A SIMPLIFIED *IN SITU* COSMOGENIC ^{14}C EXTRACTION SYSTEM

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ABSTRACT. We describe the design, construction, and testing of a new, simplified *in situ* radiocarbon extraction system at the University of Arizona. Blank levels for the new system are low ($(234 \pm 11) \times 10^3$ atoms ($1\ \sigma$; $n = 7$)) and stable. The precision of a given measurement depends on the concentration of ^{14}C, but is typically <5% for concentrations of 100×10^3 atoms g^{-1} or more. The new system is relatively small and easy to construct, costs significantly less than the original *in situ* ^{14}C extraction system at Arizona, and lends itself to future automation.

INTRODUCTION

Like other *in situ* cosmogenic nuclides (CNs), *in situ* cosmogenic radiocarbon ($t_{1/2} = 5.73$ ka; *in situ* ^{14}C) is produced in the upper few meters of the Earth's crust by the interaction of cosmic rays with target nuclei, primarily O and Si (Gosse and Phillips 2001). The potential for using *in situ* ^{14}C to determine exposure ages of Holocene landforms and quantify erosion rates in rapidly denuding areas is widely recognized (e.g. Lal 1991; Gosse et al. 1996). Also recognized is the potential for *in situ* ^{14}C to be used in concert with longer-lived and stable CNs to elucidate complex exposure histories of surficial landforms (e.g. Miller et al. 2006). Despite its potential, however, the impact of *in situ* ^{14}C on CN research to date has been limited primarily because of the difficulty of measuring *in situ* ^{14}C concentrations in terrestrial rocks.

In situ ^{14}C has been measured routinely in lunar material and meteorites for decades (Goel and Kohman 1962; Jull et al. 1998), but ^{14}C concentrations in terrestrial rocks are typically 2–3 orders of magnitude lower. Measuring the *in situ* ^{14}C inventory of terrestrial rocks, therefore, requires complete removal of contaminant ^{14}C from sources such as CO_2 and other atmospheric gases, organic matter, and secondary carbonate, which are often present in quantities equivalent to or exceeding the *in situ* component.

An early attempt to isolate *in situ* ^{14}C from terrestrial silicates (Lal and Jull 1994) achieved some success, but assumed that ^{14}C exists largely as ^{14}CO within a rock and that the ratio of ^{14}CO:$^{14}CO_2$ derived from sample dissolution in hydrofluoric acid was constant. However, the measured ^{14}CO:$^{14}CO_2$ ratio varied by a factor of 3–4 between samples, limiting the actual precision of a given measurement. Handwerger et al. (1999) took a similar approach in attempting to isolate *in situ* ^{14}C from carbonates, and encountered many of the same problems. Lifton et al. (2001) developed an extraction system and protocols that were used successfully to isolate and extract *in situ* ^{14}C from terrestrial quartz, which includes melting grains using a lithium metaborate ($LiBO_2$) flux and oxidizing liberated carbon to CO_2. Yokoyama et al. (2004) isolated *in situ* ^{14}C from quartz through step-wise heating and using a mixture of carrier gases (He-O_2-CO-CO_2). Naysmith (2007) constructed an extraction system similar to the original Arizona system and followed the Lifton method protocols with minor modifications. Finally, Hippe et al. (2009) developed an extraction system that uses an electron bombardment furnace capable of reaching temperatures of 1550–1600 °C, which allows the melting of quartz grains without the use of a fluxing agent.

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Proceedings of the 20th International Radiocarbon Conference, edited by A J T Jull
RADIOCARBON, Vol 52, Nr 2–3, 2010, p 1236–1243

At Arizona, we set out to build upon the success of Lifton et al. (2001) by designing and constructing a simplified extraction system that is smaller and cheaper to build than the original, and by altering our laboratory procedures to reduce the amount of time required for an extraction. While the Lifton method is analytically difficult and time-intensive, it avoids the problem of variable CO:CO_2 ratios in rocks, and has resolved problems with unstable background levels and reproducibility that have undermined other attempts at measuring *in situ* ^{14}C (Lal and Jull 1994; Handwerger et al. 1999). Below, we discuss our modified extraction system and analytical procedures in detail. We present our results in full so that other laboratories can avoid the pitfalls and obstacles that we encountered.

MODIFICATIONS OF THE *IN SITU* ^{14}C EXTRACTION SYSTEM AND PROCEDURES

The original extraction system at the University of Arizona described in Lifton et al. (2001), which is still in use, is a closed loop that consists of 4 major components: (1) a Thermolyne® Type 59300 tube furnace capable of temperatures >1500 °C fitted with a dedicated 5 cm o.d. mullite tube; (2) a U-shaped secondary furnace packed with 2-mm-diameter fused quartz beads set at 1020 °C; (3) a cryogenic coil trap; and (4) a dual-stage stainless-steel (SS) bellows recirculating pump (Figure 1a). The recirculating pump forces ultra-high-purity (UHP) O_2 through the secondary furnace where all carbon species liberated from a sample are fully oxidized to CO_2. Condensable gases, including CO_2, are collected with liquid nitrogen (LN) in the coil trap. The O_2 carrier gas is continuously recirculated through the closed loop to ensure that all ^{14}C atoms are oxidized and ultimately captured.

A single extraction, which formerly took ~26 hr over a period of 3 days as described in Lifton et al. (2001), can now be completed in ~18 hr over 2 days (similar to Naysmith 2007). On the first day, a quartz sleeve (replaced for every sample) is precleaned by heating it to >800 °C with a glassblower's torch, and centered in the furnace tube using clean stainless steel (SS) and quartz implements. The sleeve protects the main furnace tube from corrosive $LiBO_2$ vapors. Some 20.0 g of $LiBO_2$ are placed in a high-purity Al_2O_3 boat and slid into the center of the quartz sleeve using a precleaned quartz push rod. The extraction system is then evacuated at 170 °C for 1.5 hr to remove sorbed atmospheric gases (mostly water) until the system pressure stabilizes at $<1.3 \times 10^{-4}$ kPa. The $LiBO_2$ is subsequently melted and degassed at 1200 °C for 1 hr in an UHP O_2 atmosphere that is recirculated through a liquid nitrogen (LN) trap to remove any contaminants. The LN trap is isolated from the recirculating system and evacuated to background pressures as the furnace is allowed to cool to room temperature overnight.

On the second day, the Al_2O_3 boat with the solidified $LiBO_2$ is retrieved from the furnace and placed in a laminar flow hood, using only precleaned quartz implements (no SS). The sample grains are loaded on top of the $LiBO_2$, and the boat is returned to the furnace tube. After evacuating the extraction system at 170 °C for 1 hr, the sample is heated in 6.7 kPa of UHP O_2 at 500 °C for 1 hr to remove atmospheric contaminants. The contaminants are removed from the LN trap, which is isolated from the recirculating system, evacuated to background pressures, refilled with 6.7 kPa of UHP O_2, and then reopened to the recirculating system. The sample is then oxidized in UHP O_2 at 1100 °C for 3 hr.

The CO_2 is purified (i.e. water, halides, nitrogen, and sulfur species are removed) separately from the extraction loop (but on the same vacuum system), while the CO_2 is converted to graphite in a separate, dedicated graphitization system. Until 2001, we used BOC-Edwards Diffstak® oil diffusion pumps backed by oil-cooled rotary vane pumps to evacuate each system. These have since been replaced with turbomolecular pumps backed by dry scroll pumps to eliminate the possibility of contaminating the lines with backstreaming oil vapors.

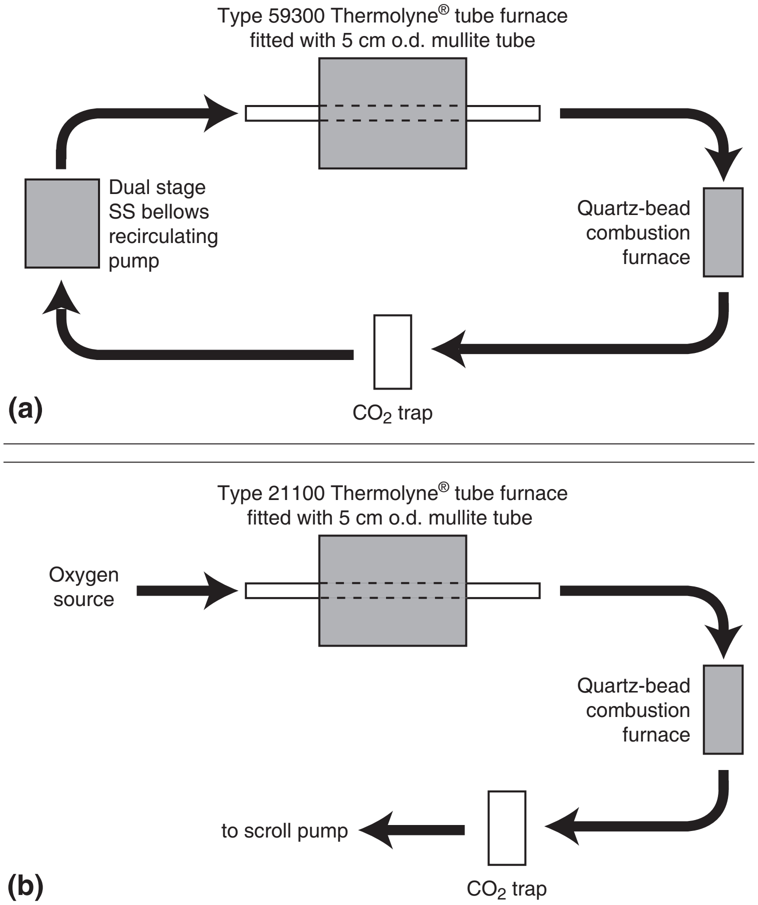

Figure 1 Schematic diagrams of (a) the original *in situ* ^{14}C extraction line (after Lifton et al. 2001) and (b) the modified extraction line in our laboratory at the University of Arizona. Dark arrows show the flow path of O_2 carrier gas, which is kept at pressures equivalent to ~6.7 kPa at 170 °C. Carbon species liberated from a sample are fully oxidized to CO_2 in the quartz-bead furnace and collected in the CO_2 trap. Contaminant gases are subsequently removed, the sample CO_2 is diluted with ^{14}C-free CO_2, and converted to graphite for analysis by AMS.

While the existing extraction system works well with the new procedures, extracting *in situ* ^{14}C from quartz with it is still very time- and labor-intensive. Our intent in modifying this design was to make an extraction system that was as small, inexpensive, and simple as possible, with the idea that ultimately multiple extraction lines could be operated via automation to maximize sample throughput. In the new system, we eliminated the recirculating pump and moved the purification apparatus to a separate vacuum system. These changes required several modifications to the extraction procedures discussed above. The primary difference is that the O_2 carrier gas now moves through the modified extraction line in a single pass, rather than being continually recirculated. We control the rate at which the gas moves through the line by opening various stopcocks very slightly. After experimenting with different flow rates, we settled on the equivalent of 2–3 times the volume of the furnace tube (at 6.7 kPa pressure) over a 1-hr period, although flow rates up to ~50% higher or lower than this do not appear to affect our results.

The modified extraction system consists of 3 major parts: (1) a Thermolyne® Type 79300 tube furnace capable of reaching 1200 °C, fitted with a dedicated 5 cm o.d. mullite tube; (2) a secondary furnace as described above; and (3) a single cryogenic coil trap (Figure 1b). The mullite tube is capped with 316 SS compression flanges and glass frits are installed at both ends of the furnace chamber beyond the flanges to protect the rest of the line from particulates. These frits also slow the movement of gases through the mullite tube, minimizing the possibility of disturbing the sample during extraction. UHP O_2 is bled through the furnace tube and carries evolved sample gases through the secondary furnace before condensable gases are collected in the coil trap. Sample purification and graphitization are conducted separately as before. All glass tubing (except the coil trap) and glass-metal connections are covered by heat tape that is maintained at 50–70 °C to minimize gas adsorption inside the line (Figure 2).

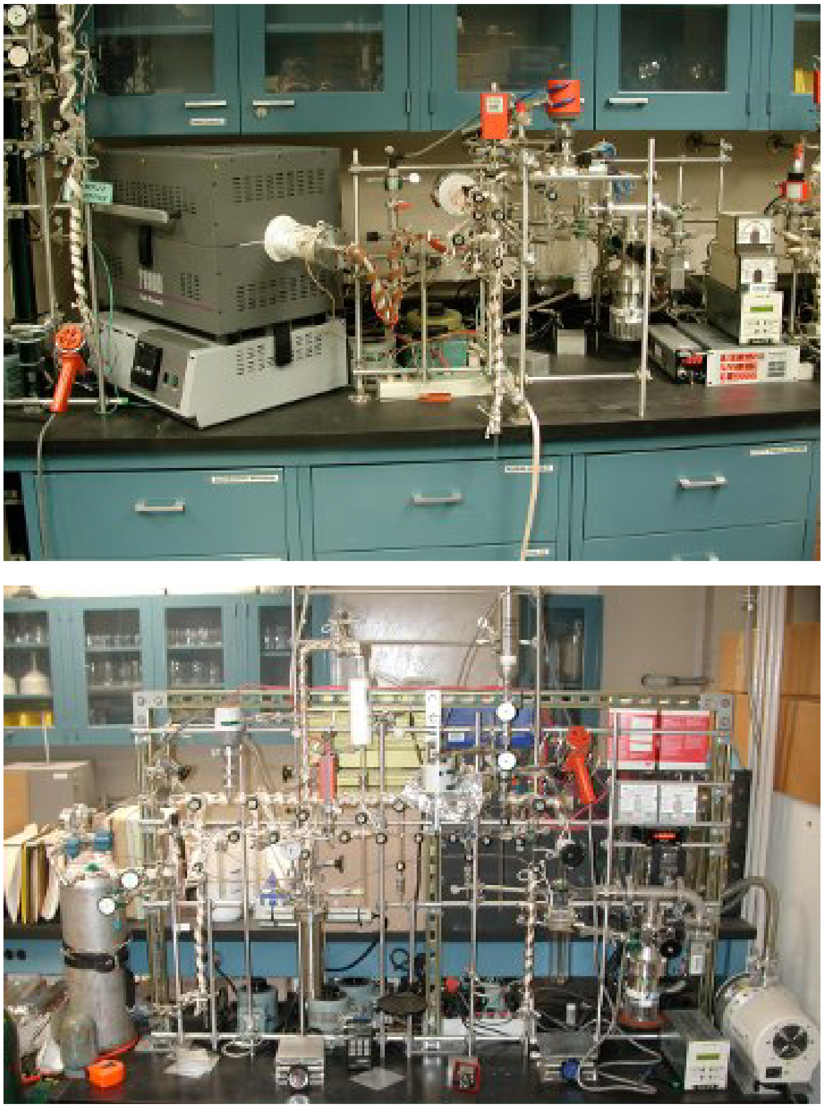

Figure 2 Photographs of the modified extraction line (upper panel) and CO_2 purification line (lower panel). All glass tubing (except coil traps and cold fingers) and glass-metal connections are covered by heat tape that is maintained at 50–70 °C to minimize gas adsorption inside the line.

Yield Test, Blank Levels, and Reproducibility

An initial concern with our modified system was that the single-pass design might not effectively trap the CO_2 evolved from a sample before it was pumped away with the O_2 carrier gas. To address this issue, a multiple-step yield test was conducted over a period of 5 days. The experiment began on day 1 with the degassing of the Al_2O_3 boat and $LiBO_2$ powder. On days 2 and 3, blank measurements were made to establish a trend in the blank levels because the blank and *in situ* ^{14}C concentration of a sample cannot be measured simultaneously (the blank value for a given extraction must be inferred or interpolated). At the beginning of each day, the temperature inside the furnace tube was raised to 170 °C and evacuated for 1 hr. The tube was then filled with ~6.7 kPa of UHP O_2, and the temperature was raised to 1100 °C. The carrier gas was then slowly bled through the system for 1 hr, and condensable gases were collected in the coil trap. The tube was allowed to cool to room temperature overnight between each step. On day 4, a small aliquot (64 μL) of CO_2 was purified, and cryogenically introduced into the furnace tube from a small cold finger immediately adjacent to the tube's downstream end. Approximately 6.7 kPa of UHP O_2 was then introduced into the tube, and allowed to mix with the CO_2 aliquot for 1 hr at 1100 °C. UHP O_2 was then bled through the system for 1 hr, and condensable gases were collected in the coil trap. An additional blank measurement was conducted on day 5 so that we could interpolate a blank level for day 4. All gases that were collected were later purified and the CO_2 yield was measured. The results indicated essentially 100% recovery of CO_2 (Table 1).

Table 1 Results of yield test on modified system.[a]

Step	Temperature and duration	Sample volume (μL)
Abbreviation in Equation 1		V_s
blank - day 2	1100 °C; 1 hr	6.2 ± 1.1
blank - day 3	1100 °C; 1 hr	3.0 ± 1.1
blank - day 4	interpolated	1.9 ± 1.1
initial CO_2	—	63.9 ± 1.4
blank - day 5	1100 °C; 1 hr	1.4 ± 1.1
total recovered CO_2	1100 °C; 1 hr	64.7 ± 1.2
recovered CO_2 - blank CO_2	—	62.8 ± 1.6
Recovery (%)		**98.2 ± 2.5**

[a] All uncertainties are given at the 1-σ level.

Blank levels for standard 2-day extractions for both systems are low and stable (Table 2). Blanks for the original and modified extraction systems have a weighted mean of $(141 \pm 13) \times 10^3$ atoms (1 σ; $n = 6$) and $(234 \pm 11) \times 10^3$ atoms (1 σ; $n = 7$), respectively. Although the blank levels for the modified extraction line are higher than those for the original line, the limiting factor in a given measurement is not the blank itself, but rather the uncertainty in the blank, which is similar for both systems.

Two series of blank measurements for step-heating experiments with the modified extraction line yielded a weighted mean total blank of $(232 \pm 19) \times 10^3$ atoms (1 σ; $n = 2$), which is indistinguishable from the weighted mean of the standard 2-day blanks on the modified line.

Our final test was to determine if we could reproduce the measured concentration of *in situ* ^{14}C in a "known" sample in the modified line relative to the total measured using the original extraction system. For this, we used a sample of quartzite (PP-4) that is used as an internal standard in our laboratory at Arizona. This sample was collected from a wave-cut bench formed during the late Pleistocene highstand of pluvial Lake Bonneville (Oviatt et al. 1992; Lifton et al. 2001). All aliquots were treated and extracted in the same manner to facilitate direct comparison.

Table 2 Summary of blank data.[a]

Lab #	Sample #	System	Temperature and duration	Fraction modern[b]	Sample volume (µL)	Diluted volume[c] (mL)	^{14}C yield (10^3 atoms)
Abbreviations in Equation 1				F_m	V_s	V_s	N
			Extraction blanks				
RN-632	AA-41990	original	1100 °C; 3 hr	0.0024 ± 0.0003	7.62 ± 0.07	1.26 ± 0.01	94 ± 14
RN-635	AA-41990	original	1100 °C; 3 hr	0.0032 ± 0.0003	9.70 ± 0.74	1.22 ± 0.01	125 ± 13
RN-636	AA-41990	original	1100 °C; 3 hr	0.0049 ± 0.0003	11.62 ± 0.80	1.21 ± 0.01	185 ± 13
RN-647	AA-41990	original	1100 °C; 3 hr	0.0035 ± 0.0005	10.82 ± 0.44	1.07 ± 0.01	119 ± 18
RN-649	AA-41990	original	1100 °C; 3 hr	0.0034 ± 0.0003	9.98 ± 0.26	1.36 ± 0.01	145 ± 11
RN-650	AA-41990	original	1100 °C; 3 hr	0.0047 ± 0.0003	12.25 ± 0.92	1.09 ± 0.01	161 ± 12
						Weighted mean	**141 ± 13**
RN-682	AA-48353	modified	1100 °C; 3 hr	0.0048 ± 0.0004	18.79 ± 0.95	1.76 ± 0.01	265 ± 24
RN-684	AA-48353	modified	1100 °C; 3 hr	0.0052 ± 0.0007	18.43 ± 0.66	1.75 ± 0.01	285 ± 40
RN-686	AA-48353	modified	1100 °C; 3 hr	0.0040 ± 0.0003	13.49 ± 0.82	1.72 ± 0.01	220 ± 19
RN-698	AA-48353	modified	1100 °C; 3 hr	0.0053 ± 0.0003	9.78 ± 0.74	1.58 ± 0.01	265 ± 17
RN-705	AA-48353	modified	1100 °C; 3 hr	0.0051 ± 0.0012	9.94 ± 1.10	1.46 ± 0.01	237 ± 57
RN-716	AA-48353	modified	1100 °C; 3 hr	0.0054 ± 0.0004	9.93 ± 1.14	1.40 ± 0.01	240 ± 19
RN-750	AA-48353	modified	1100 °C; 3 hr	0.0043 ± 0.0003	11.91 ± 1.54	1.53 ± 0.01	207 ± 12
						Weighted mean	**234 ± 11**
			Step-heated blanks				
RN-720a	AA-48353	modified	300 °C; 1 hr	0.0001 ± 0.0005	1.39 ± 1.02	1.19 ± 0.01	4 ± 20
RN-720b	AA-48353	modified	500 °C; 1 hr	0.0000 ± 0.0003	1.61 ± 1.22	1.14 ± 0.01	0 ± 13
RN-720c	AA-48353	modified	600 °C; 1 hr	0.0000 ± 0.0003	1.16 ± 1.06	1.20 ± 0.01	0 ± 10
RN-720d	AA-48353	modified	700 °C; 1 hr	0.0006 ± 0.0003	0.69 ± 1.14	1.23 ± 0.01	22 ± 9
RN-720e	AA-48353	modified	900 °C; 1 hr	0.0006 ± 0.0002	1.38 ± 1.22	1.15 ± 0.01	22 ± 7
RN-720f	AA-48353	modified	1100 °C; 1st hr	0.0025 ± 0.0003	2.08 ± 1.22	1.17 ± 0.01	95 ± 13
RN-720g	AA-48353	modified	1100 °C; 2nd hr	0.0018 ± 0.0003	2.08 ± 1.06	1.22 ± 0.01	68 ± 13
RN-720h	AA-48353	modified	1100 °C; 3rd hr	0.0006 ± 0.0003	2.08 ± 1.14	1.18 ± 0.01	22 ± 10
					Total (600–1100 °C steps)		**229 ± 26**
RN-743a	AA-48353	modified	300 °C; 1 hr	0.0006 ± 0.0003	0.70 ± 0.66	1.71 ± 0.01	32 ± 14
RN-743b	AA-48353	modified	500 °C; 1 hr	0.0009 ± 0.0003	1.63 ± 0.74	1.67 ± 0.01	46 ± 14
RN-743c	AA-48353	modified	600 °C; 1 hr	0.0000 ± 0.0002	0.93 ± 0.90	1.60 ± 0.01	0 ± 9
RN-743d	AA-48353	modified	700 °C; 1 hr	0.0000 ± 0.0002	0.93 ± 0.90	1.63 ± 0.01	0 ± 10
RN-743e[d]	AA-48353	modified	900 °C; 1 hr	—	—	—	22 ± 7
RN-743f	AA-48353	modified	1100 °C; 1st hr	0.0021 ± 0.0003	3.26 ± 0.74	1.56 ± 0.01	101 ± 13
RN-743g	AA-48353	modified	1100 °C; 2nd hr	0.0019 ± 0.0002	2.12 ± 0.25	1.53 ± 0.01	94 ± 9
RN-743h	AA-48353	modified	1100 °C; 3rd hr	0.0004 ± 0.0003	2.36 ± 0.17	1.50 ± 0.01	19 ± 16
					Total (600–1100 °C steps)		**236 ± 27**

[a] All uncertainties are given at the 1-σ level.

[b] Fraction modern values are referenced to the NIST oxalic acid I standard (Stuiver and Polach 1977).

[c] CO_2 extracted from each sample was diluted with ^{14}C-free CO_2 before conversion to graphite.

[d] Sample lost during purification; assumed same results as RN-720e.

The concentration of *in situ* ^{14}C in each extraction (given in units of atoms g^{-1}) was calculated using Equation 1:

$$N = \frac{\left(\frac{F_m A_{14} N_A V_S}{V_A} - B\right)}{M_s} \tag{1}$$

where F_m is the measured fraction modern carbon normalized to a δ^{13}C value of −25‰ $_{\mathrm{VPDB}}$ and AD 1950, A_{14} is the fractional abundance of modern carbon (1.177×10^{-12} ^{14}C atoms per ^{12}C atom), N_A is Avogadro's number, V_S is the volume of CO_2 at STP (in cm^3), V_A is the volume of 1 mol of CO_2 at STP (in cm^3), B is the number of ^{14}C atoms associated with a blank extracted in the same manner as the sample, and M_s is the mass of the sample (in grams). After measuring the volumetric yield, the extracted CO_2 was diluted with ^{14}C-free CO_2 to a total mass of 1 to 2 mL (0.5 to 1 mg C equivalent) for subsequent conversion to graphite.

Following Lifton et al. (2001), measured concentrations of *in situ* ^{14}C were normalized to production at the ground surface, open-sky conditions (i.e. no topographic shielding), and a pure SiO_2 composition using experimental cross-section data (Jull et al. 1998) and ground-level neutron spectrum measurements (Gordon et al. 2004). The magnitudes of these corrections for sample PP-4 were 7.6%, 0.6%, and 0.3%, respectively. The weighted means of the corrected *in situ* ^{14}C concentrations using the original ($(360 \pm 13) \times 10^3$ atoms g^{-1}; 1 σ; $n = 3$) and modified ($(354 \pm 10) \times 10^3$ atoms g^{-1}; 1 σ; $n = 6$) extraction systems are indistinguishable at the 1-σ level (Table 3).

Table 3 Summary of data for sample PP-4.[a]

Lab #	Sample #	System	Fraction modern[b]	Sample volume (μL)	Diluted volume[c] (mL)	^{14}C yield[d] (10^3 atoms g^{-1})
Abbreviations in Equation 1			F_m	V_s	V_s	N
RN-674	AA-34075	original	0.0480 ± 0.0008	47.90 ± 1.35	1.19 ± 0.01	359 ± 23
RN-690	AA-34075	original	0.0342 ± 0.0006	46.74 ± 0.39	1.71 ± 0.01	369 ± 24
RN-691	AA-34075	original	0.0366 ± 0.0005	46.22 ± 0.70	1.54 ± 0.01	353 ± 23
RN-758a	AA-34075	original	0.0023 ± 0.0003	5.12 ± 0.82	1.34 ± 0.01	21 ± 5
RN-758b	AA-34075	original	0.0410 ± 0.0005	49.17 ± 0.97	1.36 ± 0.01	331 ± 37
RN-758[e]	AA-34075	original	—	—	—	352 ± 37
					Weighted mean	**359 ± 13**
RN-661	AA-61595	modified	0.0401 ± 0.0009	46.88 ± 1.16	1.44 ± 0.01	343 ± 23
RN-662	AA-61595	modified	0.0412 ± 0.0009	48.77 ± 1.12	1.42 ± 0.01	350 ± 23
RN-665	AA-61595	modified	0.0454 ± 0.0018	45.27 ± 1.16	1.29 ± 0.01	350 ± 27
RN-687	AA-61595	modified	0.0360 ± 0.0006	54.49 ± 0.59	1.75 ± 0.01	381 ± 25
RN-704	AA-61595	modified	0.0369 ± 0.0005	49.59 ± 1.01	1.54 ± 0.01	339 ± 22
RN-748	AA-61595	modified	0.0461 ± 0.0006	51.08 ± 1.08	1.27 ± 0.01	367 ± 24
					Weighted mean	**354 ± 10**

[a] All uncertainties are given at the 1-σ level.

[b] Fraction modern values are referenced to the NIST oxalic acid I standard (Stuiver and Polach 1977).

[c] CO_2 extracted from each sample was diluted with ^{14}C-free CO_2 before conversion to graphite.

[d] *In situ* ^{14}C yields for sample PP-4 were normalized to production at the ground surface, open-sky conditions (no topographic shielding), and a pure SiO_2 composition using correction factors of 1.0764 ± 0.0104, 1.0064 ± 0.0474, and 1.0027 ± 0.0266, respectively.

[e] For sample RN-758, extractions were done at 600 °C for 1 hr (RN-758a) and 1100 °C for 3 hr (RN-758b). The sum of the 2 temperature steps was included in the weighted mean calculation.

CONCLUSIONS

The experimental data presented here summarize the testing of a new, simplified *in situ* ^{14}C extraction system. Blank levels for the new system are low ($(234 \pm 11) \times 10^3$ atoms (1 σ; $n = 7$)) and stable, and we are able to reproduce *in situ* ^{14}C concentrations in quartz for samples measured on the original system. The precision of a given measurement depends on the concentration of ^{14}C, but is typically <5% for concentrations of 100×10^3 atoms g^{-1} or more. While our extraction methods are analytically difficult and quite time-intensive, modification of the original Lifton method has reduced the amount of time required for an extraction from ~26 hr over 3 days to ~18 hr over 2 days. The simple design of the modified system lends itself to future automation, which is ongoing. The development of an automation system should allow us to control the extraction, CO_2 purification, and graphitization systems remotely without sacrificing precision or accuracy.

ACKNOWLEDGMENTS

We thank Rich Cruz and the Arizona AMS facility for their support. An early version of this manuscript benefited from constructive reviews from G Cook, J Schaefer, and an anonymous reviewer. The current version of this manuscript was improved by careful reviews by E Ellis, D Goodwin, J Rech, Y Yokoyama, and an anonymous reviewer.

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