS facility will produce AMS data valid also for other facilities. As commercial solutions from the company Scharlab having different LOT numbers, i.e., an identification number assigned to a particular lot of material from a single manufacturer, vary in $^{10}\text{Be}/^9\text{Be}$ by up to a factor of nine, we strongly recommend to buy a bigger quantity of a single production batch (such as $10 \times 100 \text{ ml}$ bottles of ^9Be at 1 g l^{-1}) and have them tested once at any AMS facility before first use.

At the level of customised carriers, which can be as low as 9×10^{-17} , comparisons of different solutions measured at the same facility will allow to find out the best. However, if the same customised solution is measured at different facilities, the extremely low counting statistics are influenced by the use of standards at different levels, e.g., 10^{-11} vs. 10^{-12} , and general long-term-memory of ion sources (e.g., [18]). Ineffective isobar suppression (from boron) should not influence the results at sophisticated facilities, but differences in measurement efficiencies at different facilities might result in higher detection limits when detecting "zero counts". As chemical processing will also increase the final ratio of the so-called processing blank, there are only rare cases when you have to choose between a customised " $<10^{-16}$ " or a " 5×10^{-16} " $^{10}\text{Be}/^9\text{Be}$ carrier [22]; both carriers at this low level will be suitable for nearly all applications.

Acknowledgements

We are very grateful to the scientists at the AMS facilities at Aix-en-Provence (ASTER), Dresden (DREAMS), Trondheim and Vienna (VERA) for measurements of BeO prepared from different carrier solutions; especially ASTER-Team (Georges Aumaître, Didier L. Bourlès, Karim Keddadouche), Marie-Josée Nadeau, Andreas Scharf, Martin Seiler, Carlos Vivo-Vilches, Anton Wallner and René Ziegenrucker. The ASTER AMS national facility is supported by the INSU/CNRS, the ANR through the "Projets thématiques d'excellence" program for the "Equipements d'excellence" ASTER-CEREGE action and IRD. Parts of this research were carried out at the Ion Beam Centre (IBC) at the Helmholtz-Zentrum Dresden-Rossendorf e.V., a member of the Helmholtz Association. Some AMS measurements at the VERA facility were supported by the RADIATE project under the Grant Agreement 824096319 from the EU Research and Innovation programme HORIZON 2020. We also thank the careful users for

providing carrier solutions for having them measured before they are applied to chemical processing of real samples, namely Florian Adolphi, Bodo Bookhagen, Toshiyuki Fujioka, Philipp Häuselmann, Dominik Koll, Maylin Malter, Ari Matmon, Stephanie Neuhuber, Zsófia Ruszkiczay-Rüdiger and Michal Šujan. Finally, an inspiring review by Darryl Granger is acknowledged.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

- [1] W. Kutschera, Accelerator mass spectrometry: state of the art and perspectives, *Adv. Physics X* 1 (2016) 570–595, doi:[10.1080/23746149.2016.1224603](https://doi.org/10.1080/23746149.2016.1224603).
- [2] R. Middleton, J. Klein, L. Brown, F. Tera, ^{10}Be in commercial beryllium, *Nucl. Instrum. Methods Phys. Res. B* 5 (1984) 511–513, doi:[10.1016/0168-583X\(84\)90009-0](https://doi.org/10.1016/0168-583X(84)90009-0).
- [3] S. Merchel, M. Arnold, G. Aumaitre, L. Benedetti, D.L. Bourlès, R. Braucher, V. Alfimov, S.P.H.T. Freeman, P. Steier, A. Wallner, Towards more precise ^{10}Be and ^{36}Cl data from measurements at the 10^{-14} level: Influence of sample preparation, *Nucl. Instrum. Methods Phys. Res. B* 266 (2008) 4921–4926, doi:[10.1016/j.nimb.2008.07.031](https://doi.org/10.1016/j.nimb.2008.07.031).
- [4] S. Merchel, W. Bremser, D.L. Bourlès, U. Czeslik, J. Erzinger, N.-A. Kummer, L. Leanni, B. Merkel, S. Recknagel, U. Schaefer, Accuracy of ^9Be -data and its influence on ^{10}Be cosmogenic nuclide data, *J. Radioanal. Nucl. Chem.* 298 (2013) 1871–1878, doi:[10.1007/s10967-013-2746-x](https://doi.org/10.1007/s10967-013-2746-x).
- [5] K. Nishiizumi, D. Lal, J. Klein, R. Middleton, J.R. Arnold, Production of ^{10}Be and ^{26}Al by cosmic rays in terrestrial quartz *in situ* and implications for erosion rates, *Nature* 319 (1986) 134–136, doi:[10.1038/319134a0](https://doi.org/10.1038/319134a0).
- [6] J.M. Schaefer, G.H. Denton, M. Kaplan, A. Putnam, R.C. Finkel, D.J.A. Barrell, B.G. Andersen, R. Schwartz, A. Mackintosh, T. Chinn, C. Schlüchter, High-Frequency Holocene Glacier Fluctuations in New Zealand Differ from the Northern Signature, *Science* 324 (2009) 622–625, doi:[10.1126/science.1169312](https://doi.org/10.1126/science.1169312).
- [7] K. Hahne, R. Naumann, S. Niedermann, H.-U. Wetzel, S. Merchel, G. Rugel, Geochemische Untersuchungen an Moränen des Inylchek-Gletschers im Tien Shan, *Syst. Erde GFZ J.* 3 (2) (2013) 44–49, doi:[10.2312/GFZ.syserde.03.02.7](https://doi.org/10.2312/GFZ.syserde.03.02.7).
- [8] I. Schimmelpfennig, J.M. Schaefer, N. Akçar, T. Koffman, S. Ivy-Ochs, R. Schwartz, R.C. Finkel, S. Zimmerman, C. Schlüchter, A chronology of Holocene and Little Ice Age glacier culminations of the Steingletscher, Central Alps, Switzerland, based on high-sensitivity beryllium-10 moraine dating, *Earth Planet. Sci. Lett.* 393 (2014) 220–230, doi:[10.1016/j.epsl.2014.02.046](https://doi.org/10.1016/j.epsl.2014.02.046).
- [9] S.M. Braumann, J.M. Schaefer, S.M. Neuhuber, J.M. Reitner, C. Lüthgens, M. Fiebig, Holocene glacier change in the Silvretta Massif (Austrian Alps) constrained by a new ^{10}Be chronology, historical records and modern observations, *Quat. Sci. Rev.* 245 (2020) 106493, doi:[10.1016/j.quascirev.2020.106493](https://doi.org/10.1016/j.quascirev.2020.106493).
- [10] P.R. Bierman, J.D. Shakun, L.B. Corbett, S.R. Zimmerman, D.H. Rood, A persistent and dynamic East Greenland Ice Sheet over the past 7.5 million years, *Nature* 540 (2016) 256–260, doi:[10.1038/nature20147](https://doi.org/10.1038/nature20147).
- [11] J.M. Schaefer, R.C. Finkel, G. Balco, R.B. Alley, M.W. Caffee, J.P. Briner, N.E. Young, A.J. Gow, R. Schwartz, Greenland was nearly ice-free for extended periods during the Pleistocene, *Nature* 540 (2016) 252–255, doi:[10.1038/nature20146](https://doi.org/10.1038/nature20146).
- [12] W. Schwanghart, A. Bernhardt, A. Stolle, P. Hoelzmann, B.R. Adhikari, C. Andermann, S. Tofelde, S. Merchel, G. Rugel, M. Fort, O. Korup, Repeated catastrophic valley infill following medieval earthquakes in the Nepal Himalaya, *Science* 351 (2016) 147–150, doi:[10.1126/science.aac9865](https://doi.org/10.1126/science.aac9865).
- [13] A. Wallner, M. Bichler, L. Coquard, L. Dillmann, O. Forstner, R. Golser, M. Heil, F. Käppler, W. Kutschera, C. Lederer-Woods, M. Martschini, A. Mengoni, S. Merchel, L. Michlmayr, A. Priller, P. Steier, M. Wiescher, Stellar and thermal neutron capture cross section of ^9Be , *Phys. Rev. C* 99 (2019) 015804, doi:[10.1103/PhysRevC.99.015804](https://doi.org/10.1103/PhysRevC.99.015804).
- [14] G.M. Raisbeck, F. Yiou, D. Bourlès, J. Lestringuez, D. Deboffle, Measurements of ^{10}Be with a tandemtron accelerator operating at 2 MV, *Nucl. Instrum. Methods Phys. Res. B* 5 (1984) 175–178, doi:[10.1016/0168-583X\(84\)90505-6](https://doi.org/10.1016/0168-583X(84)90505-6).
- [15] J. Heinemeier, J. Olsen, M. Klein, D. Mous, The new extended HVE 1 MV multi-element AMS system for low background installed at the Aarhus AMS Dating Centre, *Nucl. Instrum. Methods Phys. Res. B* 361 (2015) 143–148, doi:[10.1016/j.nimb.2015.05.039](https://doi.org/10.1016/j.nimb.2015.05.039).
- [16] M. Arnold, S. Merchel, D.L. Bourlès, R. Braucher, L. Benedetti, R.C. Finkel, G. Aumaitre, A. Gottdang, M. Klein, The French accelerator facility ASTER: Improved performance and developments, *Nucl. Instrum. Methods Phys. Res. B* 268 (2010) 1954–1959, doi:[10.1016/j.nimb.2010.02.107](https://doi.org/10.1016/j.nimb.2010.02.107).
- [17] S. Akhmadaliev, R. Heller, D. Hanf, G. Rugel, S. Merchel, The new 6 MV AMS-facility DREAMS at Dresden, *Nucl. Instrum. Methods Phys. Res. B* 294 (2013) 5–10, doi:[10.1016/j.nimb.2012.01.053](https://doi.org/10.1016/j.nimb.2012.01.053).
- [18] G. Rugel, S. Pavetich, S. Akhmadaliev, S.M. Enamorado Baez, A. Scharf, R. Ziegenrucker, S. Merchel, The first four years of the AMS-facility DREAMS: Status and developments for more accurate radionuclide data, *Nucl. Instrum. Methods Phys. Res. B* 370 (2016) 94–100, doi:[10.1016/j.nimb.2016.01.012](https://doi.org/10.1016/j.nimb.2016.01.012).
- [19] M. Seiler, J. Anjar, E. Værnes, M.-J. Nadeau, G. Scognamiglio, First ^{10}Be measurements at Trondheim 1 MV AMS, *Nucl. Instrum. Methods Phys. Res.* 437 (2018) 123–129, doi:[10.1016/j.nimb.2018.08.013](https://doi.org/10.1016/j.nimb.2018.08.013).
- [20] P. Steier, M. Martschini, J. Buchriegler, J. Feige, J. Lachner, S. Merchel, L. Michlmayr, A. Priller, G. Rugel, E. Schmidt, A. Wallner, E.M. Wild, R. Golser, Comparison of methods for the detection of ^{10}Be with AMS and a new approach based on a silicon nitride foil stack, *Int. J. Mass Spectrom.* 444 (2019) 116175, doi:[10.1016/j.ijms.2019.116175](https://doi.org/10.1016/j.ijms.2019.116175).

- [21] K. Nishiizumi, M. Imamura, M.W. Caffee, J.R. Southon, R.C. Finkel, J. McAninch, Absolute calibration of ^{10}Be AMS standards, Nucl. Instrum. Methods Phys. Res. 258 (2007) 403–413, doi:[10.1016/j.nimb.2007.01.297](https://doi.org/10.1016/j.nimb.2007.01.297).
- [22] K.M. Wilcken, T. Fujioka, D. Fink, R.H. Fülöp, A.T. Codilean, K. Simon, C. Mifsud, S. Kotevski, SIRIUS Performance: ^{10}Be , ^{26}Al and ^{36}Cl measurements at ANSTO, Nucl. Instrum. Methods Phys. Res. B 455 (2019) 300–304, doi:[10.1016/j.nimb.2019.02.009](https://doi.org/10.1016/j.nimb.2019.02.009).
- [23] G.J. Feldman, R.D. Cousins, Unified approach to the classical statistical analysis of small signals, Phys. Rev. D 57 (1998) 3873, doi:[10.1103/PhysRevD.57.3873](https://doi.org/10.1103/PhysRevD.57.3873).