Two ages of porphyry intrusion resolved for the super-giant Chuquicamata copper deposit of northern Chile by ELA-ICP-MS and SHRIMP

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ABSTRACT

Zircon U-Pb ages measured in situ by excimer laser ablation–inductively coupled plasma–mass spectrometry (ELA-ICP-MS) and verified by sensitive high-resolution ion microprobe (SHRIMP) on ore-bearing felsic porphyries from the Chuquicamata porphyry copper-molybdenum deposit, northern Chile, identify two discrete igneous events. The volumetrically dominant East porphyry has an age of 34.6 ± 0.2 Ma, whereas the Bench and West porphyries yield ages of 33.3 ± 0.3 Ma and 33.5 ± 0.2 Ma, respectively. The age of the East porphyry is indistinguishable from a Re-Os age for early molybdenite mineralization (35 Ma) and the oldest reported ^{40}Ar-^{39}Ar ages for hydrothermal alteration, confirming a genetic link with mineralization. Previous geological studies and ^{40}Ar-^{39}Ar and Re-Os geochronology identify two main hydrothermal events: high-temperature potassic alteration with chalcopyrite at 33.4 ± 0.3 Ma followed by lower temperature quartz-sericite alteration with pyrite at 31.1 ± 0.3 Ma. The ages of the West and Bench porphyries match the ages for potassic alteration. Younger quartz-sericite alteration may reflect an additional fourth intrusion concealed at depth. The anomalously large size of Chuquicamata appears to be due to a protracted igneous history resulting in the superposition of at least two temporally distinct magmatic-hydrothermal systems.

Keywords: Chuquicamata, porphyry copper, geochronology, Chile, zircon.

INTRODUCTION

The Chuquicamata porphyry Cu-Mo deposit of northern Chile is historically the world's largest Cu producer and, 85 years after the start of mining, is still one of the principal sources of Cu (Ossandón and Zentilli, 1997). The purpose of this study was to test the hypothesis that Chuquicamata is a supergiant deposit because two or more distinct magmatic-hydrothermal systems have contributed to its formation. That is, Chuquicamata represents the superimposition of multiple porphyry copper deposits.

Previous studies have found evidence for multiple magmatic-hydrothermal events at Chuquicamata. Geological mapping has shown that there are three texturally distinguishable productive intrusive units within the pit, the East, West and Bench porphyries (Aracena, 1981; Maksaev, 1990), suggesting that there may have been more than one period of igneous activity. This conclusion is supported by an extensive analysis of the alteration pattern at Chuquicamata (Lindsay et al., 1995), which does not show the classic concentric zonation of other porphyry ore deposits described by Lowell and Guilbert (1970). The irregular alteration pattern at Chuquicamata may have arisen through the multiple hydrothermal events documented by Lindsay et al. (1995), who reported quartz-sericite alteration superimposed on potassic alteration. The ^{40}Ar-^{39}Ar evidence (Reynolds et al., 1998) that high-temperature (>350 °C) potassic alteration is older (33.4 ± 0.3 Ma) than low-temperature (<350 °C) quartz-sericite alteration (31.1 ± 0.3 Ma) supports such an interpretation.

Although field studies provide evidence of multiple igneous and hydrothermal events, they do not distinguish whether the recognized events occurred during a single period of protracted magmatic-hydrothermal activity, or as part of two or more discrete igneous-hydrothermal events that were superimposed on each other. Despite the consistency of field evidence and Ar geochronology with the multiple porphyry hypothesis, they do not provide a definitive test. This can only be done through direct dating of the porphyries. Furthermore, because the geochemistry of the three types of porphyries is very similar and because they do not always show clear cross-cutting relationships, it is likely that the age difference between them is small. A dating technique that is both precise and resistant to thermal overprinting is required.

The porphyries were dated using excimer laser ablation–inductively coupled plasma–mass spectrometry (ELA-ICP-MS) zircon U-Pb geochronology. The accuracy of the results was confirmed by sensitive high-resolution ion microprobe (SHRIMP) analyses of the same samples. In situ zircon U-Pb analysis has two important advantages over alternative techniques for the present study. First, the U-Pb system in zircon is highly resistant to hydrothermal resetting. Second, both ELA-ICP-MS and SHRIMP are microbeam techniques that allow inherited cores, which are present in the Chuquicamata porphyries (Zentilli et al., 1994), to be identified and excluded from age calculations.

GEOLOGICAL SETTING

The Chuquicamata porphyry Cu-Mo mine is located ~240 km northeast of Antofagasta, in northern Chile (Fig. 1). The Precordillera basement and cover sequences in the Chuquicamata area comprise varied igneous, sedimentary, and metamorphic lithologies ranging in age between Ordovician and Holocene (Chong and Pardo, 1994). Eocene-Oligocene magmatism in the Chuquicamata district exploited weakened crust along the West fissure zone of strike-slip deformation (Lindsay et al., 1995; Reutter et al., 1996; Tomlinson and Blanco, 1997a, 1997b; Dilles et al., 1997; Reynolds et al., 1998). The West fissure forms part of the more extensive north-south Domeyko fault system (Maksaev and Zentilli, 1988) and juxtaposes noneconomic Fortuna Complex intrusions against ore-bearing Chuquicamata porphyries in the central part of the Chuquicamata pit.

The Chuquicamata deposit is at the southern end of the 12 by 2 km north-trending Chuquicamata Intrusive Complex (Chong and Pardo, 1994). This late Eocene–Oligocene igneous suite (Maksaev, 1990; Zentilli et al., 1994) consists of the three recognizable porphyritic units in the mine, distinguished by texture and different amounts of plagioclase, quartz, orthoclase, and biotite phenocrysts. These units are the East porphyry, a granodiorite to monzogranite that displays a close-packed texture, with medium to coarse grains

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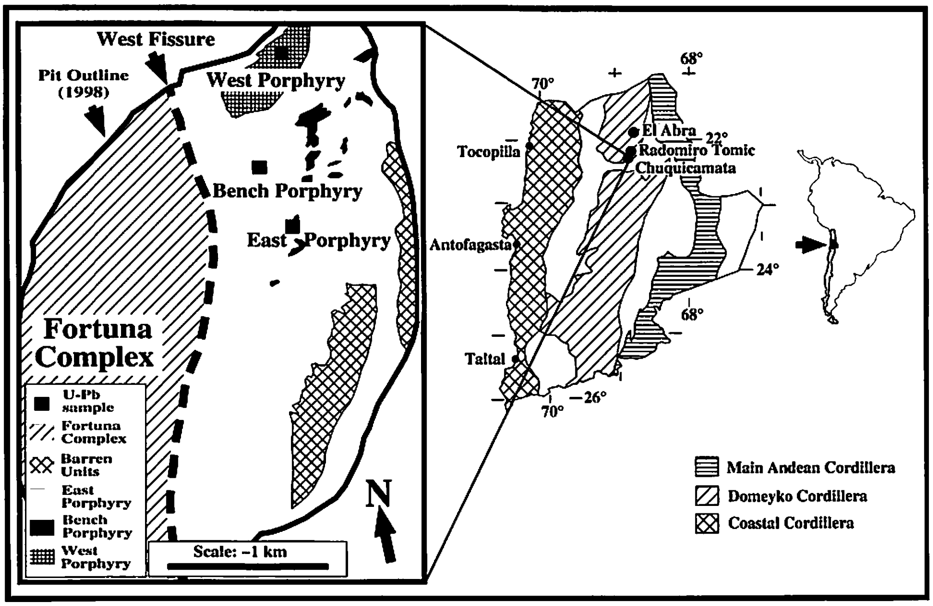

Figure 1. Location of Chuquicamata mine in Antofagasta region of northern Chile. Three core samples used in this study are marked on pit map (modified from Lindsay et al., 1995; Reynolds et al., 1998).

in scarce matrix; the West porphyry, a monzogranite to granodiorite that contains visibly spaced phenocrysts set in a saccaroidal matrix; and the Bench porphyry, a monzodiorite with a bimodal distribution of phenocryst size set in an aphanitic matrix (Aracena, 1981). Volumetrically, the East porphyry dominates the Chuquicamata Intrusive Complex, where there are only minor West and Bench porphyry occurrences. The West porphyry crops out in the north of the pit, but its boundaries have not been clearly delimited. The Bench porphyry crops out along the eastern flank of the mine as a series of north-trending dike-like bodies offset by later faults. East of the Chuquicamata Intrusive Complex, but within the mine, a poorly defined contact is made with the barren Elena Granodiorite (Lindsay et al., 1995) and East Granite.

METHOD

The zircons analyzed in this study were separated from 2 kg samples of each unit collected from drill cores (Fig. 1), mounted in epoxy and polished. Using cathodoluminescence and optical microscopy, care was taken to ensure that the least-fractured, inclusion-free zones in zircon were analyzed. Even so, both Pb loss and inheritance were encountered. Dating by ELA-ICP-MS followed a procedure modified from Horn et al. (2000,) as outlined in the Appendix. Data were acquired in three separate 8 h analytical sessions several weeks apart. Zircons from the three mine porphyries were analyzed in a rotation that included standard zircon SL13 (Claoué-Long et al., 1995) and standard silicate glass NIST SRM 612 (Pearce et al., 1997).

Zircon dating by SHRIMP followed well-established analytical procedures (Claoué-Long et al., 1995). Zircons from the three mine porphyry units and reference zircon AS3 ($^{206}Pb/^{238}U = 0.1859$; Paces and Miller, 1993) were analyzed in rotation during a single 22 h analytical session, making the measurement of any age differences independent of errors in Pb/U calibration (0.27%). Further details of the method are outlined in the Appendix.

RESULTS

The results are summarized in Tables 1 and 2 and individual spot analyses are available from the data repository.[1] The mean U-Pb zircon ages of individual intrusions differ slightly between the two analytical methods, the SHRIMP ages being 1.5%–2.5 % older. This most likely arises through a combination of the analytical biases specific to each instrument, and procedural differences, in particular the standards and methods used to correct for instrument drift, elemental and isotopic fractionation, and common lead. The SHRIMP Pb/U calibration used AS3, whereas the ELA-ICP-MS used silicate glass NIST 612 to monitor drift and U/Th fractionation, and zircon standard SL13 for Pb isotope, Pb/U, and Pb/Th fractionation. Despite the small differences in absolute age, both techniques are in close agreement in terms of the relative age difference between the three porphyries. The results of this study show that it is now possible to date Tertiary zircon by ELA-ICP-MS with an aggregate precision and accuracy that is comparable with SHRIMP, a significant advance on previous ICP-MS studies (cf. Compston, 1999).

DISCUSSION

Both SHRIMP and ELA-ICP-MS resolve two igneous events at Chuquicamata (Fig. 2). The oldest and volumetrically dominant intrusion, the East porphyry, yields individual grain ages by both techniques that are slightly more scattered than expected from analytical uncertainty, reflecting either a small age spread, or more likely, some Pb loss (Fig. 3). The mean ELA-ICP-MS $^{206}Pb/^{238}U$ age of 34.6 ± 0.2 Ma correlates well with the highest ^{40}Ar-^{39}Ar ages of Reynolds et al. (1998) and a Re-Os molybdenite age of 34.8 ± 0.2 Ma (Mathur et al., 2000; J. Ruiz, 2000, personal commun.) for early mineralization (Fig. 4). Intrusion of the Bench and West porphyries took place between 0.9 to 1.5 m.y. later, at 33.3 ± 0.3 and 33.5 ± 0.2 Ma, respectively. The difference in measured age between the oldest and youngest intrusions is significant at the 95% confidence level. The ages of the Bench and the West porphyries cannot be resolved by either analytical technique (Fig. 2), nor are the two units distinguishable geochemically. They are texturally different, however, which in the past has been used as the basis to separate them. The new U-Pb data suggest that the Bench and West porphyries represent contemporaneous intrusions from a common magma source, both >1 m.y. younger than the East porphyry. This second period of porphyry intrusion can be correlated with the period of potassic alteration documented by Reynolds et al. (1998) that has an average ^{40}Ar-^{39}Ar age of 33.4 ± 0.3 Ma. Lower ^{40}Ar-^{39}Ar ages of 31.1 ± 0.3 Ma for quartz-sericite alteration (Reynolds et al., 1998) and a Re-Os isochron

TABLE 1. $^{206}Pb/^{238}U$ AGES FOR MINE PORPHYRIES

	No. of analyses	SHRIMP Age* (Ma)	MSWD	No. of analyses	ELA-ICP-MS Age (Ma)	MSWD
East	15	35.2 ± 0.4	2.77	68	34.6 ± 0.2	3.29
West	10	34.0 ± 0.3	1.00	73	33.5 ± 0.2	1.96
Bench	13	34.1 ± 0.3	1.15	83	33.3 ± 0.3	7.92

Note: The SHRIMP (sensitive high-resolution ion microprobe) uncertainties include Pb/U calibration, but the ELA-ICP-MS (excimer laser-ablation–inductively coupled plasma–mass spectrometry) do not.
*All uncertainties are 95% confidence limits.

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[1] GSA Data Repository item 2001042, SHRIMP U-Th-Pb isotopic data for zircon from the Chuquicamata copper deposit, is available from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, editing@geosociety.org, or at www.geosociety.org/pubs/ft2001.htm.

TABLE 2. MINE PORPHYRY AGES FROM THREE ELA-ICP-MS ANALYTICAL SESSIONS

	Day 1	MSWD	Day 2	MSWD	Day 3	MSWD
East*	34.5 ± 0.3	3.26	34.5 ± 0.4	3.64	34.4 ± 0.4	2.85
West*	33.8 ± 0.4	4.07	33.6 ± 0.3	1.46	33.6 ± 0.2	1.66
Bench	33.8 ± 0.4	3.01	33.0 ± 0.4	2.12	33.6 ± 0.4	2.25

Note: Ages are Ma. ELA-ICP-MS is excimer laser-ablation–inductively coupled plasma–mass spectrometry.

*Apparent small discrepancies between the mean ages for the East and West porphyries for the individual days and the pooled days are due to different grains being culled when data from only one analytical session are considered; see Data Repository material (footnote 1 in text).

age for associated pyrite at 31 ± 2 Ma (Mathur et al., 2000) indicate an even younger hydrothermal event.

The agreement between zircon U-Pb, molybdenite Re-Os, and the highest biotite ^{40}Ar-^{39}Ar ages in the East porphyry requires that this unit cooled from near 800 °C (the zircon crystallization temperature), through 500 ± 50 °C (the inferred closure temperature for Re-Os in molybdenite; Suzuki et al., 1996), to below 300 ± 50 °C (the closure temperature for Ar in biotite) in <0.5 m.y. Furthermore, the similarity between the U-Pb zircon ages of the Bench and West porphyries and the average ^{40}Ar-^{39}Ar age of potassic alteration suggests that these porphyries cooled from ~800 °C to below 300 °C in a comparable interval of time. This evidence suggests a cooling rate of at least 1000 °C/m.y. (Fig. 5), which is an order of magnitude faster than the rate estimated by Reynolds et al. (1998) based on the ^{40}Ar-^{39}Ar system. Note that neither cooling trajectory passes through the age of youngest sericitic alteration. We suggest that this youngest period of hydrothermal alteration may have been produced by a fourth, perhaps unexposed, igneous intrusion ca. 31.4 Ma. The extrapolation of the inferred youngest cooling curve passes through the He apatite closure temperature of 75 ± 25 °C ca. 31 Ma (McInnes et al., 1999), thus constraining all subsequent fluid-rock interactions in the deposit to very low temperatures.

The cooling rates inferred for the Chuquicamata porphyries agree well with the thermal calculations of Cathles et al. (1997) for kilometer-scale shallow-level intrusions. For example, a 2 km wide by 3 km tall igneous body emplaced 4 km below the surface will cool through the first 75% of its thermal history at an average rate of 5000 °C/m.y. by conduction alone. Hydrothermal circulation will result in convective cooling rates 2–10 times faster than this, depending on the permeability of the intrusion and its country rocks. Such thermal models predict that the lifetime of a hydrothermal system driven by a shallow-level intrusion will be ~0.1 m.y. or less. Because the Chuquicamata mine porphyries were emplaced at least 1 m.y. apart, each must have been accompanied by a temporally and, to some extent, spatially distinct magmatic-hydrothermal system. It is reasonable to conclude that the superposition of two or more such magmatic-hydrothermal systems generated the irregular alteration zoning and extraordinary size of the Chuquicamata deposit.

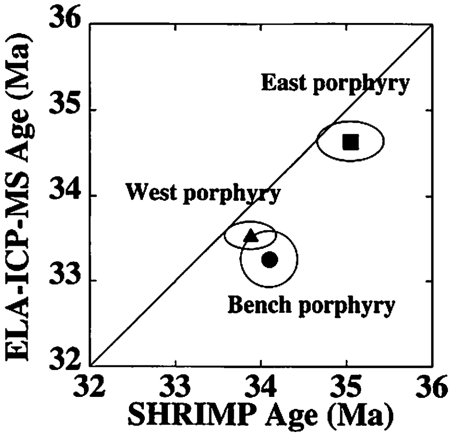

Figure 2. Sensitive high-resolution ion microprobe (SHRIMP) and Excimer laser-ablation–inductively coupled plasma–mass spectrometry (ELA-ICP-MS) U-Pb geochronology both resolve two igneous intrusive events. East porphyry intruded at ~34.6 Ma, and Bench and West porphyries intruded ca. 33.3 and 33.5 Ma, respectively.

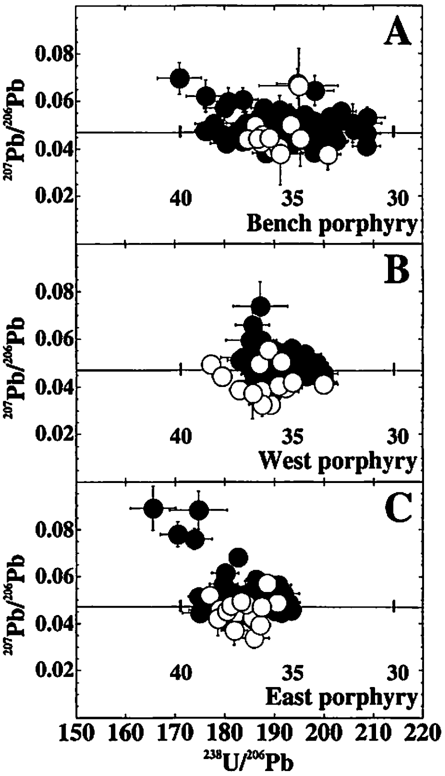

Figure 3. Concordia plots of Bench, West, and East porphyries by sensitive high-resolution ion microprobe (SHRIMP) (white) and Excimer laser-ablation–inductively coupled plasma–mass spectrometry (ELA-ICP-MS) (black), see text. ELA-ICP-MS data are uncorrected for common Pb, whereas SHRIMP data have been ^{204}Pb corrected. Only data included in age calculation are shown for the ELA-ICP-MS.

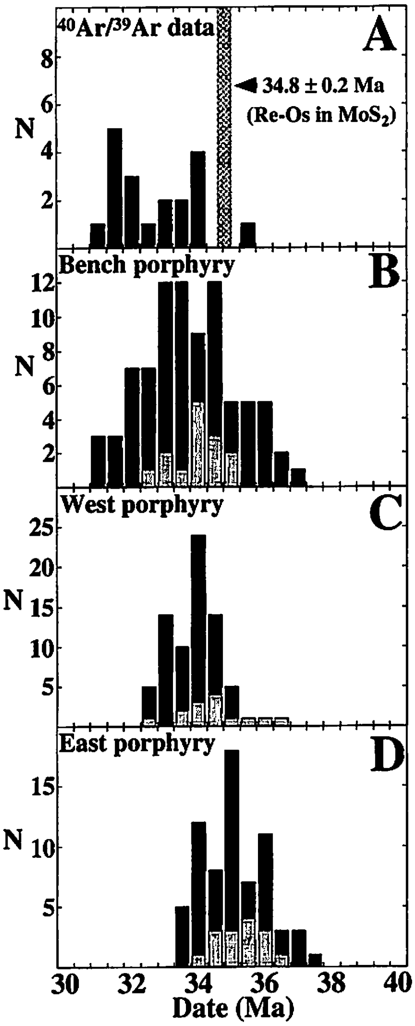

Figure 4. A: ^{40}Ar-^{39}Ar age data (modified from Reynolds et al., 1998) and Re-Os Mo-mineralization age data (J. Ruiz, 2000, personal commun.; Mathur et al., 2000) for Chuquicamata mine porphyries. B–D: Bench, West, and East porphyry age data by sensitive high-resolution ion microprobe (SHRIMP) (gray) and by Excimer laser-ablation–inductively coupled plasma–mass spectrometry (ELA-ICP-MS) (black), see text. SHRIMP analyses have been superimposed on ELA-ICP-MS data for ease of comparison. Only data included in age calculation are shown for the ELA-ICP-MS.

APPENDIX

Laser ablation was conducted by a pulsed ArF LambdaPhysik LPX 120I UV Excimer laser operated at a constant voltage, between 21

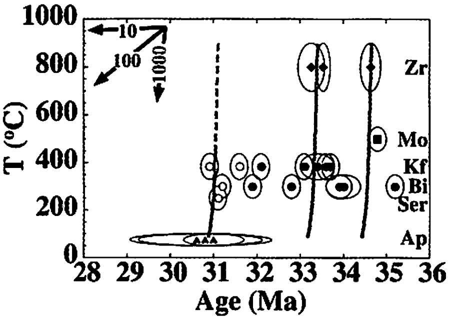

Figure 5. Thermal (*T*, temperature) history of Chuquicamata Intrusive Complex based on new zircon U-Pb ages and previous data as discussed in the text (Zr—zircon, Mo—molybdenite, Kf—potassium feldspar, Bi—biotite, ser—sericite, Ap—apatite). Age error ellipses are 2 σ, except for apatite ages that equal 1 σ. Near vertical lines represent inferred cooling histories of ca. 34.6 Ma, ca. 33.4 Ma, and ca. 31 Ma intrusive events. Arrows shown in top left indicate cooling rates in °C/m.y.

and 23 kV, at 5 Hz. The spot diameter was 29 μm. The ablated material was carried by He-Ar gas from a custom-designed sample cell and flow homogenizer to a Fissions VG PlasmaQuad II+ ICP-MS. Raw count rates for ^{10}B, ^{29}Si, ^{96}Zr, ^{206}Pb, ^{207}Pb, ^{208}Pb, ^{232}Th, and ^{238}U were collected in time-resolved mode. Because of a relatively high ^{204}Hg blank, ^{204}Pb was not measured. The integration time for the 3 Pb isotopes was 102.4 ms, whereas for the other isotopes it was 20.48 ms sampling at 1 point per peak. Data were acquired for 20 s with the laser off and 40 s with the laser on, giving ~120 mass scans for a penetration depth of ~20 μm.

Corrections were made for mass bias drift, isotopic fractionation, and common Pb. After triggering, it took three to four mass scans for the counts to reach a steady signal, so these initial data were excluded. Depth-dependent interelement fractionation of Pb, Th, and U, documented by previous workers (e.g., Hirata and Nesbitt, 1995; Horn et al., 2000), were corrected by reference to standard zircon SL13. Measured $^{207}Pb/^{206}Pb$, $^{206}Pb/^{238}U$, and $^{208}Pb/^{232}Th$ ratios in SL13 were averaged over the course of the analytical session and used to calculate correction factors. These correction factors were then applied to each sample to correct simultaneously for instrumental mass bias and depth-dependent elemental and isotopic fractionation. Common Pb was subtracted after drift, mass bias, and depth-dependent fractionation corrections. The common Pb corrections initially were based on the difference between the measured and expected $^{208}Pb/^{206}Pb$, given the measured $^{208}Pb/^{232}Th$, according to methods of Compston et al. (1984). This showed most analyses to be concordant within analytical uncertainty, so the $^{206}Pb/^{238}U$ ratios were then calculated by correcting for common Pb using $^{207}Pb/^{206}Pb$ and assuming concordance. Data that were more than 5% discordant were not used in age determinations. The remaining data were examined using cumulative probability plots. Old outliers were interpreted to be inherited grains and young outliers were attributed to lead loss.

The mean square of weighted deviates (MSWD) for all the ELA-ICP-MS samples, and the East porphyry by SHRIMP, are above the limit for a single population (Table 1). The MSWD for the SHRIMP data is reduced if the sample is interpreted to contain zircons with two ages, one at 34.4 ± 0.2 Ma and the other at 35.5 ± 0.2 Ma. However this interpretation is not unique and it is inconsistent with the unimodal distribution of the ELA-ICP-MS data. The high MSWD of the ELA-ICP-MS data is due in part to unquantified errors associated with standardization and, in part, to pooling data from three analytical sessions with variable drift. The MSWD values for individual days are appreciably less and range between 1.46 and 4.07.

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