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A new ^{7}Be AMS capability established at CAMS and the potential for large datasets

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ABSTRACT

We have developed a ^{7}Be measurement capability at the Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Lab. This capability is tested with dilutions of a commercial ^{7}Be tracer, a new ^{7}Be reference material produced at CAMS (which we verify to be free of ^{10}Be), and with experimental targets from a variety of sample matrices. Our results indicate that ^{7}Be can be measured at high precision for low activities (~1% for 0.1 Bq; ~10% for 0.001 Bq), with an estimated detection limit of ~0.0001 Bq (^{7}Be/^{9}Be of $\sim 7 \times 10^{-17}$; equivalent to $\sim 10^{3}$ atoms)—more than two orders of magnitude lower than that typical of counting techniques. Furthermore, because ^{10}Be and ^{7}Be sample preparation are identical, both isotopes can be measured on the same target with no additional laboratory preparation.

1. Introduction

^{7}Be is produced in the stratosphere and upper troposphere during cosmic ray collisions with heavier nuclei (mostly oxygen and nitrogen) [1], and is quickly scavenged from the atmosphere to Earth's surface by rain and snow. Because of its short half-life ($t_{1/2}$ = 53 days) and strong adsorption to fine particles, ^{7}Be is a powerful tracer of soil, watershed, and atmospheric processes over short timescales, useful for examining a variety of climate and land-use activities on surface material fluxes [2]. Recent work has demonstrated the potential of ^{7}Be as a tool for investigating rates of soil erosion [3], establishing watershed sediment budgets [4], fingerprinting runoff and suspended sediment [5], and water mass mixing in the ocean [6]. However, environmental samples are typically measured via gamma counting, which requires large sample sizes (100–200 L for suspended sediment and ocean water, ~10 L for rain water, ~100 g for soil) and long measurement times (1–2+ days/sample). A recent review identified these logistics as prohibiting the large datasets and low-level measurements needed for proper characterization of ^{7}Be inventories in space (surface sampling) and in depth in a soil column [2]. Measurements of ^{7}Be by accelerator mass spectrometry (AMS) techniques have been shown to be significantly (10–100 times) more sensitive than counting approaches [7–10]. Thus, ^{7}Be measured with AMS has the potential to produce high-resolution datasets while collecting and processing much smaller samples.

In this paper we demonstrate the feasibility and capability of measuring ^{7}Be on the HVEC 10 MV FN Tandem Van de Graaff accelerator at CAMS. We also demonstrate that a recently developed method for making high-purity ^{7}Be [11] is essentially free of ^{10}Be. This makes it ideal as a reference material for AMS and permits low-level measurement of both ^{7}Be and ^{10}Be on the same AMS target after a brief system reconfiguration.

2. Methods

Between August 2016 and January 2017, we conducted three experimental AMS runs analyzing (1) a dilution series prepared from a commercial ^{7}Be tracer, (2) dilutions of a ^{7}Be reference material produced in-house using the CAMS isotope production line, and (3) five environmental test samples. ^{10}Be was also measured on the test samples and the CAMS ^{7}Be reference material to assess the practicality of measuring ^{10}Be and ^{7}Be on the same target and to characterize ^{10}Be in the reference material.

2.1. Experimental AMS setup

^{7}Be and ^{10}Be AMS targets were prepared and measured following existing protocols for ^{10}Be target preparation at CAMS [12]. BeO powders (containing between 250–350 μg Be) were crushed and mixed

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Table 1
Accelerator settings used for experimental measurement of ^{7}Be and ^{10}Be in this work; settings for routine measurement of ^{10}Be are shown for comparison.

Isotope	Injected beam		Terminal voltage	Charge state		
	(rare)	(stable)	(MV)	^{7}Be or ^{10}Be (post-accelerator)	^{9}Be (HE Faraday cup)	Post-stripped[a] (to gas detector)
Experimental settings						
^{7}Be	$^{7}Be^{16}O^{-}$	$^{9}Be^{16}O^{-}$	8.3	+3	+4	+4
^{10}Be	$^{10}Be^{16}O^{-}$	$^{9}Be^{16}O^{-}$	8.3	+3	+3	+4
Routine settings						
^{10}Be	$^{10}Be^{16}O^{-}$	$^{9}Be^{16}O^{-}$	8.3	+3	+3	+3[b]

[a] A 10 μg/cm^2 carbon foil was used for all post-stripping.
[b] Was not post-stripped.

2:3 by volume (~1:1 by mass) with Nb powder (Alfa Aesar Puratronic -325 mesh, 99.99%). This mixture was packed into stainless steel targets (#55 hole) designed for CAMS ion sources. CAMS uses a high-intensity modified Middleton cesium source [13,14] able to produce > 30 μA $^{9}Be^{3+}$ beam currents during routine measurement of ^{10}Be.

The accelerator settings used for our experiments, along with settings used for routine ^{10}Be measurement are detailed in Table 1. BeO^{-} is selected from the anion beam generated by the source and then injected into the accelerator at 8.3 MV. Injection of $^{7}Be^{16}O^{-}$ or $^{9}Be^{16}O^{-}$, and $^{10}Be^{16}O^{-}$ or $^{9}Be^{16}O^{-}$ is controlled by fast electrostatic bouncing such that the stable:rare isotope measurement time is 1:3000. Following stripping using a carbon stripper foil at the terminal, $^{9}Be^{+4}$ or $^{9}Be^{+3}$ is selected with an analyzing magnet and measured in an off-axis Faraday cup while $^{7}Be^{+3}$ or $^{10}Be^{+3}$, respectively, is post-stripped to the +4 charge state and measured in a gas-filled (P10) detector. Post-stripping in this fashion for ^{7}Be is necessary in order to remove isobaric ^{7}Li—the most abundant stable isotope of Li. This post-stripping approach has been demonstrated previously in other AMS systems and is effective since formation of $^{7}Li^{+4}$ is not possible [7–10]. For ^{10}Be, post-stripping is typically not used to remove isobaric ^{10}B since (1) standard chemical preparation methods [e.g. 15,16] and materials produce targets suitably low in boron such that ^{10}B can be reduced acceptably by an absorber foil at the entrance of the detector, and (2) transmission through the post-stripper reduces efficiency by ~10% as determined by loss in gated count rate. However, for the experiments in this study the post-stripper was left in so that accelerator settings could be quickly reconfigured for ^{7}Be or ^{10}Be.

2.2. Commercial ^{7}Be dilution series

We prepared a dilution series from a ^{7}Be tracer solution procured from Eckert & Ziegler Isotope Products Lab (product #8007). This solution had a ^{7}Be activity of 7.77 ± 0.30 × 10^4 Bq/g (error represents relative expanded uncertainties) on reference date 13-Aug-2015, or 5.58 ± 0.21 × 10^2 Bq/g on 26-Aug-2016 when it was used for the first experiment. To facilitate AMS analysis, each dilution was spiked with a ^{9}Be carrier routinely used for high-level ^{10}Be sample preparation at CAMS ([Be] = 990 ± 3 ppm; $^{10}Be/^{9}Be = 1.35 \pm 0.06 \times 10^{-14}$). Enough ^{9}Be carrier was added to make two AMS targets from each dilution (~500 μg Be), and then split to ensure identical $^{7}Be/^{9}Be$ between duplicate targets. At time of measurement, the expected $^{7}Be/^{9}Be$ for this series ranged from 1790 – 126 × 10^{-15} (Table 2; 7BE082016). Although the ^{9}Be carrier was not characterized for ^{7}Be, we assume it did not contribute significant ^{7}Be to our measurements since it was prepared more than 30 ^{7}Be half-lives prior to these measurements. Following carrier addition, beryllium was precipitated from each solution as BeOH by bringing to pH = 9 with dilute NH_4OH. This precipitant was then dissolved in 3 drops of 5% HNO_3, transferred to a quartz crucible, and converted to BeO by heating in a muffle furnace at 850 °C for 1 min.

2.3. CAMS ^{7}Be reference material

We created a ^{7}Be material with the CAMS isotope production line using the method of [11]. In this approach, a 4:1 (by mass) mixture of LiOH and Al powder is pressed into disc (~5 mm diameter; ~0.3 mm thick). The disc is mounted in a chamber at the end of the beamline and bombarded with 7 MeV protons. This energy level was chosen to

Table 2
$^{7}Be/^{9}Be$ results for reference materials and BeO blanks from the three experimental AMS runs. Mean value and standard deviation denoted by μ ± σ.

Referece material	$^{7}Be/^{9}Be$ Expected (x10^{-15})[a]	$^{7}Be/^{9}Be$ AMS measured μ ± σ (x10^{-15})[b]		# of meas.	Total ^{7}Be counts	Total ^{7}Be meas. time (s)	Total $^{9}Be^{+4}$ Q (nC)
Commercial tracer dilution series (26-Aug-2016)							
7BE082016-A	1790	1790	20	4	20193	249	642
	1790	1800	20	5	25179	376	803
7BE082016-B	1010	1010	10	4	10121	223	575
	1010	1020	10	4	12060	297	675
7BE082016-C	507	525	6	4	7033	340	766
	507	509	6	4	8059	388	905
7BE082016-D	254	256	4	4	4038	365	904
	254	256	5	4	4036	328	896
7BE082016-E	126	128	3	3	2658	826	1188
	126	132	2	4	4021	720	1739
BeO blank[c]	0	0.069	0.035	3	4	975	3230
CAMS reference material (08-Dec-2016)							
7BE122016-A	3080	3130	30	2	20289	147	399
	3080	3170	30	3	30387	201	635
7BE122016-B	1540	1520	10	7	35389	517	1490
	1540	1520	20	7	35326	482	1501
	1540	1610	10	7	35406	542	1446
	1540	1550	10	7	35539	508	1496
	1540	1490	10	7	35451	496	1543
7BE122016-C	310	317	8	3	6039	495	1177
	310	324	8	3	6067	525	1237
7BE122016-D	160	167	4	3	3037	379	1122
	160	168	7	2	2010	270	779
BeO blank[c]	0	0.044	0.044	2	1	714	1463
CAMS reference material (19-Jan-2017)							
7BE012017-A	2830	2890	70	3	15169	133	398
7BE012017-B	1400	1420	20	3	9086	212	485
	1400	1430	20	3	9072	194	485
	1400	1410	20	3	9052	205	491
	1400	1360	30	3	9066	190	505
	1400	1380	20	3	9064	229	500
7BE012017-C	305	309	8	3	3030	244	744
7BE012017-D	153	157	3	3	3019	500	1468
BeO blank[c]	0	0.039	0.039	2	1	601	1961

[a] Adjusted for ^{7}Be decay to reflect values on measurement date.
[b] Ratios for each run normalized to primary dilution (shaded gray); uncertainties are the larger of counting uncertainty or the standard deviation of individual acquisitions.
[c] Commercial BeO powder from Alfa Aesar.

optimize production of ^{7}Be via ^{7}Li(p,n) [17] and is lower than the energy necessary to produce ^{10}Be via O(p,x) [18]. After irradiation for ~4 h, the disc was dissolved and the Li and Al fractions were removed using cation exchange chromatography [11]. This procedure produced $3.41 \pm 9 \times 10^{11}$ atoms ($5.14 \pm 0.12 \times 10^{4}$ Bq) of ^{7}Be as characterized by a co-axial HPGe detector (reference date 12-Dec-2016). Two sets of four dilutions (see Table 2; series 7BE122016 and 7BE012017) of this material were prepared for AMS as described in Section 2.2. These were used as the reference material for unknown targets in the December 2016 and January 2017 experimental ^{7}Be AMS runs.

2.4. Preparation of test samples

During or immediately following atmospheric precipitation events, samples of rain, snow, as well as sand and gravel exposed to rain were collected for AMS analysis of ^{7}Be and ^{10}Be. These consisted of (1) rain water collected from two different storms in Livermore, CA, (2) snow collected from an undisturbed surface in central Utah (10 km west of Price, UT), (3) samples of sand and gravel stored outside in exposed bins from a concrete supplier in Livermore, CA, and (4) tap water sourced from the Livermore, CA Municipal Water utility. A spread of sample types and sizes were chosen to help inform logistics for future environmental sampling.

Following collection, samples and process blanks were spiked with 300–350 µg ^{9}Be carrier. After evaporating water samples down to < 5 mL, each sample was transferred to a 50 mL centrifuge tube and leached overnight in 40 mL aqua regia. Leachates were extracted from particulates with a transfer pipette following centrifugation. Remaining particulates were rinsed and vortexed three times in 40 mL 1% HNO_3, and these fractions were similarly extracted and added to their respective leachates. The leachates were evaporated to dryness in Teflon beakers and residues were converted to fluoride salts during three cycles of re-dissolution in 5 mL concentrated HF following evaporation to dryness. These residues were transferred to clean centrifuge tubes with 20 mL deionized water, vortexed to break up the fluoride cakes, and heated to 60 °C overnight. Leachates were removed via transfer pipette following centrifugation, and this leaching step was repeated twice more. This "fluoride cake" approach takes advantage of the relatively high solubility of BeF_2 in water compared to most other fluoride compounds and helps to significantly reduce the total cation load while maintaining a reasonable (70–90%) Be extraction efficiency [e.g. 19]. Following fluoride cake leaching, the samples were evaporated to dryness and beryllium was extracted with ion-exchange chromatography using standard procedures [16]; the beryllium fraction was then precipitated as BeOH and prepared for AMS as described in Section 2.1.

AMS measurements of ^{7}Be/^{9}Be were made against the CAMS reference material (7BE122016-B for the 08-Dec-2016 run; 7BE012017-B for the 19-Jan-2017 run); measurements of ^{10}Be/^{9}Be were made against the CAMS primary ^{10}Be standard (07KNSTD3110, nominal ^{10}Be/^{9}Be = 2850×10^{-15} [20]). For these ^{10}Be measurements, no boron correction was necessary. This is because post-stripping to the highest charge state for beryllium ($^{10}Be^{+4}$) effectively removes the ^{10}B isobar, which is efficiently stripped to $^{10}B^{+5}$ at the selected beam energy (28.11 MeV). We limited acquisition times for the first isotope to < 1200 s to ensure sufficient material remained in the targets for measurement of the second isotope. Although the BeO-Nb mixture used for these targets is exhausted in the CAMS source after 9000 s, approximately two-thirds of the target is typically sputtered after 3000–4000 s [21].

3. Results and discussion

3.1. Reference materials

Measurements of ^{7}Be/^{9}Be from dilutions of both the CAMS and Eckert & Ziegler ^{7}Be reference materials agree well with expected values over a range of 126 – 3080×10^{-15} (Table 2; Fig. 1) and produced count rates between 4.3 and 144 atoms ^{7}Be/s for these end-member ratios, respectively. Counting uncertainties near ~2% or better were obtained for each target. Three BeO blanks, one measured during each experimental AMS run, yielded ^{7}Be/^{9}Be values of 0.069, 0.044, and 0.039×10^{-15}, consistently 3–4 orders of magnitude below the lowest ratio prepared in each dilution series (Fig. 2), and resulting in only 6 atoms of ^{7}Be counted between the three targets (~0.0026 atoms ^{7}Be/s). Because the Alfa Aesar BeO powder used in these blanks was purchased in 2011 and kept sealed and away from sources of ionizing radiation, the material should be completely devoid of ^{7}Be. From the BeO blank ratios, relative ratios of reference materials in corresponding runs, and known atoms of ^{7}Be in reference targets, we calculate a ^{7}Be background of 1100, 700, and 600 atoms as recorded by the three BeO blank targets. Thus, we adopt a value of 10^{3} atoms as the effective detection limit for the CAMS system.

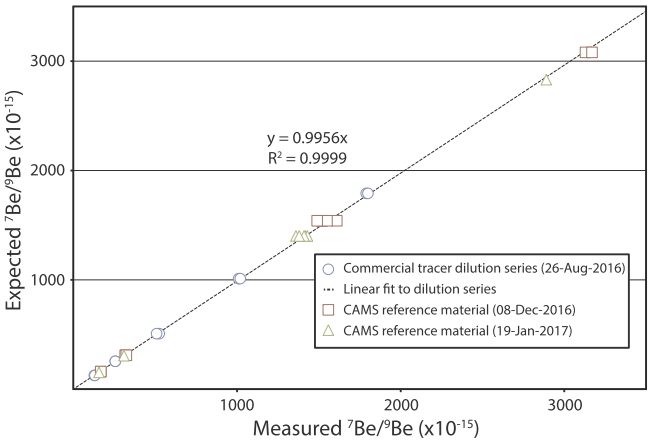


Fig. 1. Measured vs. expected ^{7}Be/^{9}Be for dilutions of commercial tracer and CAMS ^{7}Be reference materials (see Table 2). Error bars too small to be shown.

The ^{7}Be tracer obtained from Eckert & Ziegler contained concentrations of ^{10}Be unsuitably high for measurement with the CAMS AMS system as the $^{10}Be^{+4}$ beam overwhelmed the electronics in the gas-filled detector. Thus, only ^{7}Be was acquired for those targets. Moreover, the ^{10}Be/^{9}Be in BeO blanks measured afterward was elevated 35–50% above nominal values (1.2×10^{-14}; Fig. 3), indicating a significant ^{10}Be memory effect and requiring cleaning of the source and sample handling areas before further ^{10}Be acquisition. Thus, the Eckert

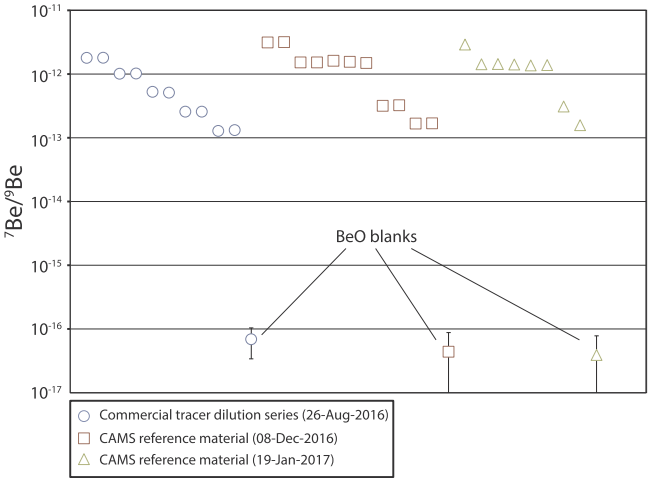


Fig. 2. ^{7}Be/^{9}Be results for all reference materials and BeO blanks measured during the three experimental AMS runs (error bars are 1σ and are smaller than symbols for ratios greater than 10^{-13}). Ratios in BeO blank material are all in the mid 10^{-17} range—more than three orders of magnitude lower than the lowest-ratio reference dilutions.

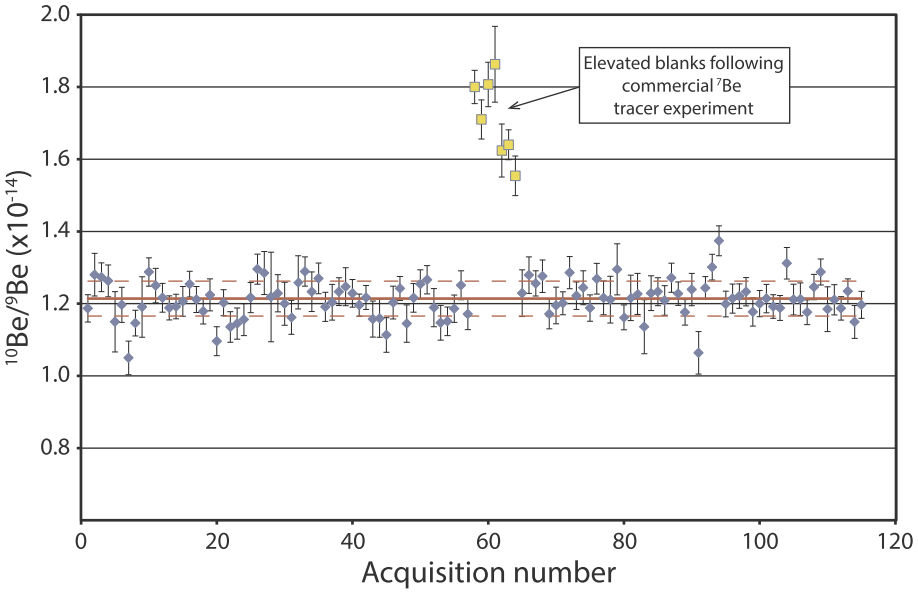


Fig. 3. Long-term (Nov 2015 – Aug 2017) ^{10}Be measurement record of commercial BeO blank material (n = 115) measured at CAMS, with a nominal ^{10}Be/^{9}Be of $1.21 \pm 0.05 \times 10^{-14}$ (solid red line; 1σ uncertainty – dashed red lines). Blue diamonds represent BeO blank measurements from routine ^{10}Be runs; error bars represent the larger of the counting uncertainty or the standard deviation of all target acquisitions. Yellow squares represent BeO blanks run after the commercial ^{7}Be dilution experiment and indicate a significant contamination from the very high ^{10}Be levels in the ^{7}Be material. Blanks returned to nominal levels after two cleanings of the ion source and sample handling areas. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and Ziegler material is unacceptable for AMS systems that routinely measure ^{10}Be and prohibits measuring ^{10}Be and ^{7}Be in the same target. This was the rationale for producing and testing our own reference material for the later experiments.

During the 08-Dec-2016 run, the ^{10}Be content in the CAMS ^{7}Be reference material was measured on one of the 7BE122016-C targets, yielding a ^{10}Be/^{9}Be of $14 \pm 1 \times 10^{-15}$ (Table 3). This is indistinguishable from the known value in the ^{9}Be carrier that was added to this target during isotope dilution (^{10}Be/^{9}Be = $13.5 \pm 0.6 \times 10^{-15}$). Furthermore, the BeO blank and ^{9}Be carrier blank measured during the 19-Jan-2017 run—where ^{7}Be measurements were made before ^{10}Be—exhibited no elevation in ratio (^{10}Be/^{9}Be = 10×10^{-15} and 1.4×10^{-15}, respectively; Table 3). This indicates no ^{10}Be memory, at least not at the low 10^{-15} level, from the entire suite of ^{7}Be targets that were sputtered beforehand. These observations imply that the CAMS ^{7}Be reference material is viable as an AMS standard and virtually free of ^{10}Be. In the future, additional measurements of the concentrated stock solution (following ^{7}Be depletion) will test this assertion.

3.2. Test samples

Both ^{7}Be/^{9}Be and ^{10}Be/^{9}Be were successfully measured on all test samples and were distinguishable from the process blanks (with the exception of the tap water sample). Count rates and counting uncertainties for resolvable samples were between 0.013 and 9.7 atoms/s (22–1.8%, 1σ) for ^{7}Be, and from 1.3 to 123 atoms/s (3.4 to < 1%, 1σ) for ^{10}Be. Fig. 4 shows the average ^{9}Be current per acquisition for all targets where both ^{7}Be and ^{10}Be were measured (Table 3). The targets typically produced initial average ^{9}Be^{+3} currents between 18 and 30 μA and ^{9}Be^{+4} currents ~33% lower, between 6 and 10 μA (presumably reflecting the relative charge state distribution after terminal stripping). The order of isotopic measurement had no impact on target longevity and all targets sufficiently endured measurement of both ^{7}Be and ^{10}Be.

Blank-corrected ^{7}Be and ^{10}Be concentrations for the samples calculated both at time of measurement and at time of collection, are shown in Table 4. These results indicate the following.

(1) The adopted chemical preparation methods for the water, sand, and gravel samples produced adequate beryllium yields.

Table 3
^{7}Be/^{9}Be and ^{10}Be/^{9}Be results for unknown samples and reference materials measured for both isotopes.

Sample ID	Measured ^{7}Be/^{9}Be μ ± σ[d] (×10^{-15})		# of meas.	Total ^{7}Be counts	Total ^{7}Be meas. time (s)	Total ^{9}Be^{+4} Q (nC)	Measured ^{10}Be/^{9}Be μ ± σ[d] (×10^{-15})		# of meas.	Total ^{10}Be counts	Total ^{10}Be meas. time (s)	Total ^{9}Be^{+3} Q (nC)
08-Dec-2016 run												
TapH20	0.24	0.14	2	3	600	788	16	1	2	452	600	5042
Rain-A	0.62	0.36	3	20	900	1467	26	1	2	854	600	5910
Sand	0.41	0.13	3	12	900	1757	3170	40	3	10121	82	578
Gravel	9.3	0.5	4	337	1200	2436	2020	30	4	7060	104	636
Process blank A[a]	0.26	0.10	3	8	900	1791	23	2	2	682	600	5277
7BE122016-C	317	8	3	6039	495	1177	14	1	2	413	600	5142
BeO blank	0.044	0.044	2	1	714	1463	12	1	3	524	900	7786
KNSTD3110[c]	0.075	0.075	1	1	300	854	2870	20	8	40539	381	2585
19-Jan-2017 run												
Rain-B	44	2	4	1594	1200	2754	119	4	3	1197	900	1818
Snow	299	6	3	3005	311	766	587	15	4	4015	373	1250
Process blank B[b]	0.20	0.10	2	4	600	1368	2.4	0.3	2	49	600	3682
BeO blank	0.039	0.039	2	1	601	1961	10	1	2	241	600	4264
^{9}Be carrier blank[b]	ND	ND	2	0	909	1238	1.4	0.3	3	12	900	1536

[a] High-ratio ^{9}Be carrier (990 ± 3 ppm Be); nominal ^{10}Be/^{9}Be = $13.5 \pm 0.6 \times 10^{-15}$ (uncertainties 1σ).
[b] Low-ratio ^{9}Be carrier (425 ± 1 ppm Be); nominal ^{10}Be/^{9}Be = $0.7 \pm 0.3 \times 10^{-15}$ (uncertainties 1σ).
[c] Primary standard used for routine measurement of ^{10}Be at CAMS (contains no 7Be).
[d] Uncertainties reported as the larger of counting uncertainty or the standard deviation of individual acquisitions.

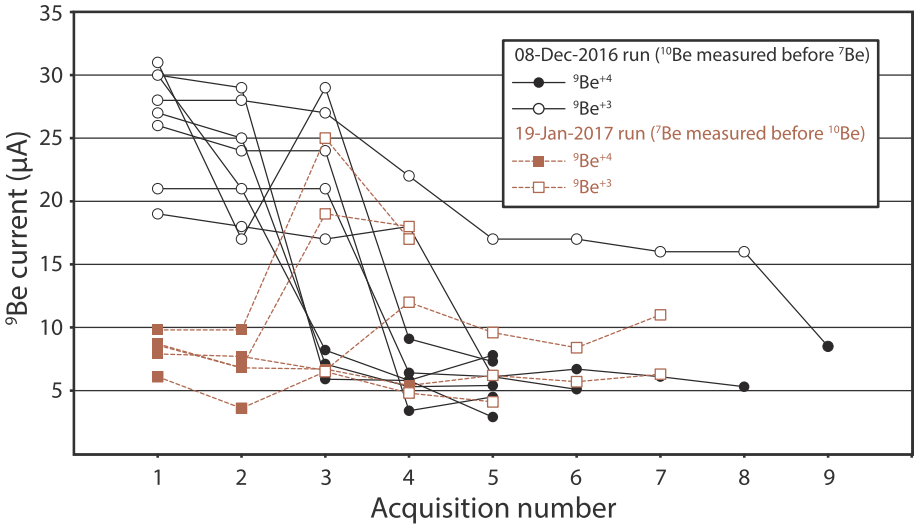


Fig. 4. Evolution of average ^{9}Be currents for all experimental targets where measurements were made for both ^{10}Be and ^{7}Be. $^{9}Be^{+3}$, selected for ^{10}Be measurements, produces approximately 3x the current of $^{9}Be^{+4}$, selected for ^{7}Be measurements. Order of isotopic measurement was reversed between the December and January runs.

(2) Sample sizes of ~1 L for rainwater or snow were more than sufficient for measuring both ^{7}Be and ^{10}Be to 2–4% precision (even after a ~1 month hiatus between collection and measurement) while 40 mL of rainwater was sufficient to resolve both isotopes above background. The precipitation samples yielded $^{10}Be/^{7}Be$ values at collection time of 4.1, 1.7, and 1.4 for Rain-A, Rain-B, and Snow, respectively. These values are consistent with the range typical in precipitation [22,23].

(3) The samples of the exposed commercial sand (~5 g) and gravel (~10 g), had ^{7}Be concentrations of 810 and 24,000 atoms/g and ^{10}Be concentrations of 14,000 × 10^{4} and 467 × 10^{4} atoms/g, respectively. Although the samples were not from sources with a natural context, these results point toward the order of magnitude range in concentrations that can be encountered when sampling soils.

(4) In all samples, the levels of ^{10}Be were significantly greater than that of ^{7}Be (most notably so for the particulate samples). This suggests that the partitioning of acquisition time between the isotopes can be heavily weighted toward ^{7}Be to obtain similar counting statistics for both isotopes. This tactic may be especially useful when measuring soil samples because long meteoric accumulation times are likely to produce very high levels of ^{10}Be compared to ^{7}Be (as was observed in the sand and gravel samples) due to their very different half-lives (~1.4 Ma vs. 53 days).

3.3. Total system efficiency

We did not conduct a dedicated experiment to ascertain total system efficiency (S_{eff} = atoms counted/atoms in target x 100) for measuring ^{7}Be on the CAMS AMS system. This would require sputtering a series of characterized ^{7}Be targets to exhaustion as was previously done for ^{10}Be at CAMS [12]. However, we can estimate S_{eff} for ^{7}Be in two ways.

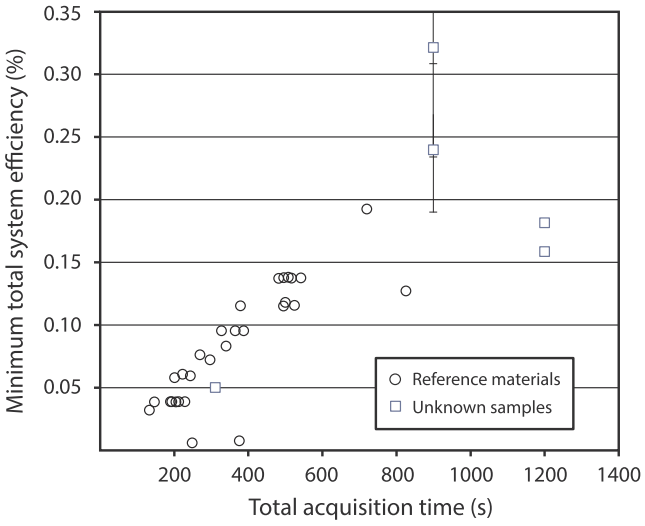


Fig. 5. Estimate of minimum total system efficiency (gated counts/^{7}Be atoms in target) plotted against acquisition time for all targets with measureable quantities of ^{7}Be. Targets were not run to exhaustion, so no maximum or average efficiency can be calculated. Error bars are 1σ and are too small to be shown for most targets.

(1) S_{eff} for routine ^{10}Be measurement at CAMS is ~1.2% [12]. This estimate does not include the efficiency loss through post-stripping $^{10}Be^{3+}$ to $^{10}Be^{4+}$, which was an additional implementation for our

Table 4
^{7}Be and ^{10}Be concentrations calculated in test samples.

Sample ID	Collection date	Mass (g)	^{9}Be spike (μg)	[^{10}Be] (×10^{4} atoms g^{-1}) μ ± σ		[^{7}Be] at t_m (×10^{2} atoms g^{-1}) μ ± σ		^{7}Be at t_m (×10^{3} atoms)	t_m-t_c[a] (days)	[^{7}Be] at t_c (×10^{2} atoms g^{-1}) μ ± σ	
08-Dec-2016 run											
TapH20	30-Oct-16	39.75	345	DL	DL	DL	DL		40	DL	DL
Rain-A	30-Oct-16	39.73	348	0.144	0.012	2.1	1.2	8.3	40	3.5	2.1
Sand	29-Nov-16	5.26	350	1400	20	7.1	2.7	3.7	10	8.1	3.1
Gravel	29-Nov-16	10.08	353	467	7	211	12	213	10	240	14
19-Jan-2017 run											
Rain-B	15-Dec-16	1039	299	0.236	0.008	8.45	0.35	878	36	13.5	0.6
Snow	23-Dec-16	984	300	1.19	0.03	60.9	1.2	5993	28	87.7	1.7

[a] t_m = time of measurement; t_c = time of collection.

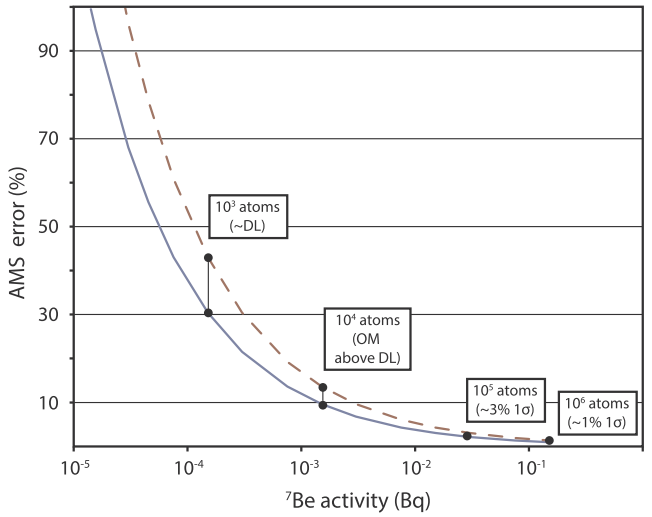


Fig. 6. Expected AMS counting statistics vs. activity assuming 1.07% (solid blue line) and 0.5% (dashed red line) total system efficiency. CAMS ^{7}Be detection limit (DL) is ~1000 atoms with a recommended sample minimum of 10,000 atoms—one order of magnitude (OM) above the DL. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

experimental settings. However, in both configurations $^{9}Be^{+3}$ is measured in the same off-axis Faraday cup immediately upstream of the post-stripper (Table 1). Thus, S_{eff} for $^{10}Be^{+4}$ can be inferred from the change in the gated ^{10}Be count rate between routine and experimental settings. The change in count rate can be estimated from raw $^{10}Be/^{9}Be$ measurements (in gated ^{10}Be counts per nC ^{9}Be) of our primary ^{10}Be standard (07KNSTD3110: $^{10}Be^{+3}/^{9}Be^{+3} = 0.0058$; $^{10}Be^{+4}/^{9}Be^{+3} = 0.0052$). Thus, for $^{10}Be^{+4}$, $S_{eff} = 0.0052/0.0058*1.2\%$, which is ~1.08%. If we assume that the post-stripping efficiency of stripping $^{7}Be^{+3}$ to $^{7}Be^{+4}$ is similar, then this estimate for S_{eff} would also apply to $^{7}Be^{+4}$.

(2) We can determine minimum $^{7}Be^{+4}$ S_{eff} values for all ^{7}Be reference material targets since we know the total atoms present in these target at time of measurement (note: we can also do this for our test sample targets, but this value includes an uncharacterized chemical yield efficiency and would therefore be less accurate). We consider these values far-limiting minima since none of the targets were exhausted for ^{7}Be. In Fig. 5 we plot minimum S_{eff} values against acquisition time and infer that, for the reference targets, we averaged ~0.15% ^{7}Be counting efficiency in the first 600 s. Fig. 2 of [12] reports the cumulative ^{10}Be counts per time for exhausted targets—targets that are both identical in composition to our experiment and sputtered in the same source. In that experiment they report ~25,000 ^{10}Be counts out of the exhausted total of 180,000 in the first 600 s of acquisition. Thus, for BeO-Nb targets in the CAMS source, ~14% of the total counting is accomplished in the first 600 s. Based on this observation, the empirical S_{eff} for $^{7}Be^{+4}$ should be 0.15%/14% = 1.07%. This estimate agrees well with the independent estimate made from assuming similar post-stripping yields for $^{7}Be^{+4}$ and $^{10}Be^{+4}$. Thus, our best estimate for total system efficiency for measuring ^{7}Be with the CAMS AMS system is ~1.07%. We intend to verify this in later experiments.

Fig. 6 plots the expected AMS counting uncertainty as a function of ^{7}Be activity assuming $S_{eff} = 1.07\%$ and $S_{eff} = 0.5\%$. These values provide an illustration of both an estimate for both the best possible statistics that can be obtained for a ^{7}Be sample at CAMS (calculated above) and also a conservative low-endmember estimate that reflects expected precisions for unknowns, given fair to poor beryllium yields through the chemistry and realistic target performance. This reduction in performance is because (1) targets are almost never run to exhausting during data acquisition, (2) efficiency is lost during target warm-up before data acquisitions (120 s initial burn-in, plus 20 s each for additional acquisitions), and (3) optional measurement of ^{10}Be on the same target will directly cut into ^{7}Be efficiency—although this is mitigated by the typically much higher abundance of ^{10}Be in meteoric samples such that equivalent ^{10}Be precision can be obtained from a relatively small fraction of the target material.

4. Conclusion

(1) A method for measuring ^{7}Be via AMS has been established at CAMS with a total system efficiency of just over 1% and a background detection limit of ~1000 atoms. This capability permits rapid measurement of ^{7}Be (10–20 min/sample) and is two orders of magnitude more sensitive than standard decay counting techniques. This allows a significant downscaling of sample size while greatly increasing sample throughput. Furthermore, this method is compatible with existing ^{10}Be measurement protocols at CAMS such that both isotopes can be measured on the same target with no additional preparation.

(2) We created a ^{7}Be reference material following the method of [11]. This method produced a material free of ^{10}Be and sufficient for use in AMS systems.

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