

1 **The magma and metal source of giant porphyry-type**  
2 **ore deposits, based on lead isotope microanalysis of**  
3 **individual fluid inclusions**

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18 **Abstract**

19 The global supply of Mo and much of Cu and Au comes from porphyry-type ore deposits  
20 associated with hydrous magmas of broadly calc-alkaline composition, thought to be  
21 generated by contemporaneous subduction zone processes. Molybdenum is generally  
22 considered to be derived from the continental crust while Cu and Au are sourced in the  
23 mantle wedge above subducting slabs. Here we show that neither contemporaneous  
24 subduction nor derivation of Mo from crustal sources is required to explain the genesis of  
25 porphyry-Cu-Mo-Au deposits on Proterozoic lithosphere in the eastern Rocky Mountains.

26 Uniform Pb isotope ratios measured by LA-MC-ICP-MS in individual fluid inclusions from  
27 distinct Cu-Au and later Mo ore-forming stages at Bingham Canyon, USA, demonstrate a  
28 common metal source. Uranogenic Pb isotope ratios are particularly non-radiogenic ( $17.494 <$   
29  $^{206}\text{Pb}/^{204}\text{Pb} < 17.534$ ;  $15.553 < ^{207}\text{Pb}/^{204}\text{Pb} < 15.588$ ) and plot to the left of the geochron and  
30 above the mantle Pb evolution line. In  $^{207}\text{Pb}/^{206}\text{Pb}$  vs.  $^{208}\text{Pb}/^{206}\text{Pb}$  space, the fluid Pb isotope  
31 data cluster at the non-radiogenic end of a mixing line described by >80 feldspar data from  
32 igneous rocks intimately associated with magmatic-hydrothermal ore formation, which  
33 extends to modern depleted mantle or upper crust. Forward Monte Carlo simulations require  
34 three events for the U-Th-Pb isotope evolution of the fluid: (1) Late Archean formation of  
35 enriched crust is followed by (2) preferential extraction of Pb from this aged crust into a  
36 subduction fluid characterized by drastically reduced U/Pb that metasomatized lithospheric  
37 mantle at ~ 1.8 Ga. This mantle reservoir then evolved to produce the retarded uranogenic Pb  
38 isotope signatures of the Bingham Canyon Cu-Mo-Au deposit in the Cenozoic (3).

39 Similarly retarded uranogenic Pb isotope data characterize the giant porphyry-Mo and  
40 Climax-type Mo deposits of Henderson, Questa, Butte, and SE Arizona that occur in  
41 Proterozoic sutures of the central and eastern Rocky Mountains. We propose that Cenozoic  
42 melting of subcontinental lithospheric mantle metasomatized by subduction fluids during early  
43 Proterozoic amalgamation of terranes to the Wyoming Craton provides the metal endowment  
44 and subduction flavour to the giant magmatic-hydrothermal Cu-Mo-Au ore deposits in western  
45 North America, which together constitute the world's major molybdenum ore province.

46 **Keywords:**

47 Proterozoic subduction metasomatism,  
48 subcontinental lithospheric mantle,  
49 Post-collisional ore deposit,  
50 lead isotopes,  
51 laser ablation ICPMS

## 52 **1. Introduction**

53 Porphyry-type ore deposits form from hydrothermal fluid circulation in and around small  
54 intrusions atop large, intermediate to felsic, subvolcanic magma chambers that source the  
55 ore-forming fluid and most of the metals (e.g., Burnham, 1979; Dilles, 1987; Halter et al.,  
56 2002, 2005; Williams-Jones and Heinrich, 2005). Magma mixing, fractional crystallization and  
57 assimilation characterize these magmatic systems, sustained through repeated injection of  
58 basic mantle-derived magma providing heat and chemical components (Hildreth, 1981;  
59 Hattori and Keith, 2001; Maughan et al., 2002; Halter et al., 2005). Upper-crustal processes of  
60 ore formation are fairly well understood, but far less is known about the deep source  
61 characteristics of ore-forming magmas and their metals. This is partly due to hydrothermal  
62 alteration in subvolcanic intrusions hosting the ore deposits, which erases much of the earlier  
63 bulk geochemical and isotopic record. The debate is therefore alive, whether specific magma  
64 sources are required to form economically significant deposits of Cu, Mo and Au, but also of  
65 W, Sn, and platinum-group elements including Re. Empirical evidence for lithosphere-scale  
66 source control includes the observation that some regions of the Earth seem particularly well  
67 endowed with certain types of ore deposits. Four of the world's six largest molybdenum  
68 deposits (Climax, Henderson, Butte, and Bingham Canyon) occur in the western USA within  
69 less than 1% of the Earth's land surface (Singer et al., 2005). If lithosphere-scale source  
70 characteristics are essential for such extraordinary metal accumulation, the potential of finding  
71 new ore resources of the same type would be largely limited to such provinces. Alternatively,  
72 if fluid processes in magmatic-hydrothermal systems in the shallow crust alone would account  
73 for the formation of these economic geochemical anomalies, any area with a suitable  
74 magmatic-hydrothermal architecture could become a target for exploration.

75 Fluid evolution and ore mineral precipitation in hydrothermal systems are recorded by  
76 multiple generations of fluid inclusion assemblages and mineral inclusions, and their trapping  
77 sequence can be established through careful thin-section petrography. At concentrations of  
78 several thousand  $\mu\text{g Pb per g}$  of fluid (e.g., Landtwing et al., 2005; Seo et al., 2009; Klemm et  
79 al., 2008), the hydrothermal fluid is by far the most Pb enriched phase conserved in the high-  
80 temperature magmatic-hydrothermal ores; co-existing minerals contain traces of Pb only.  
81 Compositional modification of fluid inclusions after their entrapment in non-reactive quartz is  
82 unlikely or can be recognized by petrography or microthermometry, whereas trace amounts of  
83 Pb in porphyry-stage minerals (feldspars, Cu-Fe-sulphides) are easily modified by later,  
84 cooler fluids that commonly deposit galena (PbS) from a potentially different lead source.  
85 Direct analysis of Pb-rich high-temperature fluids therefore provides hitherto unavailable,  
86 controlled access to the most pristine Pb isotopic signatures of magmatic-hydrothermal ore-  
87 forming systems.

88 As a new and direct tracer of magmatic fluid source, we report the first in-situ measured  
89 Pb isotopic data of individual fluid inclusions from Mo and Cu-Au mineralized veins and  
90 magmatic feldspar phenocrysts for the porphyry-Cu-Au-Mo deposit at Bingham Canyon, Utah  
91 (Fig. 1), one of the world's largest and best-studied porphyry-type volcano-plutonic systems  
92 (e.g., Stacey et al., 1968; Waite et al., 1997; Maughan et al., 2002; Redmond et al., 2004;  
93 Redmond and Inaudi, 2010; Landtwing et al., 2005, 2010). We use radiogenic isotope  
94 systematics, because these retain a memory of processes which predate the formation of the  
95 fluid-generating magmas. Lead isotope systematics are particularly powerful because three  
96 radioactive decay chains produce distinct Pb isotopes ( $^{238}\text{U} \rightarrow ^{206}\text{Pb}$ ,  $^{235}\text{U} \rightarrow ^{207}\text{Pb}$  and  $^{232}\text{Th}$   
97  $\rightarrow ^{208}\text{Pb}$ ). Besides source fingerprinting, Pb isotopes also provide time-integrated information  
98 on Pb/U/Th ratios in the melt source, elements that are particularly sensitive to fractionation  
99 during fluid-assisted processes in the mantle and crust. Monte Carlo simulations are used to  
100 evaluate radiogenic Pb growth scenarios able to reproduce our measured Pb isotope  
101 signatures of the ore-forming fluid. Consistent with independent geological and geophysical  
102 constraints, we propose that ore-forming magmas and their metal endowment, including  
103 notably those sourcing porphyry-type Cu-Mo, Cu-Mo-Au and even large Climax-type Mo  
104 deposits in the eastern Rocky Mountains of the USA are derived from anciently

105 metasomatized subcontinental lithospheric mantle. Contrary to current views, significant input  
106 from evolved lower crustal sources, especially for Mo, appears unlikely.

## 107 **2. Bingham geology and sample characterization**

108 The giant Cu-Au-Mo deposit at Bingham Canyon formed in the late Eocene as part of a  
109 now partly eroded volcano-plutonic complex composed of calc-alkaline to shoshonitic and  
110 subordinate alkaline magmatic rocks (e.g., Maughan et al., 2002; Waite et al., 1997;  
111 Redmond and Einaudi, 2010). Hosted by a sequence of shallow monzonitic intrusions, it is  
112 situated in the Oquirrh Mountains in the easternmost part of the Basin and Range province.  
113 The Oquirrh mountains sit on the western projection of the Uinta axis, a Late Proterozoic  
114 basin extending along an even older continental suture (Presnell, 1997). To the east follows  
115 the Cheyenne Belt, which delineates the suture between Archean crust to the north (the  
116 Wyoming Province) and juvenile, accretionary, arc-like, paleo-Proterozoic terranes in the  
117 south (Dueker et al., 2001; Karlstrom et al., 2004; Mueller and Frost, 2006; Fig. 1).

118 In the Bingham Canyon deposit, alternating dyke intrusion and veining phases document  
119 an intimate relationship between magmatic and hydrothermal activity (Fig. 2). Cross-cutting  
120 relationships demonstrate that all Cu ± Au veins are older than a distinct last generation of  
121 veins containing most of the molybdenite (MoS<sub>2</sub>), which formed after the last stage of dyke  
122 intrusion (Redmond et al., 2004; Redmond and Inaudi, 2010).

123 Samples of quartz - chalcopyrite (Cu-Au stage) and quartz - molybdenite (Mo stage) veins  
124 contain numerous pseudosecondary fluid inclusion assemblages, which can be  
125 petrographically related to distinct phases of Cu-sulphide and molybdenite mineralization (Fig.  
126 2). The trapping history of the fluid inclusions plus extensive microthermometric and  
127 compositional data are reported in Landtwing et al. (2005, 2010). Both pulses of ore formation  
128 occurred between 425 - 350 °C from two-phase aqueous fluids (vapour and brine), which  
129 formed by phase separation from a single-phase magmatic-hydrothermal fluid of ~7 wt-%  
130 NaCl<sub>equivalent</sub> bulk salinity.

131 Late Eocene shallow intrusive rocks hosting the Bingham porphyry-type Cu-Au±Mo deposit  
132 are almost pervasively altered, with the exception of relicts of large magmatic feldspar pheno-  
133 or xenocrysts. In-situ Pb isotope analyses were thus carried out on K-feldspar and plagioclase

134 from Quartz-Monzonite Porphyry (QMP), Biotite Porphyry (BP) and Quartz Latite Porphyry  
135 (QLP), three intrusions that, from old to young, bracket the hydrothermal Cu±Au input into the  
136 Bingham Canyon deposit (Waite et al., 1997). These late Eocene - early Oligocene magmas  
137 intruded Paleozoic quartzite and limestone. Co-existing plagioclase and K-feldspar were also  
138 analyzed in order to test for Pb isotope disequilibrium between different minerals in the same  
139 rock. One rock sample (D404-1676) was also tested for the effects of potassic alteration of K-  
140 feldspar on Pb isotope systematics.

141 Melanephelinite and shoshonite lavas of the less-altered volcanic section of mafic and  
142 variably alkaline magmas in Rose Canyon, some 9 km to the SSE of the Bingham Canyon  
143 mine, have been interpreted to be directly related to the ore-forming magmas (Maughan et al.,  
144 2002). Four bulk rock samples were thus also analyzed for Pb isotope ratios.

### 145 **3. Analytical techniques**

146 For in-situ Pb isotope analysis we use a new method of laser-ablation multiple-collector  
147 inductively-coupled-plasma mass-spectrometry (LA-MC-ICP-MS) developed at ETH Zurich  
148 (Pettke et al., 2008; electronic appendix ES 1). A Geolas ArF excimer laser system (Lambda  
149 Physik, Germany; Günther et al., 1998) is coupled to two different MC-ICP-MS instruments  
150 (Nu Instruments, Wrexham, UK). The pulsed laser beam opens and liberates the entire  
151 contents of individual inclusions in a controlled manner, and the fast transient Pb isotopic  
152 signals are simultaneously recorded on Faraday detectors in static mode. Mass bias  
153 correlation parameters between measured  $^{205}\text{Tl}/^{203}\text{Tl}$  and Pb isotope ratios are established by  
154 measurements on NIST SRM 610 glass and then applied for within-run correction of the fluid  
155 inclusion analyses employing aspirated, desolvated Tl (Baxter et al., 2006). Isobaric  
156 interference by Hg on mass 204 is subtracted based on measured  $^{202}\text{Hg}$  intensities; other  
157 interferences were insignificant at the analytical precision obtained, even for SRM 610 glass,  
158 a trace element doped (nominally 500 µg/g) standard reference material. The analytical  
159 procedure has been successfully tested on synthetic Na-K-Pb-Cl fluid inclusions of known Pb  
160 isotopic composition (SRM 981), yielding accurate ratios for individual inclusions containing  
161 as little as 0.01 ng total Pb. The external precision achieved on our synthetic fluid inclusion  
162 analyses converges to ± 0.08% (2 s.d.) for  $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{207}\text{Pb}/^{206}\text{Pb}$  and to ± 0.4 % for Pb

163 isotope ratios containing mass 204. Magmatic feldspar phenocrysts and xenocrysts were  
164 analyzed with the same routine.

165 The same feldspar crystals were analyzed for their major and trace element compositions,  
166 using a GeolasPro laser system combined with a Perkin Elmer Elan DRC-e ICP-MS  
167 instrument at the University of Bern (see Pettke, 2008, for instrumental details and  
168 optimization strategies). Data quantification was based on SRM 612 for external calibration,  
169 and internal standardization was done by summation of all major element oxides to 100 wt-%.  
170 The accuracy of the data is routinely better than  $\pm 3\%$  (2 s.d.).

171 Five bulk rock Pb isotopic analyses were also performed. Rocks were crushed and then  
172 powdered in agate mills. 100 mg aliquots of rock powder were digested in a mixture of 9:1  
173 HF:HNO<sub>3</sub> at 180 °C in Savillex screwtop beakers. Lead was extracted from the samples  
174 employing mixed HNO<sub>3</sub>-HBr ion exchange procedures (Galer, 1986). Measurements were  
175 done on a Nu Plasma MC-ICP-MS at the University of Bern in static mode, using Faraday  
176 cups and employing TI addition for within-run correction of mass bias. Measurements on SRM  
177 981 performed during the same session were identical to the reference values.

#### 178 **4. Results**

179 24 individual fluid inclusions from 8 assemblages in 5 samples were investigated in two  
180 analytical sessions (Table 1). The precise results (Fig. 3) document identical Pb isotopic  
181 signatures for the successive (Fig. 2) Cu-Au *and* Mo ore stages at Bingham Canyon. The Pb  
182 isotopic signatures are conspicuously non-radiogenic, with averages for individual fluid  
183 inclusion assemblages restricted to  $17.494 < {}^{206}\text{Pb}/{}^{204}\text{Pb} < 17.534$ ,  $15.553 < {}^{207}\text{Pb}/{}^{204}\text{Pb} <$   
184  $15.588$ ,  $38.22 < {}^{208}\text{Pb}/{}^{204}\text{Pb} < 38.31$ ,  $0.88805 < {}^{207}\text{Pb}/{}^{206}\text{Pb} < 0.88955$ ,  $2.1803 < {}^{208}\text{Pb}/{}^{206}\text{Pb} <$   
185  $2.1853$ . Such a non-radiogenic signature is rare for Phanerozoic rocks and has never been  
186 reported for classical subduction-related calcalkaline magmas, with or without associated  
187 porphyry-type ore deposits. Data on element concentrations of Mo, Cu, Au, Pb, U and Th are  
188 also reported in Table 1. For Bingham ore fluids, measured U/Pb ratios are  $\sim 0.001$  and  
189 Th/Pb ratios are  $\ll 0.001$ ; hence, correction for in-situ grown radiogenic Pb since fluid  
190 inclusion formation at ca. 38 Ma (Parry et al., 2001) is insignificant. We thus report measured  
191 Pb isotope ratios.

192 The fluid inclusion results lie at the non-radiogenic end of a linear trend in  $^{207}\text{Pb}/^{206}\text{Pb}$  vs.  
193  $^{208}\text{Pb}/^{206}\text{Pb}$  space defined by ~80 in-situ analyses on feldspar phenocrysts and xenocrysts  
194 from the associated intrusions (Fig. 4; Electronic Supplementary Table ES2). This linear trend  
195 extends towards more radiogenic values of present-day depleted mantle. Some K-feldspar  
196 phenocrysts host melt and fluid inclusions. Inclusion-poor and inclusion-rich analyses  
197 randomly align along the same Pb isotopic trend; the one analysis on a sample devoid of  
198 inclusions lies in the central part of the trend (Fig. 5A). Lead isotope systematics in co-existing  
199 K-feldspar and plagioclase often show Pb isotope disequilibrium within the same rock (Fig.  
200 5B), their data conforming to the same trend, however.

201 Compositionally, the feldspars are strikingly uniform (Table ES2). Most importantly, all  
202 feldspar analyses have U/Pb and Th/Pb abundance ratios smaller than 0.0003 and 0.0002,  
203 respectively, thus requiring no correction for in-situ grown Pb since 38 Ma. Anorthite contents  
204 in plagioclase vary little across the successive intrusions, from An22 to An25. K-feldspar  
205  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios are variable. Inclusion-free K-feldspar (e.g., sample 4890-2080) has  
206  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  up to 0.28, while inclusion-rich feldspar domains from hydrothermally overprinted  
207 sample D404-1676 are characterized by  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ~0.14, consistent with ore-related potassic  
208 alteration. Contents of Ba are 1 wt-% on average, in line with the conspicuously high Ba  
209 contents of the bulk rocks (Maughan et al., 2002).

210 Bulk rock Pb isotope data for the melanephelinite and shoshonite rock suites, which occur  
211 peripherally to the Bingham Canyon mine and were considered to be co-sanguineous with the  
212 ore-forming magma (Waite et al., 1997, Maughan et al., 2002), do not plot on the Bingham Pb  
213 isotope trend (Fig. 4) but are enriched in  $^{208}\text{Pb}$  relative to  $^{207}\text{Pb}$ . The melanephelinite samples  
214 show the most radiogenic Pb isotope compositions measured on Bingham magmas,  
215 interpreted to reflect the most significant addition of present-day common mantle Pb, this  
216 being in tune with their largest content of compatible elements. However, the Pb isotopic  
217 composition of the propylitically altered pre-mineralization Equigranular Monzonite sample  
218 from the Bingham Canyon pit plots on the mixing trend defined by the in-situ feldspar and fluid  
219 inclusion analyses.

## 220 **5. Discussion: Source of Bingham ore fluids**



221 The fluid Pb isotope signatures for the chemically distinct and temporally successive Cu-  
222 Au and Mo mineralization stages of the Bingham Canyon deposit are identical, implying a  
223 common metal source. The ore fluids occupy the non-radiogenic end of a dominantly binary  
224 mixing array populated by feldspar phenocryst data from the ore-related porphyry intrusions  
225 (Figs. 4, 5). These subvolcanic intrusions are too small to be the source of the ore fluids,  
226 indicating that the ore fluids and the subvolcanic intrusions originate from a common, much  
227 larger but only partially homogenized magma reservoir (cf. Halter et al., 2005). The more  
228 radiogenic lead isotope component of the feldspar phenocryst mixing array may be of mid- to  
229 upper crustal derivation (cf. Stacey et al., 1968) or may represent addition of a common  
230 mantle component. The non-radiogenic mixing component dominating in the ore fluids thus  
231 represents a deep endmember component and probably relates to the magma source that  
232 also contributed the ore metals. The origin of this deep and retarded uraniumogenic lead  
233 component plotting to the left of the geochron (Fig. 6A) is the focus of the following  
234 discussion.

### 235 **5.1 A lower crustal magma and metal source?**

236 The classic view on Cretaceous to Tertiary evolved magmatism in Precambrian crustal  
237 domains of the Rocky Mountains, including the Bingham – Uinta district, interprets the  
238 dominant parental melt to be derived from evolved lower continental crust assembled during  
239 the Proterozoic (e.g., Stacey et al., 1968; Zartman, 1974; Farmer and De Paolo, 1983, 1984).  
240 Arguments for this lower-crustal melt source are based on combined radiogenic isotope  
241 systematics (variably negative present-day  $\epsilon_{Nd}$ , moderately radiogenic Sr, and non-radiogenic  
242 Pb isotope ratios) and the often peraluminous character of the most evolved magmas.

243 Figure 6 compares xenolith Pb isotope data for lower crust and lithospheric mantle below  
244 western North America with the Bingham data. Lower crustal xenolith data scatter strongly  
245 and plot above the MORB-source mantle evolution line, for the Archean Wyoming Craton at  
246 variably low  $^{206}Pb/^{204}Pb$ , and for Proterozoic crust at variably higher  $^{206}Pb/^{204}Pb$ . Xenoliths of  
247 metasomatized subcontinental lithospheric mantle from the Wyoming Craton and the adjacent  
248 Great Falls Tectonic Zone tend to have  $^{206}Pb/^{204}Pb$  ratios intermediate to these groups and  
249 cluster around the Bingham data, with slightly elevated  $^{207}Pb/^{204}Pb$  and less radiogenic

250  $^{206}\text{Pb}/^{204}\text{Pb}$  compared to Proterozoic crustal xenoliths. Galena and feldspar data from the  
251 Bingham - Oquirrh region plot at high  $^{207}\text{Pb}/^{204}\text{Pb}$  for a given  $^{206}\text{Pb}/^{204}\text{Pb}$ , identified by Stacey  
252 et al. (1968) as mixing between subordinate upper crustal radiogenic Pb and a more remote  
253 Pb source assumed to be ancient lower crust. While the scatter in the feldspar  $^{206}\text{Pb}/^{204}\text{Pb}$   
254 data might be due to upper-crustal contamination or represent subordinate contribution of  
255 modern mantle Pb (as indicated by the extension of the feldspar data towards Aleutian arc  
256 lava signatures), the source of the endmember component dominating the fluid data remains  
257 ambiguous. Lead, Nd and Sr isotope systematics alone do not allow a clear distinction  
258 between a lower crustal and a modified lithospheric mantle source of the parental magmas.  
259 As an alternative to lower-crustal melt generation, other workers have favoured a  
260 metasomatized lithospheric mantle source for mafic magma components with comparable  
261 radiogenic isotope systematics, occurring in Proterozoic crustal domains characterized by  
262 lithospheric thinning (e.g., Kempton et al., 1991; Harry and Leeman, 1995; Hawkesworth et  
263 al., 1995; Johnson et al., 1990; Johnson, 1991; Hart et al., 1997).

## 264 **5.2 An ancient lithospheric mantle magma and metal source for Bingham Canyon**

265 In the following section, we show that the ore-fluid endpoint can be explained by a  
266 quantitative U-Th-Pb evolution model, in which the parental magma is derived by melting a  
267 subcontinental lithospheric mantle that had been metasomatized by subduction fluids about  
268 1.8 billion years ago. The model reproduces the present-day Pb isotope ratios measured in  
269 the ore fluids, while satisfying independent geochemical and geodynamic boundary  
270 conditions. Two long-lasting evolution stages bounded by three events are required by the  
271 uraniumogenic Pb isotope signatures to have affected the source region of the Bingham  
272 magmas, as indicated schematically in Figure 7 and tested with Monte Carlo simulations  
273 (Table 2). Thorogenic Pb systematics contribute complementary information only, but are  
274 consistent with the model. One-stage evolution models cannot satisfy uraniumogenic Pb isotope  
275 systematics and are therefore rejected. Variations of  $\pm 0.2$  Ga in the timing of events 1 and 2  
276 are easily tolerated in our preferred model, variations that exceed those permitted by the  
277 known geological evolution of the region. Figure 8 depicts our preferred Pb evolution model,  
278 which leads to the conspicuously non-radiogenic Pb isotopic composition of the ore-forming  
279 fluid that lies to the left of the geochron and above the MORB source mantle evolution line

280 (Kramers and Tolstikhin, 1997), in  $^{207}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  representation,  
281 together with the three major events marked by solid stars.

282 **Event 1:** Late Archean juvenile crust formation is widespread in western North America  
283 (Carlson and Irving, 1994; Chamberlain et al., 2003; Souders and Frost, 2006; Frost et al.,  
284 2006a). This was the first essential event, leading to elevated  $\mu$  values ( $^{238}\text{U}/^{204}\text{Pb}$ ) because  
285 of the higher incompatibility of U relative to Pb during shallow partial mantle melting. Melt  
286 extraction from a MORB-source mantle (Kramers and Tolstikhin, 1997) during the late  
287 Archean (Table 2, simulation 1) would necessitate very high  $\mu_1$  values ( $>14.0$ ) for newly  
288 formed crust (stage 1), too high to be compatible with significant fractions of mantle melting.  
289 U-Pb fractionation factors between such a MORB source mantle and its melt products as  
290 required by the model would become lower and thus more plausible for significantly older  
291 separation ages (e.g., 3.2 Ga; simulation 2), but there is no independent indication for  
292 significant mid-Archean crust formation in the southern Wyoming Province (e.g., Chamberlain  
293 et al., 2003; Frost et al., 2006a). Consequently, melt extraction from an already evolved Pb  
294 isotope reservoir is indicated, i.e., a high- $\mu$  late Archean mantle, modified perhaps by  
295 recycling of older crustal materials. Our simulation 5 (Table 2) returns a plausible  $\mu$  value of  
296 8.5 for this mantle reservoir, consistent with the enriched mantle reservoir proposed for the  
297 late Archean Wyoming Province ( $8.5 > \mu > 8.65$ ; Frost et al., 2006b; Mueller and Frost, 2006).  
298 The late Archean crust produced by Event 1 subsequently evolved in isolation for about 1 Ga  
299 (stage 1 from 2.8 - 1.8 Ga; Fig. 8), resulting in time-integrated Pb isotope signatures that plot  
300 well above the MORB-source mantle of Kramers and Tolstikhin (1997) or a primitive mantle  
301 evolution line in  $^{207}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  space.

302 **Event 2:** Subduction and accretion of arc terranes to the Wyoming Craton along its southern,  
303 western and north-western margins assembled Laurentia around 1.8 Ga (e.g., Chamberlain,  
304 1998; Mueller et al., 2002; Karlstrom et al., 2004, Foster et al., 2006; Mueller and Frost,  
305 2006). The Cheyenne Belt (Fig. 1) marks the suture between Archean crust to the North and  
306 juvenile 1.8-1.7 Ga Proterozoic island arc crust. A north-dipping, high velocity mantle anomaly  
307 beneath the southern rim of the Wyoming Craton is still visible today, most likely representing  
308 an ancient slab fragment subducted at  $\sim 1.8$  Ga (Dueker et al., 2001; Karlstrom et al., 2004).  
309 We propose that this Event 2 metasomatized a depleted mantle wedge above the northward

310 subducted Cheyenne slab, by fluids that devolatilized from late Archean crustal material  
311 produced in Event 1 (Fig. 7). Slab fluids are characterized by enrichment in light rare earth  
312 elements (LREE) and large ion lithophile elements (LILE), most importantly K, Pb, Rb, Ba and  
313 Sr (e.g., Kessel et al., 2005). The resulting lowered Sm/Nd and moderately elevated Rb/Sr  
314 ratios are consistent with the present-day negative  $\epsilon_{Nd}$  and moderately radiogenic Sr isotopic  
315 ratios measured in Bingham magmas (e.g., Maughan et al., 2002, Waite et al., 1997). Most  
316 importantly, this subduction metasomatism led to drastically reduced  $\mu$  values in the mantle  
317 beneath Bingham (simulations 1 - 5 in Table 2), as the enrichment of Pb  $\gg$  U is diagnostic for  
318 an aqueous fluid component released during devolatilization of crust at cool, sub-solidus  
319 conditions and shallower than about 150 km (Kessel et al., 2005). Mass balance modelling  
320 demonstrates that addition of aqueous slab fluids to depleted mantle will dominate the  
321 incompatible element inventory of the metasomatized mantle product and thus its U-Th-Pb  
322 system. Consequently, we interpret the elevated  $^{207}\text{Pb}$  signature as compared to MORB-  
323 source mantle Pb isotopic composition as a signature of subducted late Archean crust,  
324 transferred to the overlying mantle wedge at 1.8 Ga. Phlogopite-bearing and partially veined  
325 (veining dated at  $\sim$ 1.8 Ga) dunite and harzburgite xenoliths from Eocene minette dikes  
326 intruded within and along the NW margin of the Wyoming craton in the Great Falls Tectonic  
327 Zone (Fig. 1; Carlson and Irving, 1994) may represent samples of such a metasomatized  
328 lithospheric mantle analogue. After terrane amalgamation in Event 2, the metasomatized  
329 material evolved in isolation as subcontinental lithospheric mantle for  $\sim$ 1.7 billion years (stage  
330 2; Fig. 8), until the onset of Cenozoic magmatic activity.

331 **Event 3:** Cenozoic magmatism at Bingham Canyon probably involved dominant melting of  
332 this ancient metasomatized mantle plus a smaller contribution from ordinary (depleted)  
333 mantle, to generate the binary lead isotope mixing trend recorded by the igneous feldspars  
334 and the ore fluids (Fig. 4). It has been tentatively argued that the calcalkaline to shoshonitic  
335 subduction flavour of late Eocene Bingham magmas may be related low-angle subduction of  
336 the Farallon Plate beneath western North America (Maughan et al., 2002; Waite et al., 1997).  
337 However, the lead isotopes show little, if any, contribution of Pb from modern oceanic  
338 lithosphere, and the subduction front was  $>$ 500 km to the west of Bingham, farther away than  
339 what can be reconciled with common subduction zone geometries and subsequent basin-

340 and-range extension. Moreover, preservation of a Proterozoic lithospheric keel >200 km thick  
341 beneath the eastern Rocky Mountains (Karlstrom et al., 2004) is inconsistent with a model of  
342 low-angle subduction of the Farallon plate reaching as far east as Bingham. We therefore  
343 propose that incipient extensional tectonics during late Eocene triggered partial melting of  
344 subcontinental lithospheric mantle domains below the Bingham area, and that the modern  
345 magmas and fluids acquired their subduction signature from an event that occurred some 1.7  
346 Ga earlier (Fig. 7). Metasomatized mantle rocks are expected to melt selectively at lower  
347 temperature than enclosing depleted peridotite (Leeman and Harry, 1993). High degrees of  
348 melting of metasomatized mantle rocks (e.g., phlogopite + orthopyroxene + clinopyroxene ±  
349 amphibole - veined harzburgite as sampled in xenoliths from the Great Falls Tectonic Zone;  
350 Carlson and Irving, 1994; Downes et al., 2004) can produce voluminous mafic magmas of  
351 mildly alkaline character (Pilet et al., 2008) rich in incompatible elements, consistent with Pb  
352 isotope data as well as the arc-like trace element signature and the conspicuous Ba  
353 enrichment that characterize Bingham magmas (Maughan et al., 2002, Waite et al., 1997). It  
354 is conceivable that substantial enrichments in highly refractory elements such as Ni, Cr and  
355 Mg (Maughan et al., 2002, Waite et al., 1997) and the subordinate radiogenic Pb component  
356 originated from interaction of the lithospheric melts with non-metasomatized portions of  
357 depleted subcontinental lithospheric mantle. Our fluid Pb isotope results clearly demonstrate,  
358 that modern mantle derived Pb is, at most, a minor component in these magmas, consistent  
359 with the Nd-Sr-Pb systematics of the previous studies.

## 360 **6. Ancient, metasomatized mantle: key to a global ore province**

361 Four of the six largest molybdenum deposits in the world (Henderson, Climax, Bingham  
362 Canyon, Butte) and several other world-class Mo ± Cu deposits are located in the western  
363 USA. We propose that the geodynamic and geochemical source model developed for  
364 Bingham Canyon also applies to these other giant Mo-rich ore deposits, and that such an  
365 extended lithospheric pre-history may be a defining hallmark and essential metallogenic  
366 component of the premier global molybdenum province.

367 All four giant deposits and several other Mo-rich ore districts in this province are located  
368 along Proterozoic lithospheric boundaries (Fig. 1). The ancient suture zones are delineated by

369 velocity contrasts in mantle tomography images resulting from compositional heterogeneities  
370 (Dueker et al., 2001; Karlstrom et al., 2004). Climax and Henderson lie in the Colorado  
371 Mineral Belt, interpreted to follow a NE striking Proterozoic arc-arc suture. The Mo-deposit at  
372 Questa, New Mexico, lies on the eastern flank of the Rio Grande Rift (Johnson et al., 1990),  
373 near the Jemez suture zone believed to represent the reactivated boundary between the 1.8-  
374 1.7 Ga Yavapai and Mazatzal Provinces. Near the SW end of this lineament, in the southern  
375 Eastern Transition Zone of the Basin and Range province (e.g., Kempton et al., 1991), Mo-  
376 rich Cu porphyries define the northern part of the SE Arizona district (SEAZ in Fig. 8). Finally,  
377 the giant Cu-Mo deposit at Butte, Montana (Doe et al., 1968), occurs at the southwest end of  
378 the Great Falls Tectonic Zone, a Proterozoic collisional suture between two Archean terranes  
379 (e.g., Mueller et al., 2002). These Mo-rich ore deposits occur in different crustal domains, but  
380 they all are located on or near Proterozoic crustal boundaries atop volatile-rich lithospheric  
381 mantle (e.g., Carlson and Irving, 1994; Downes et al., 2004; Karlstrom et al., 2004). Ore-  
382 forming Cenozoic magmatism occurred during early stages of tectonic perturbation (extension  
383 and/or asthenospheric upwelling) of these ancient lithospheric structures, with little or no  
384 temporal or spatial relationship to modern subduction.

385 All of these Mo-rich deposits and the associated magmas are characterized by retarded  
386 uraniumogenic Pb isotope signatures (Fig. 8). In any one magmatic-hydrothermal ore district, the  
387 ore minerals from the largest deposits and their most intimately associated igneous rocks  
388 show the most extreme uraniumogenic Pb isotope retardation compared to smaller deposits and  
389 less-directly ore-related magmas (Stacey et al., 1968; Doe et al., 1968; Bouse et al., 1999;  
390 Johnson et al., 1990; Stein and Hannah, 1985). Molybdenum endowment therefore seems to  
391 be correlated with the contribution of this retarded source component. The origin of this  
392 component has been suggested to be the lower crust, mainly because the molybdenum-only  
393 porphyries (Climax, Henderson, Questa) are all associated with highly silicic and partly  
394 peraluminous intrusions and variably non-radiogenic Nd and moderately radiogenic Sr isotope  
395 signatures (e.g., Farmer and DePaolo, 1984). However, several lines of evidence suggest  
396 that metasomatized subcontinental lithospheric mantle is at least a permissive alternative  
397 source, as previously suggested by Kempton et al. (1991), Johnson (1991) and Hawkesworth  
398 et al. (1995). All of the mineralized igneous complexes include at least minor mafic (~basaltic)

399 components (e.g., Johnson et al., 1990; Waite et al., 1997). Lower-crustal xenoliths in SE  
400 Arizona have distinctly more radiogenic  $^{206}\text{Pb}/^{204}\text{Pb}$  (Esperanca et al., 1988; Kempton et al.,  
401 1990) than the Mo-rich porphyries in this district (Bouse et al., 1999). Peraluminous granite  
402 magmas do not necessarily require sediment assimilation; they can also be generated by  
403 fractional crystallization of typical mantle-wedge melts of picobasaltic composition (Muntener  
404 and Ulmer, 2006). The volume of basaltic melt required to generate progressively Mo-  
405 enriched felsic magmas (e.g., Carten et al., 1993) is consistent with partial melting of  
406 metasomatized subcontinental lithospheric mantle during early stages of extension, as  
407 discussed by Harry and Leeman (1995) for the Proterozoic Rocky Mountains domain. The  
408 isotopic composition of sulphur in giant molybdenum deposits is also consistent with  
409 fractionation of the associated rhyolitic magmas from mantle melts (Henderson, Questa:  $+1 <$   
410  $\delta^{34}\text{S} < 5.3 \text{‰}$ ; Stein and Hannah, 1985).

411 We propose that magmas originating from a metasomatized subcontinental lithospheric  
412 mantle are the decisive ingredient for the formation of giant Mo-rich porphyry deposits and  
413 this unique molybdenum ore province. Parental melt compositions were probably similar to  
414 basalts from the Eastern Transition Zone, characterized by high LIL/HFS element ratios and  
415 retarded uraniumogenic Pb signatures of an age of ca. 1.8 Ga (Kempton et al., 1991). Mafic  
416 magmatism commonly occurred late in the evolution of mineralized complexes (e.g.,  
417 Maughan et al., 2002; Johnson et al., 1990), inconsistent with progressive melting of felsic  
418 crust. Sustained mantle magma input (e.g., Harry and Leeman, 1995; Hildreth and Moorbath,  
419 1988) allows for protracted fractional crystallization promoting enrichment of the incompatible  
420 ore metals in the residual melt. Assimilation of lower crust lithologies, possibly indicated by  
421 more radiogenic  $^{208}\text{Pb}/^{204}\text{Pb}$  and less retarded  $^{206}\text{Pb}/^{204}\text{Pb}$  (Fig. 6), can be an ancillary  
422 consequence of long-lived magmatic systems (e.g., Hildreth and Moorbath, 1988), rather than  
423 a prerequisite to province-scale molybdenum enrichment as commonly inferred (e.g., Farmer  
424 and DePaolo, 1984; Bouse et al., 1999).

425 Our genetic model for Mo-rich porphyry deposits may also apply to the Gangdese belt in  
426 the Tibetan orogen. Here, world-class porphyry Cu-Mo deposits formed from high-K  
427 calcalkaline magmatism some 50 million years after arc magmatism (Hou and Cook, 2009).  
428 These systems are characterized by mantle-like  $\delta^{34}\text{S}$  but less radiogenic Nd, slightly more

429 radiogenic Sr, and enriched  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios compared to present-day mantle (Qu et al.,  
430 2007), but lack the retarded uraniumogenic Pb isotope ratios due to the short time span between  
431 mantle metasomatism and melt extraction.

432 Lithospheric mantle domains enriched by ancient subduction metasomatism occur down to  
433 at least 200 km (e.g., Scambelluri et al., 2008) and may be a world-wide factor determining  
434 the distribution of major metal provinces and giant ore deposits. Based on global xenolith  
435 geochemistry and geophysical data, Griffin et al. (2009) concluded that the roots of most  
436 Proterozoic shields consist of refertilized Archean lithospheric mantle that has escaped  
437 reintegration into the convecting mantle, because strongly depleted but subsequently  
438 subduction-metasomatized mantle rocks are buoyant (cf. Carlson and Irving, 1994). The  
439 cause for selective mantle enrichment in Mo, but possibly also in S, Cu and Au is not clear. It  
440 could relate to the redox evolution of the latest Archean to early Proterozoic hydrosphere,  
441 which favoured the enrichment of chalcophile metals and gold in predominantly reduced  
442 deep-sea shales (e.g., Holland, 2005), prior to their subduction in a period of intensive  
443 continental (re)construction during the paleo-Proterozoic. Irrespective of the mechanism of  
444 this ancient mantle source enrichment, our results endorse modern approaches to global  
445 mineral exploration and resource assessment, which place increasing weight on mapping the  
446 chemical and thermal prehistory of large-scale lithospheric mantle domains, and on identifying  
447 their boundaries by geophysical methods and mantle xenolith data (Begg et al., 2009).

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655 **Figure Captions**

656 Fig. 1: Sketch of SW Laurentia (W USA) identifying major crustal segments (modified from  
657 Karlstrom et al., 2004, Foster et al., 2006). Archean crustal blocks (grey) are delimited  
658 by Proterozoic accretionary tectonic zones. Segments to the south of the Cheyenne  
659 Belt (CB) are terranes accreted during assembly of Laurentia, and sutures represent  
660 early Proterozoic subduction zones. The dash-dotted line separates terranes  
661 accreted to the Laurentian crustal block in the Phanerozoic. Filled black circles  
662 denote Bingham (Bh), situated on the Uinta Axis, Butte (Bt), Henderson (H) in the  
663 Colorado Mineral Belt, Questa (Q) on the eastern rim of the Cenozoic Rio Grande Rift  
664 (omitted for clarity), and the northern SE Arizona (SEAZ) ore district. F-L SZ refers to  
665 the Farewell Mt. - Lester Mt. Suture Zone.

666 Fig. 2: (A) Polished drill core cut showing the temporal relationships between successive  
667 intrusions and veining events at Bingham (Landtwing, 2004). The Quartz Monzonite  
668 Porphyry (QMP) is veined by stockwork quartz sulphide veins (Cu-Au vein) and then  
669 intruded by the Quartz Latite Porphyry (QLP). Hair quartz veinlets (not visible  
670 macroscopically) penetrated the rock before late quartz-molybdenite veining (Mo-  
671 vein) occurred. (B) Thick section photograph showing fluid inclusions and  
672 molybdenite crystals (black, labelled  $\text{MoS}_2$ ) in a quartz crystal from a quartz-  
673 molybdenite vein. The blow-up (C) identifies 3 brine inclusions analyzed for Pb  
674 isotope ratios that lie on the same plane as the black molybdenite crystal in the top-  
675 right corner. Labels correspond to analysis numbers in Table 1.

676 Fig. 3:  $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  results for individual fluid inclusions from Bingham. A  
677 uniform Pb isotopic composition characterizes the texturally distinct Cu-Au (full  
678 symbols) and Mo (empty symbols) ore stages. Dashed lines separate inclusions from  
679 different samples (labels shown at the top of the diagram), identical symbols identify  
680 inclusions from the same fluid inclusion assemblage. Error bars are two standard  
681 error measurement uncertainties (2 s.e.m.), some of them smaller than symbol size.

682 Fig. 4: Graph of  $^{207}\text{Pb}/^{206}\text{Pb}$  vs.  $^{208}\text{Pb}/^{206}\text{Pb}$  for Bingham Cu-Mo-Au and other Mo-rich  
683 magmatic-hydrothermal ore deposits from the eastern Rocky Mountains. Bingham  
684 feldspar and fluid data define a linear trend (black solid line), which extrapolates to  
685 the more radiogenic values of the Butte porphyry-type Cu-Mo deposit and associated



686 magmatic rocks and to values measured for present-day depleted mantle (dotted  
687 area; Kramers and Tolstikhin, 1997), indicating a mixing relationship. Note that the Pb  
688 compositions of the fluid inclusions are uniform and cluster tightly at the non-  
689 radiogenic end (top right corner) of the Bingham array. Data for melanephelinite and  
690 shoshonite samples (Maughan et al., 2002) show more enriched  $^{208}\text{Pb}$ , interpreted to  
691 reflect source heterogeneities. The white star represents a propylitically altered pre-  
692 mineralization monzonite sample (Last Chance Stock) from Bingham. Data for lower-  
693 temperature galena of distal vein deposits at Bingham (Stacey et al., 1967), data  
694 fields for the least radiogenic Miocene hornblende andesite samples from the Latir  
695 Volcanic Field associated with Questa (Johnson et al., 1990) and for porphyry-Cu-Mo  
696 deposits from the northern SE Arizona district (SEAZ; Bouse et al., 1999) are shown  
697 for comparison. Analytical uncertainties (2 s.e.m.) are smaller than symbol size.

698 Fig. 5: In-situ Pb isotope data for feldspar phenocrysts and xenocrysts. Plot (A) shows K-  
699 feldspar data variably affected by the presence of polyphase inclusions in the ablated  
700 sample volume. The dashed arrows indicate the shift from inclusion-poor to inclusion-  
701 rich parts of the analytical signal of an individual spot ablation. Overall, there is no  
702 clear trend in Pb isotope data since the one analysis devoid of inclusions lies in the  
703 central part of the mixing trend. Plot (B) reveals distinct Pb isotopic difference  
704 between K-feldspar and plagioclase, indicating that magma mixing is at least partially  
705 responsible for the variable Pb signatures of the samples. Inter-sample and intra-  
706 sample Pb isotope disequilibrium characterizes the magmatic rocks at Bingham,  
707 reflecting primary variability (i.e., magma mixing) and, for K-feldspar, partial  
708 equilibration during magmatic-hydrothermal overprinting. Analytical uncertainties are  
709 2 s.e.m., often smaller than symbol size.

710 Fig. 6: Comparison of Pb isotope data for xenoliths from lower crust and lithospheric mantle  
711 of the US Archean to Proterozoic lithosphere with the Bingham fluid inclusion and  
712 feldspar results. Data for SCLM are metasomatized mantle xenoliths (Carlson and  
713 Irving, 1994; Carlson et al., 2004), data for lower crust are represented by granulitic  
714 xenoliths (Proterozoic: Esperanca et al., 1988; Kempton et al., 1990; Carlson et al.,  
715 2004; Wyoming Craton: Bolhar et al., 2007; Mirnejad and Bell, 2008), Bingham

716 galena data are from Stacey et al. (1967), and Aleutian arc lava data (Singer et al.,  
717 2007) represent modern subduction zone magmatism. MORB-source mantle is after  
718 Kramers and Tolstikhin (1997). (A) Uranogenic diagram, (b) thorogenic diagram. Note  
719 that the Bingham data cluster tightly near the metasomatized mantle xenoliths from  
720 the Great Falls Tectonic Zone (Carlson and Irving, 1994) while crustal xenoliths vary  
721 strongly and are generally less (Wyoming Craton) or much more (Proterozoic  
722 terrains) radiogenic.

723 Fig. 7: Sketch illustrating our favourite scenario with 3 events (marked by stars on horizontal  
724 time axis) and two stages (dashed arrows) required to explain the Pb isotopic  
725 signatures of Bingham ore fluids. Juvenile crust, formed in the late Archean (event 1),  
726 evolves in isolation (stage 1) until this crust is subducted northwards during assembly  
727 of Laurentia in the early Proterozoic. Subduction fluids are released and  
728 metasomatize the overlying depleted mantle (dotted region, event 2) that evolves in  
729 isolation (stage 2) until partial melting of this metasomatized subcontinental  
730 lithospheric mantle is triggered by mild extensional tectonics in the late Eocene (event  
731 3) to generate Bingham magmatism. Note that Cenozoic subduction is far to the west.  
732 WC refers to the Wyoming craton, CC to continental crust, and Prot. SCLM to  
733 Proterozoic subcontinental lithospheric mantle.

734 Fig. 8: (A) Forward modelling of the uranogenic (A) and thorogenic (B) two-stage Pb isotope  
735 evolution (stage 1, thick lines; stage 2, thin lines) that reproduces the measured Pb  
736 isotope signatures of Bingham ore fluids (grey star at 38 Ma).  $\mu$  values denote  
737  $^{238}\text{U}/^{204}\text{Pb}$ ,  $\kappa$  values  $^{232}\text{Th}/^{238}\text{U}$ . Ages are in million years (Ma) before present, the  
738 black stars marking the beginning of stages 1 and 2, respectively. The dashed MORB  
739 source mantle evolution line (Kramers and Tolstikhin, 1997) represents convecting,  
740 depleted mantle, the dotted line (A only) depicts primitive mantle. Data for other Mo-  
741 enriched porphyry deposits of the eastern Rocky Mountains, US (Area 1 of Zartman,  
742 1997), are shown for comparison (Butte: Doe et al., 1968; Henderson: Stein and  
743 Hannah, 1985; hornblende andesite associated with Questa: Johnson et al., 1990;  
744 SEAZ: northern SE Arizona district Cu-Mo centres; Bouse et al., 1999). Variability in  
745 thorogenic Pb at relatively uniform  $^{206}\text{Pb}/^{204}\text{Pb}$  is interpreted to reflect heterogeneities

746 in Proterozoic subduction metasomatism as subduction devolatilization may generate  
747 large variations in Th/U of product fluids (Kessel et al., 2005).

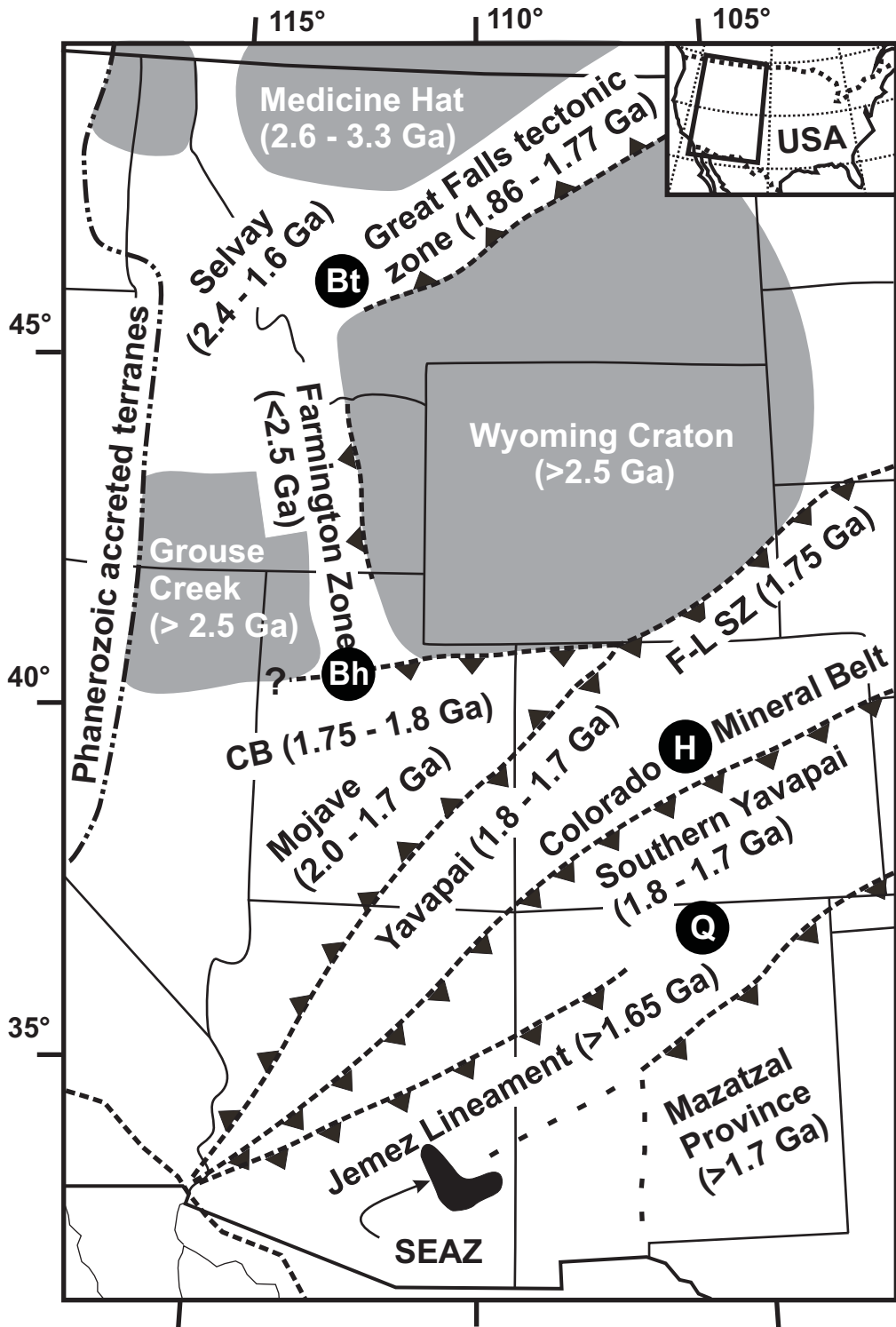


Figure 1

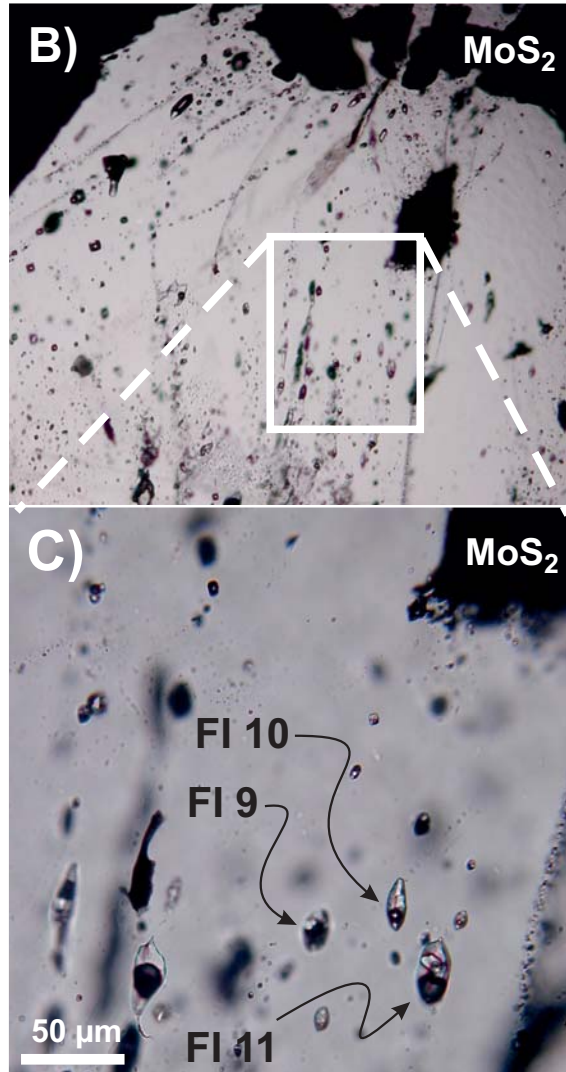
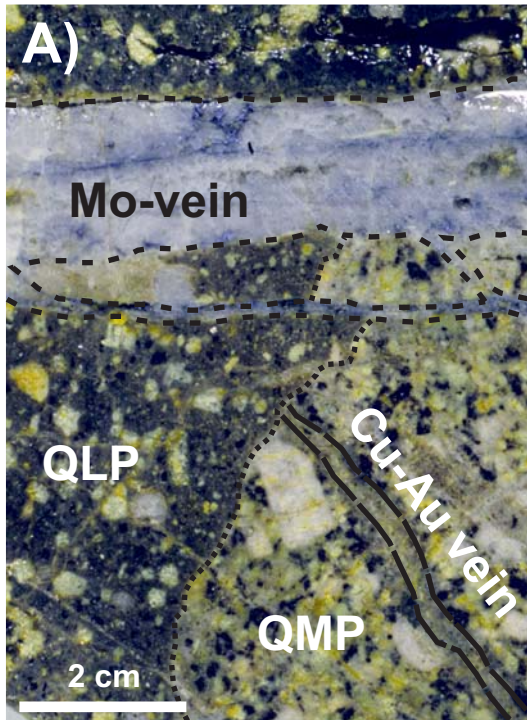


Figure 2

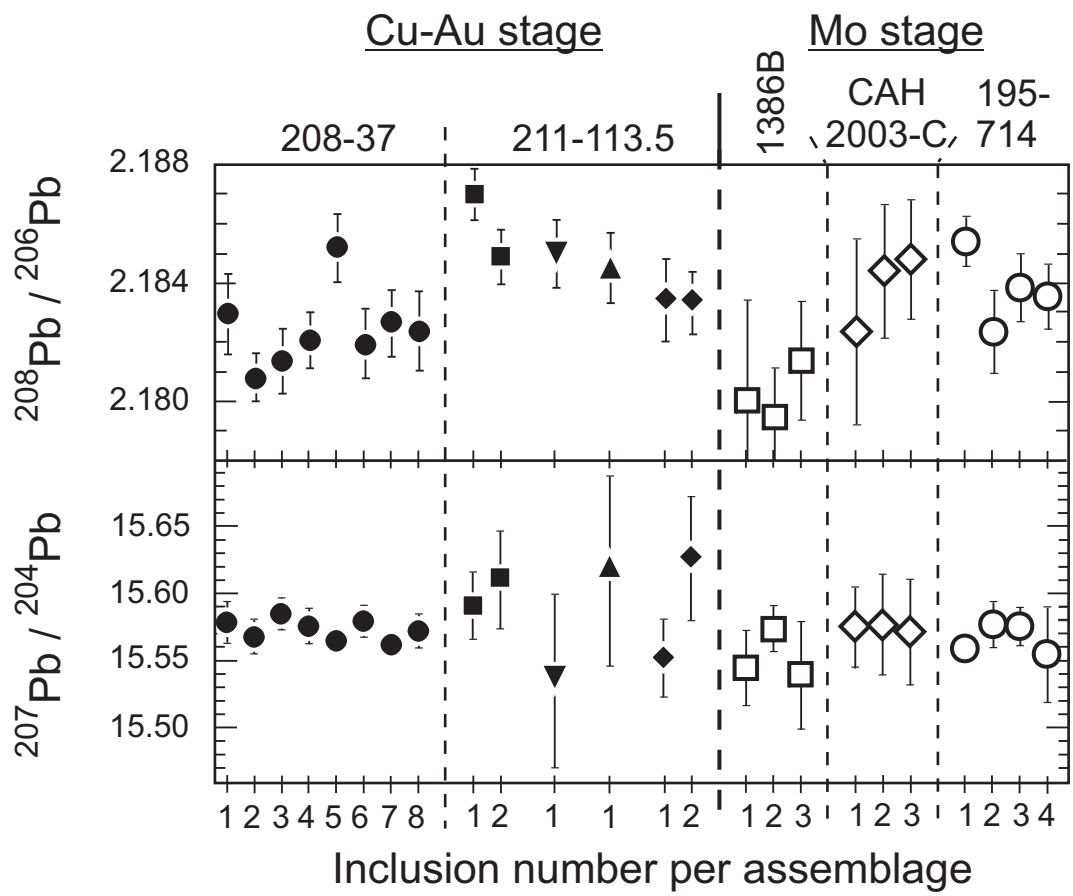
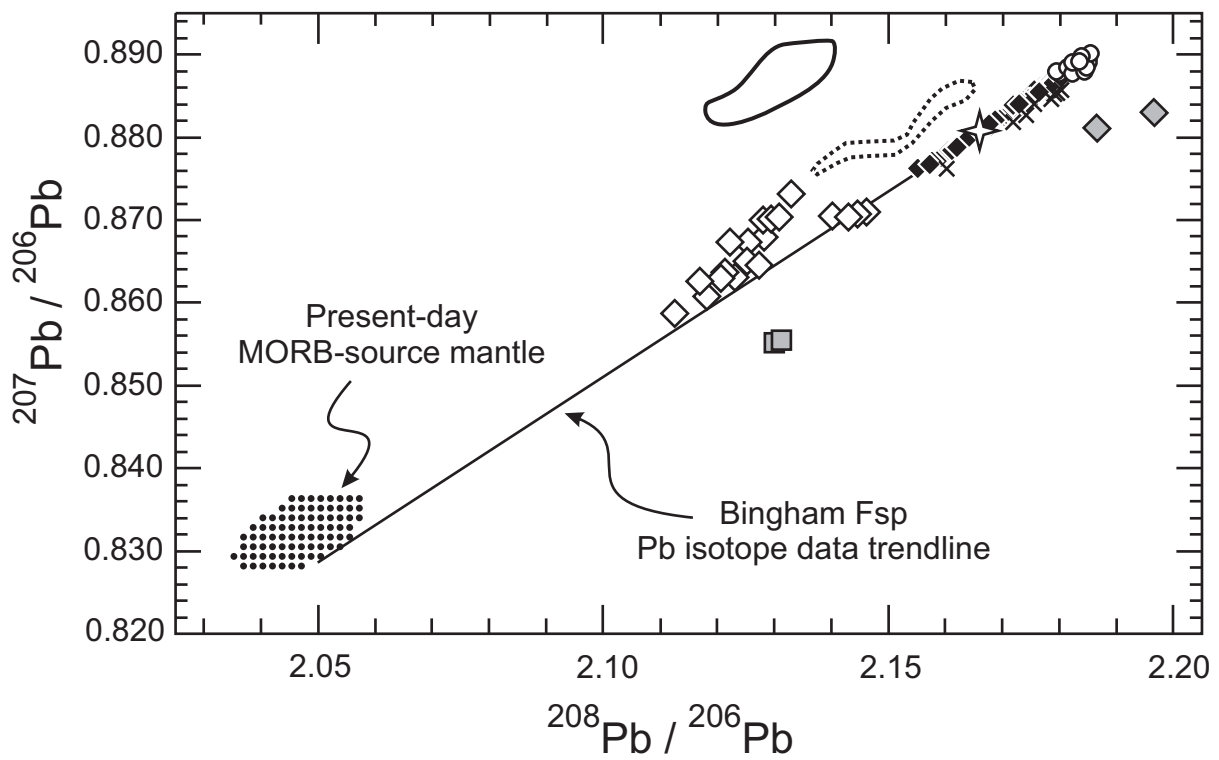


Figure 3



- |            |                                 |           |   |
|------------|---------------------------------|-----------|---|
| Bingham: ○ | Ore fluids                      | Butte: ◇  | Magmatic rocks                                  |
| ×          | Galena                          | Questa: ◌ | Magmatic rocks                                  |
| ◆          | Feldspars                       | SEAZ: ⋯   | Magmatic rocks and hydrothermal high-T sulfides |
| ◇          | Shoshonite                      |           |   |
| ■          | Melanephelinite                 |           |   |
| ✦          | Equigranular Monzonite, altered |           |   |

Figure 4

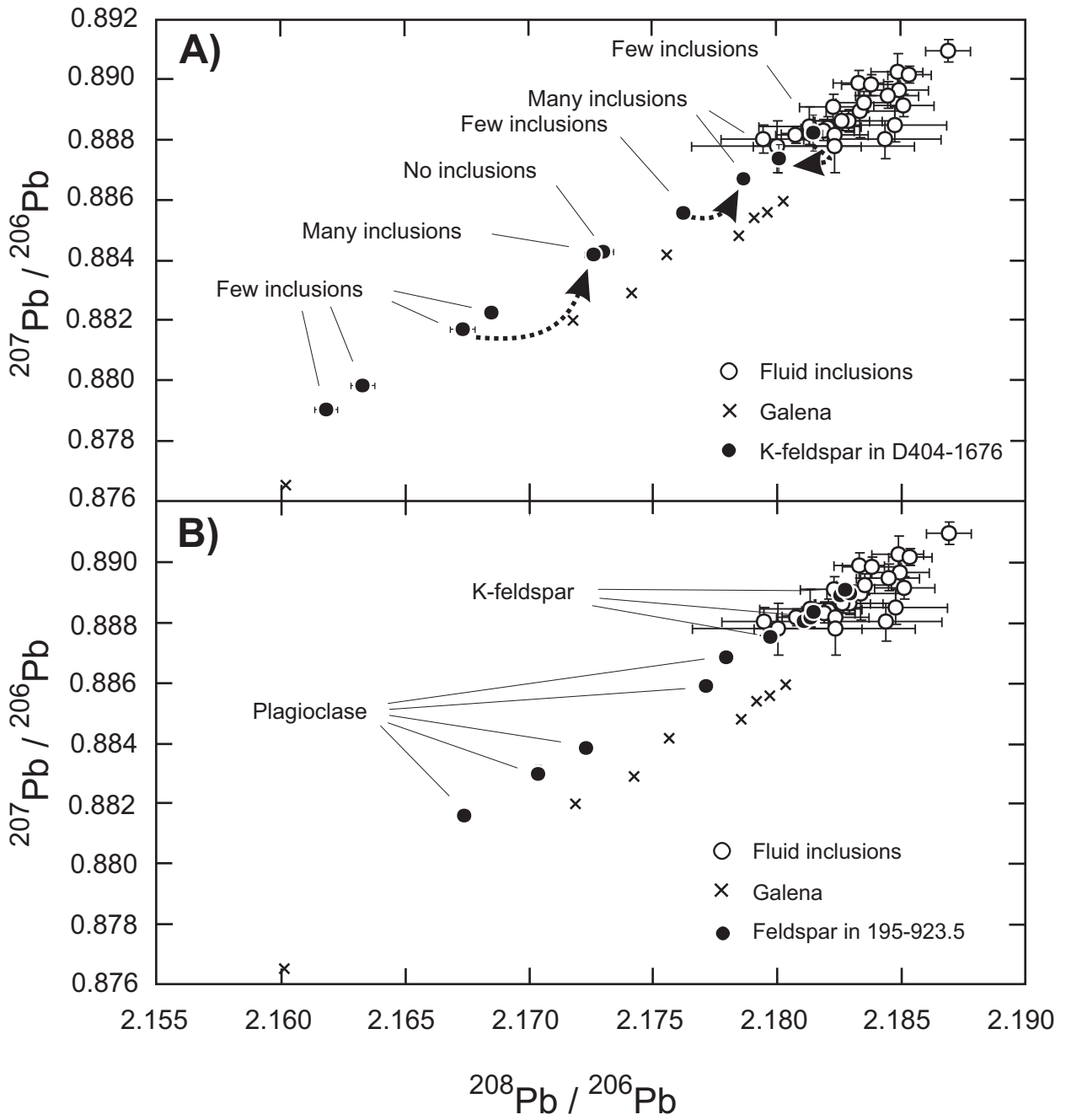


Figure 5



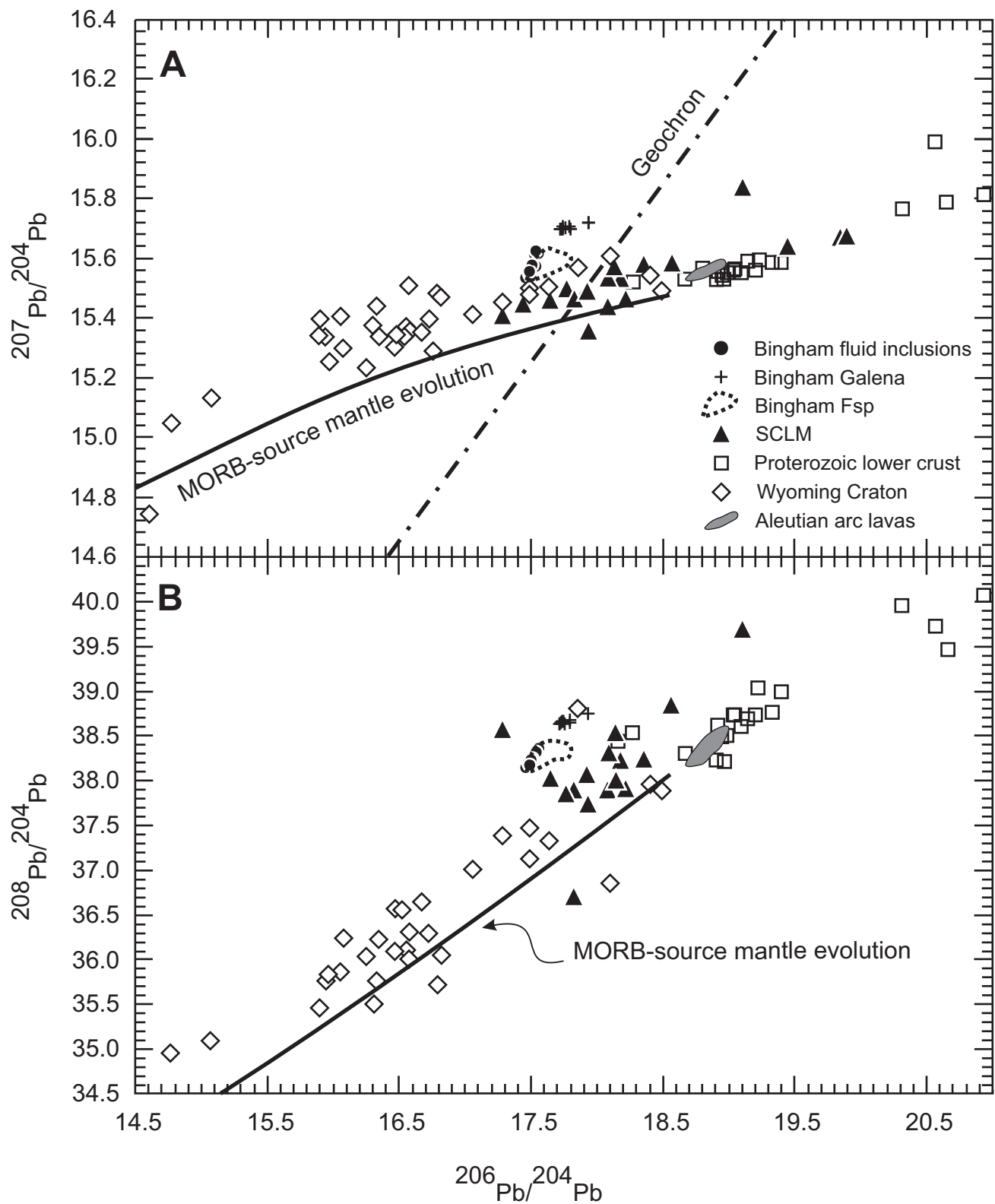


Figure 6

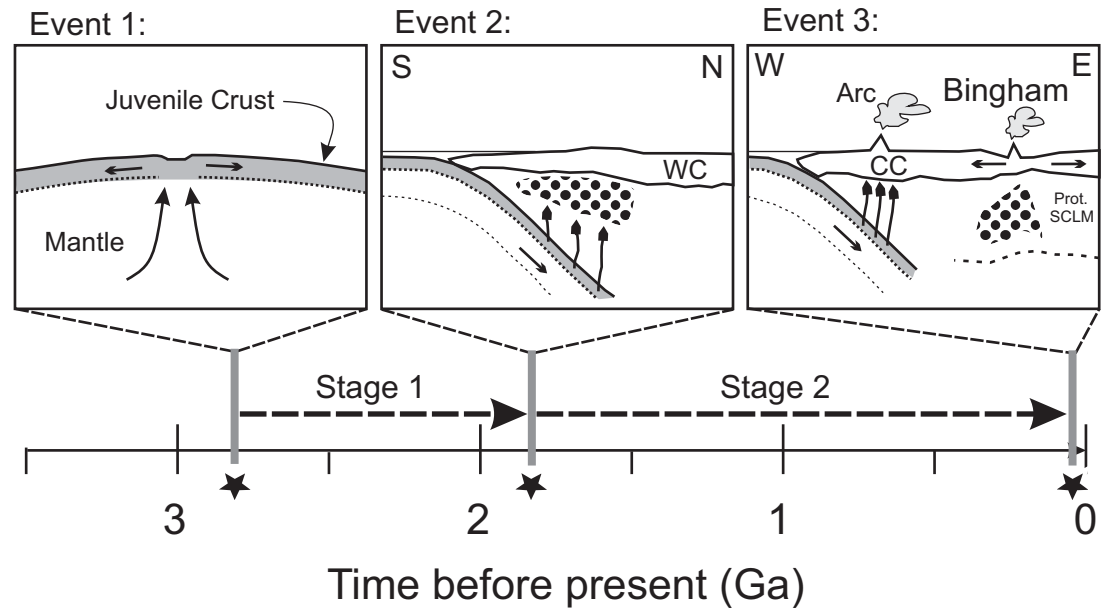


Figure 7

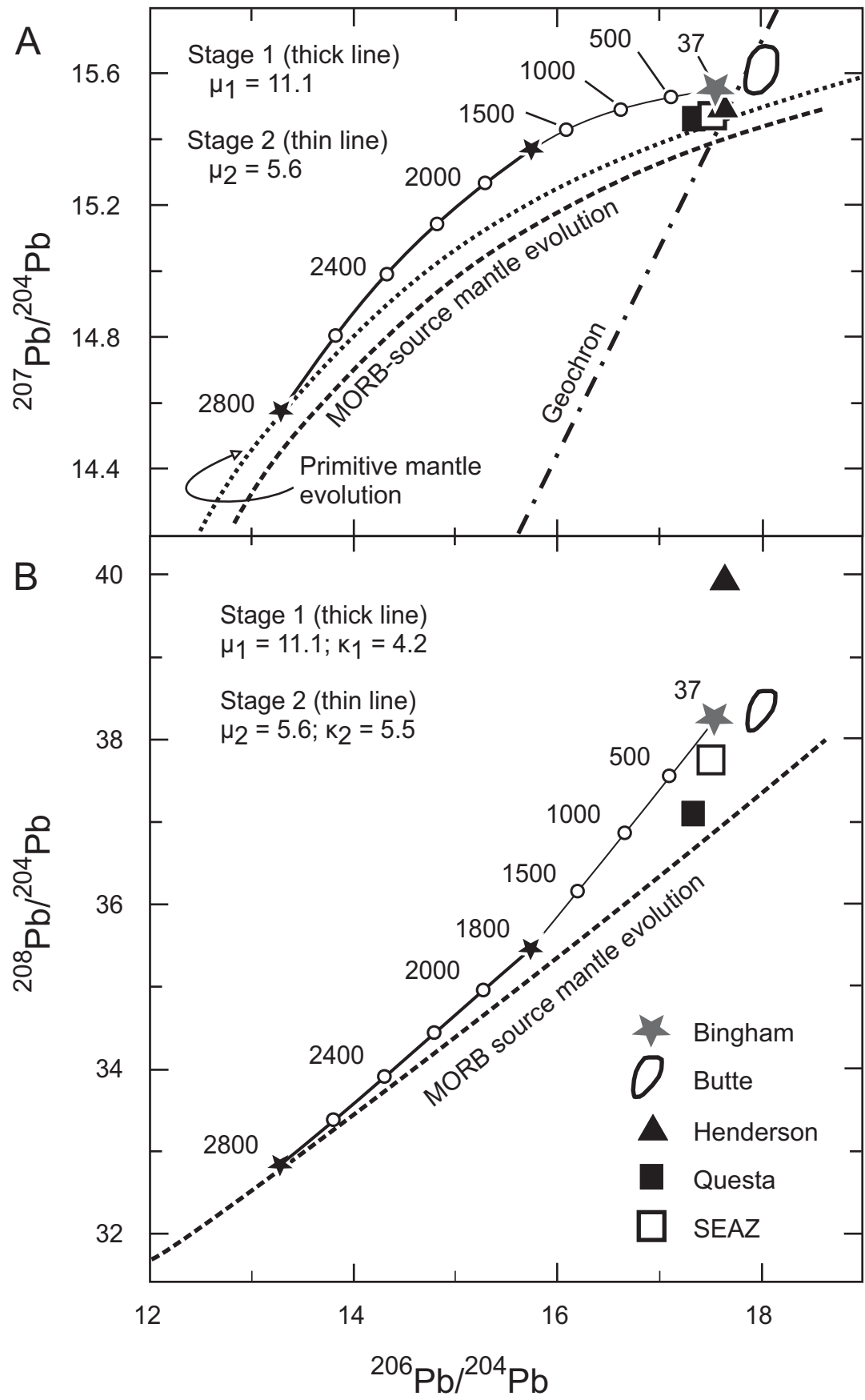


Figure 8

**Table 1: Pb isotope data of individual fluid inclusions (brines) from Cu-Au and Mo stage mineralizations at Bingham Canyon, USA**

Sample: FI number	<sup>208</sup> Pb/ <sup>206</sup> Pb	2 s.e.m. (meas)	<sup>207</sup> Pb/ <sup>206</sup> Pb	2 s.e.m. (meas)	<sup>206</sup> Pb/ <sup>204</sup> Pb	2 s.e.m. (meas)	<sup>207</sup> Pb/ <sup>204</sup> Pb	2 s.e.m. (meas)	<sup>208</sup> Pb/ <sup>204</sup> Pb	2 s.e.m. (meas)	Sample:	Mo (µg/g)	Cu (µg/g)	Au (µg/g)	Pb (µg/g)	U (µg/g)	Th (µg/g)
<b>Porphyry Cu-Au stage</b>																	
208-37: 1	2.1829	0.0014	0.88860	0.00039	17.518	0.016	15.578	0.015	38.281	0.043							
208-37: 2	2.1808	0.0008	0.88811	0.00029	17.529	0.014	15.568	0.013	38.232	0.037							
208-37: 3	2.1813	0.0011	0.88817	0.00036	17.539	0.013	15.586	0.012	38.278	0.033							
208-37: 4	2.1821	0.0011	0.88838	0.00036	17.526	0.014	15.575	0.013	38.259	0.037							
208-37: 5	2.1852	0.0012	0.88911	0.00036	17.505	0.012	15.565	0.010	38.264	0.027							
208-37: 6	2.1819	0.0012	0.88829	0.00036	17.530	0.012	15.578	0.012	38.266	0.033							
208-37: 7	2.1826	0.0011	0.88860	0.00033	17.509	0.008	15.562	0.008	38.225	0.024							
208-37: 8	2.1824	0.0013	0.88813	0.00039	17.523	0.012	15.573	0.013	38.269	0.037							
<b>Assemblage average</b>	<b>2.1824</b>		<b>0.88842</b>		<b>17.522</b>		<b>15.573</b>		<b>38.259</b>		<b>208-37</b> §	<b>86</b>	<b>11,400</b>	<b>0.077</b>	<b>3,610</b>	<b>n.m.</b>	<b>n.m.</b>
<b>Assemblage 2 s.d.</b>	<b>0.0026</b>		<b>0.00068</b>		<b>0.022</b>		<b>0.016</b>		<b>0.041</b>			<b>28</b>	<b>6,700</b>	<b>0.022</b>	<b>520</b>	<b>n.m.</b>	
211-113.5M: 19 <sup>§</sup>	2.1870	0.0009	0.89093	0.00036	17.519	0.029	15.591	0.025	38.293	0.065							
211-113.5M: 20 <sup>§</sup>	2.1849	0.0010	0.89023	0.00062	17.529	0.046	15.611	0.036	38.281	0.093							
211-113.5M: 21 <sup>§</sup>	2.1850	0.0012	0.88961	0.00070	17.456	0.071	15.534	0.065	38.142	0.156							
211-113.5M: 22 <sup>§</sup>	2.1845	0.0013	0.88945	0.00044	17.559	0.081	15.618	0.073	38.355	0.181							
<b>Sample average</b>	<b>2.1853</b>		<b>0.89006</b>		<b>17.516</b>		<b>15.588</b>		<b>38.268</b>		<b>211-19, trail G</b> †	<b>42</b>	<b>26,100</b>	<b>n.m.</b>	<b>2,420</b>	<b>3</b>	<b>&lt;1</b>
<b>Sample 2 s.d.</b>	<b>0.0022</b>		<b>0.00135</b>		<b>0.087</b>		<b>0.076</b>		<b>0.180</b>			<b>9</b>	<b>8,300</b>	<b>n.m.</b>	<b>270</b>	<b>1</b>	
211-113.5N: 23 <sup>§</sup>	2.1834	0.0014	0.88893	0.00088	17.494	0.035	15.551	0.029	38.186	0.080							
211-113.5N: 24 <sup>§</sup>	2.1833	0.0010	0.88984	0.00042	17.536	0.052	15.624	0.047	38.314	0.111							
<b>Assemblage average</b>	<b>2.1834</b>		<b>0.88938</b>		<b>17.515</b>		<b>15.587</b>		<b>38.250</b>		<b>211-19, trail F</b> †	<b>39</b>	<b>11,000</b>	<b>n.m.</b>	<b>3,100</b>	<b>5</b>	<b>n.m.</b>
<b>Assemblage 2 s.d.</b>	<b>0.0001</b>		<b>0.00128</b>		<b>0.060</b>		<b>0.103</b>		<b>0.181</b>			<b>22</b>	<b>7,500</b>	<b>n.m.</b>	<b>440</b>	<b>3</b>	
<b>Molybdenite stage</b>																	
1386B: 9	2.1800	0.0034	0.88773	0.00087	17.488	0.028	15.547	0.028	38.204	0.087							
1386B: 10	2.1795	0.0016	0.88798	0.00047	17.517	0.017	15.572	0.017	38.230	0.049							
1386B: 11	2.1814	0.0021	0.88843	0.00063	17.493	0.044	15.540	0.039	38.179	0.104							
<b>Assemblage average</b>	<b>2.1803</b>		<b>0.88805</b>		<b>17.500</b>		<b>15.553</b>		<b>38.204</b>		<b>JH2_BT2</b> *	<b>88</b>	<b>1,190</b>	<b>1.28</b>	<b>3,800</b>	<b>n.m.</b>	<b>n.m.</b>
<b>Assemblage 2 s.d.</b>	<b>0.0020</b>		<b>0.00070</b>		<b>0.032</b>		<b>0.034</b>		<b>0.051</b>			<b>87</b>	<b>950</b>	<b>1.03</b>	<b>970</b>		
CAH-2003-C: 12	2.1823	0.0032	0.88773	0.00085	17.543	0.030	15.574	0.030	38.307	0.077							
CAH-2003-C: 13	2.1844	0.0023	0.88799	0.00063	17.525	0.044	15.575	0.038	38.312	0.112							
CAH-2003-C: 14	2.1848	0.0021	0.88847	0.00057	17.535	0.046	15.571	0.039	38.313	0.102							
<b>Assemblage average</b>	<b>2.1838</b>		<b>0.88806</b>		<b>17.534</b>		<b>15.574</b>		<b>38.311</b>		<b>JH2-a7</b> *	<b>33</b>	<b>18,900</b>	<b>0.31</b>	<b>3,000</b>	<b>n.m.</b>	<b>n.m.</b>
<b>Assemblage 2 s.d.</b>	<b>0.0026</b>		<b>0.00074</b>		<b>0.019</b>		<b>0.004</b>		<b>0.007</b>			<b>32</b>	<b>7,500</b>	<b>0.12</b>	<b>230</b>		
195-714: 15	2.1854	0.0009	0.89012	0.00029	17.479	0.010	15.559	0.009	38.210	0.025							
195-714: 16	2.1823	0.0014	0.88906	0.00042	17.509	0.020	15.577	0.017	38.246	0.051							
195-714: 17	2.1838	0.0012	0.88979	0.00036	17.503	0.016	15.576	0.014	38.232	0.038							
195-714: 18	2.1835	0.0011	0.88921	0.00036	17.486	0.038	15.555	0.036	38.176	0.085							
<b>Assemblage average</b>	<b>2.1838</b>		<b>0.88955</b>		<b>17.494</b>		<b>15.567</b>		<b>38.216</b>								
<b>Assemblage 2 s.d.</b>	<b>0.0025</b>		<b>0.00099</b>		<b>0.029</b>		<b>0.023</b>		<b>0.061</b>								

**Notes:** Fluid inclusions were analyzed using a Nu Plasma 1700 ICP-MS, except for samples 211-113.5, analyzed using a Nu Plasma ICP-MS  
 For inclusions, uncertainties are reported as twice the standard error of the mean of measurement (2 s.e.m.)  
 For assemblage or sample averages, the uncertainty given is twice the standard deviation of the population (2 s.d.)  
 Sample average denotes the average fluid composition of inclusions belonging to three fluid inclusion assemblages of that sample  
 n.m. not measured  
 § Data from this study. See Pettke (2008) for analytical details  
 † Data from Landtwing (2004)  
 \* Data from Seo et al. (2009)

**Table 2: Parameters explored in Monte Carlo simulations of Pb isotope evolution to reproduce the measured Bingham fluid Pb isotope data**

Simulation number	$\mu$ post-diff	T1 (Ga)	$^{206}\text{Pb}/^{204}\text{Pb}$ init (at T1)	$^{207}\text{Pb}/^{204}\text{Pb}$ init (at T1)	$^{208}\text{Pb}/^{204}\text{Pb}$ init (at T1)	$\mu_1$	$\kappa_1$	T2 (Ga)	$\mu_2$	$\kappa_2$
1	—	2.8 ±0.1	12.88 - 13.21	14.25 -14.42	32.44 - 32.75	14.0 - 18.0	3.7 - 4.7	1.8 ±0.1	2.3 - 3.9	5.3 - 10.2
2	—	3.2 ±0.1	12.13 - 12.51	13.79 - 14.04	31.72 - 32.09	11.1 - 12.6	3.7 - 4.7	1.8 ±0.1	3.8 - 5.0	4.8 - 7.9
3	—	2.8 ±0.1	12.88 - 13.21	14.25 -14.42	32.44 - 32.75	14.0 - 18.0	4.5 - 5.5	1.8 ±0.1	2.3 - 3.9	2.5 - 6.9
4	8.0 ±0.2	2.8 ±0.1	12.82 - 13.37	14.16 - 14.58	32.24 - 33.07	11.6 - 19.8	3.7 - 4.7	1.8 ±0.1	1.2 - 5.3	4.8 - 13.1
5	8.5 ±0.2	2.8 ±0.1	13.03 - 13.63	14.41 - 14.85	32.50 - 33.30	7.3 - 13.9	3.7 - 4.7	1.8 ±0.1	4.1 - 7.7	4.6 - 7.0
6	9.0 ±0.2	2.8 ±0.1	13.26 - 13.87	14.65 - 15.12	23.66 - 33.52	1.1 - 8.2	3.7 - 4.7	1.8 ±0.1	7.1 - 10.6	4.5 - 5.6

**Parameters used for modelling Pb evolution:**

The target Pb isotope signature for the simulations is that measured in Bingham fluid inclusions:

$$^{206}\text{Pb}/^{204}\text{Pb} = 17.515, \quad ^{207}\text{Pb}/^{204}\text{Pb} = 15.573, \quad ^{208}\text{Pb}/^{204}\text{Pb} = 38.252$$

Simulations 1-3 derive the starting Pb compositions for stage 1 from the model of Kramers and Tolstikhin(1997)

$\mu$ - and  $\kappa$ -values prior to core segregation are  $0.85 \pm 0.15$  and  $3.92 \pm 0.10$ , respectively

$\mu$  post-diff       $\mu$  value after Earth's core segregation at  $4.52 \pm 0.010$  Ga (Kleine et al., 2002, Schoenberg et al., 2002)

$\kappa$  post-diff       $\kappa$  value after Earth's core segregation, set at  $3.92 \pm 0.20$

T1                      Start of stage 1 (in billion years before present)

Pb init                Range of initial Pb isotopic compositions at T1

$\mu_1$  and  $\kappa_1$         Range of  $\mu$  and  $\kappa$  values of stage 1

T2                      Start of stage 2 set to 1.8 billion years before present

$\mu_2$  and  $\kappa_2$         Range of  $\mu$  and  $\kappa$  values of stage 2

# Supporting Online Material:

## The magma and metal source of giant porphyry-type ore deposits, based on lead isotope microanalysis of individual fluid inclusions

by Thomas Pettke, Felix Oberli and Christoph A. Heinrich

All LA-MC-ICP-MS Pb isotope analyses were performed at ETH Zurich using a GeoLas 200Q (Lambda Physik, Germany) laser system with computer-controlled sample stage connected to either a Nu Plasma 1700 or a Nu Plasma MC-ICP-MS instrument (Nu Instruments Ltd, Wrexham, UK). Table ES 1 summarizes the operating conditions for LA-ICP-MS analysis of Pb isotopes.

**Table ES 1:**  
**LA-MC-ICP-MS instrument and data acquisition parameters**

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### 193 nm ArF Excimer laser Compex 110I

- Energy density on sample ( $\text{J}/\text{cm}^2$ )	ca. 16, homogeneous across the ablation crater
- Pulse duration (ns)	ca. 15
- Repetition rate (Hz)	SRM (line scan): 6; SRM, FI and FSP (single spot): 10
- Shooting mode	$1 \mu\text{m s}^{-1}$ (line scan), single spot (SRM, FI, FSP)
- Pit sizes ( $\mu\text{m}$ )	SRM: 60 or 90; FI: 8 - 80; FSP: 80 - 120
- Ablation cell volume ( $\text{cm}^3$ )	FI: 1, variable for SRM and FSP (1 - 16)
- Helium Cell gas flow ( $\text{l min}^{-1}$ )	0.6

### Nu Plasma 1700 MC-ICP-MS \*

- Desolvator unit	DSN-100 desolvation nebulizer system
- Membrane Ar flow ( $\text{l min}^{-1}$ )	2.9 - 3.1 (2.70 - 3.20)
- Auxiliary Ar flow ( $\text{l min}^{-1}$ )	0.85 - 0.90 (0.75 - 1.00)
- Coolant Ar flow ( $\text{l min}^{-1}$ )	13.0 (13.0 - 15.0)
- Power (W)	1,450 (1,100 - 1,550) fwd.; <2 reflected power
- Accelerating voltage (kV)	6
- Detector mode	Faraday, using $10^{11} \Omega$ feedback resistors
- Interface pressure (mbar)	0.8 - 1.0
- Mass resolution (10% valley)	ca. 700

### Data acquisition parameters during transient signal analysis

- Acquisition mode	static, time-resolved
- Integration time	0.2 s
- Masses analyzed	200, 202, 203, 204, 205, 206, 207, 208

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#### Notes:

Values reported in parentheses are the ranges explored during method development

SRM refers to SRM 610 glass from NIST

FI refers to fluid inclusions

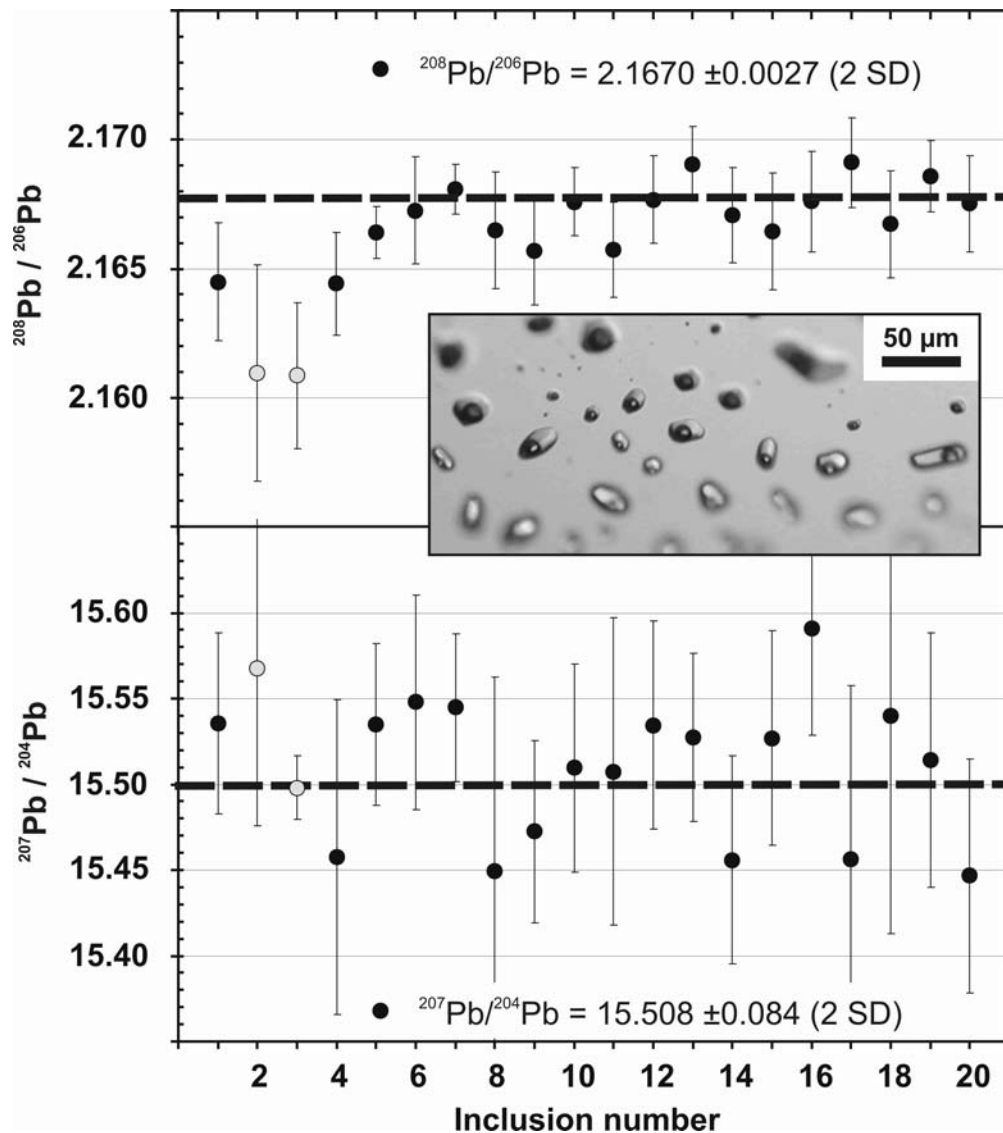
FSP refers to plagioclase or K-feldspar phenocrysts

\* Operating conditions used at Nu Plasma MC-ICP-MS are similar to those reported here, with the exception of acceleration voltage (4kV) and mass resolution (ca. 400).

The MC-ICP-MS instrument was optimized daily for maximum sensitivity and perfect peak flatness and coincidence by admixing a desolvated aerosol generated from a 30 ppb Pb - 32 ppb Tl solution to the He flow from the LA chamber. Minor retuning was then performed by laser ablation on SRM 610 in line scan mode. The analyses were performed in static time-resolved mode, collecting the analytes simultaneously in Faraday cups calibrated daily for their preamplifier gains. For sample analysis, a background signal with laser off was acquired, then the Tl-solution was aspirated, and once the Tl signal was stable, LA was started, superimposing the sample signal on the Tl signal from the desolvating unit. Each ablation chamber loading was bracketed by 3 SRM 610 analyses to monitor instrument conditions.

Transient signal data reduction was done by revisiting the individually stored readings using modified Nu Instruments transient signal software, followed by off-line evaluation on Excel spreadsheets. After background correction of the analyte signals, Hg interference correction on raw mass 204 was effected based on the measured  $^{202}\text{Hg}$  beam and a  $^{204}\text{Hg}/^{202}\text{Hg}$  ratio of 4.32, adjusted to the fractionated state by use of an exponential mass bias coefficient derived from the measured  $^{205}\text{Tl}/^{203}\text{Tl}$  ratio and its common value of 2.3871 (Dunstan et al., 1980). Final  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios (corrected for mass bias) do not correlate with  $^{202}\text{Hg}/\text{Pb}_{(\text{total})}$  ratios (not shown), thus demonstrating proper correction for Hg interference. Mass bias correction was exclusively done in within-run mode, using the Baxter et al. (2006) approach. Measurements of SRM 610 from NIST in line scan mode from >10 analytical sessions were used to define the mass bias, i.e., the instrument-specific *InTl - InPb* relationship for each Pb isotope ratio. For each sample analysis, the measured  $^{205}\text{Tl}/^{203}\text{Tl}$  isotope ratio was then used to correct for mass bias employing the *InTl - InPb* relationship established on the SRM 610 measurements. The sample Pb isotope ratios were obtained through individual processing of readouts from 0.2 s integration time slices. Data from each time-slice are individually treated for interference and mass bias correction, and the means calculated from these individual results then provide the isotope ratios of the sample.

Test measurements were performed on individual inclusions from synthetic fluid inclusions standards (prepared by A. Audetat, University of Bayreuth) containing ca. 5000  $\mu\text{g}$  SRM 981 Pb per g of NaCl-KCl solution of ca. 17 wt-% bulk salinity. Accurate Pb isotope data can be obtained on fluid inclusion assemblages with 2 SD uncertainties on inclusion-to-inclusion reproducibility of 0.06 % ( $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios) and 0.3% (Pb isotope ratios involving mass 204), respectively (Fig. S1). This is only possible, however, given controlled ablation of the entire fluid inclusion and integration of the entire transient signal (Pettke et al., 2008), in order to minimize bias originating from differences in signal decay parameters among the Faraday amplifiers used to record fast transient signals in multi-collector mode (more details to be published elsewhere). Acceptably reproducible results ( $\pm 1\%$  and  $5\%$ , respectively) were obtained for inclusions containing as little as 0.005 ng Pb using our setup with Faraday detectors.



**Fig. S1:**

Thallium-normalized  $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios of individual synthetic fluid inclusions (inset) containing ca. 5000  $\mu\text{g}$  SRM 981 Pb per gram of Na-K-Cl fluid. Average measured isotopic ratios are reported with their 2 standard error uncertainty and reproduce the reference values of the standard (Baker et al., 2004) shown by the thick dashed lines. Analyses represented by grey symbols were excluded from the data set because fluid inclusion ablation was not well controlled.

## References

- Baxter D. C., Rodushkin I., Engstrom E., Malinovsky D., 2006. Revised exponential model for mass bias correction using an internal standard for isotope abundance ratio measurements by multi-collector inductively coupled plasma mass spectrometry. *J. Anal. Atom. Spectrom.* 21, 427-430.
- Dunstan L. P., Gramlich J. W., Barnes I. L., Purdy W. C., 1980. Absolute isotopic abundance and the atomic weight of a reference sample of thallium. *J. Res. National Bureau of Standards* 85, 1-10.
- Pettke T., Oberli F., Audetat A., Wiechert U., Harris C. R., Heinrich C. A., 2008. Precise and accurate lead isotopic analysis of fast transient signals by laser-ablation MC-ICP-MS. *Geochim. Cosmochim. Acta* 72, A741-A741.



**Table ES 2: Pb isotope data of feldspar (LA-MC-ICP-MS) and bulk rocks (solution MC-ICP-MS) from Bingham Canyon, and U-Th-Pb concentration data (LA-ICP-MS)**

Spot Number	Phase	Element conc. analysis Comments	Crater size (µm)	Pb (µg/g)	Th (µg/g)	U (µg/g)	Pb isot. analysis Comments	208Pb/206Pb	1 s.e.m. (meas)	207Pb/206Pb	1 s.e.m. (meas)	206Pb/204Pb	1 s.e.m. (meas)	207Pb/204Pb	1 s.e.m. (meas)	208Pb/204Pb	1 s.e.m. (meas)
<b>A57-5922.6 (Mine grids: -2403.21 E, +3329.77 N): Quartz Monzonite Porphyry</b>																	
1	Plag An23	clean	60	21.4	<0.005	<0.002	clean	2.17818	0.00010	0.88713	0.00009	17.587	0.016	15.599	0.014	38.309	0.035
2	Plag An24	clean	90	22.3	<0.002	<0.002	clean	2.17819	0.00013	0.88693	0.00011	17.540	0.017	15.561	0.016	38.223	0.039
3	Plag						clean	2.17992	0.00009	0.88779	0.00009	17.524	0.015	15.562	0.014	38.211	0.034
4	Plag An25	clean	90	22.9	21.6	<0.002	clean	2.17274	0.00016	0.88451	0.00013	17.574	0.022	15.545	0.020	38.183	0.050
5	Plag						clean	2.17399	0.00011	0.88451	0.00009	17.629	0.015	15.596	0.013	38.331	0.033
6	Plag An23	clean	60	22.6	21.5	<0.005	clean	2.17631	0.00010	0.88625	0.00016	17.593	0.034	15.591	0.030	38.261	0.075
7	Plag An24	clean	90	23.5	22.3	<0.002	clean	2.17716	0.00012	0.88646	0.00011	17.618	0.018	15.625	0.017	38.371	0.041
8	Plag						clean	2.16766	0.00013	0.88153	0.00011	17.662	0.020	15.574	0.018	38.288	0.044
<b>D195-567 (Mine grids: -1438.73 E, +447.51 N): Quartz Latite Porphyry</b>																	
1	AKF	clean	90	61.4	<0.002	<0.002	clean	2.17545	0.00011	0.88553	0.00008	17.626	0.015	15.611	0.013	38.342	0.033
2	AKF						clean	2.17344	0.00012	0.88448	0.00005	17.604	0.008	15.572	0.007	38.270	0.017
3	AKF						clean	2.17346	0.00009	0.88446	0.00006	17.601	0.011	15.568	0.010	38.257	0.024
4	AKF	clean	90	44.5	<0.004	<0.002	clean	2.18213	0.00009	0.88881	0.00005	17.517	0.007	15.573	0.006	38.239	0.015
5	AKF	clean	60	43.9	<0.002	<0.006	clean	2.18047	0.00010	0.88797	0.00007	17.544	0.012	15.587	0.011	38.264	0.027
6	Plag An23	clean, next to 120 µm pit	60	20.8	<0.007	<0.005	clean	2.18218	0.00010	0.88870	0.00008	17.516	0.015	15.563	0.014	38.216	0.034
6	Plag An24	clean	90	21.4	<0.002	0.002											
7	Plag An24	clean	60	22.5	<0.002	<0.005	clean	2.17392	0.00010	0.88451	0.00009	17.648	0.014	15.611	0.013	38.370	0.032
<b>D195-923.5 (Mine grids: -1438.73 E, +447.51 N): Quartz Latite Porphyry</b>																	
1	AKF						some inclusions	2.17970	0.00013	0.88755	0.00012	17.575	0.018	15.596	0.016	38.307	0.041
2	AKF						inclusions	2.18108	0.00009	0.88808	0.00005	17.530	0.007	15.570	0.006	38.237	0.015
3	AKF	clean	60	36.2	<0.008	<0.004	clean	2.18133	0.00009	0.88819	0.00007	17.579	0.013	15.615	0.012	38.352	0.029
4	AKF	many inclusions, next to pit	90	61.6	<0.004	0.004	many inclusions	2.18144	0.00010	0.88838	0.00005	17.533	0.006	15.578	0.005	38.255	0.013
4	AKF	many inclusions, next to pit	90	48.4	<0.002	0.010											
5	AKF	clean	60	39.4	<0.006	<0.008	few inclusions	2.18252	0.00010	0.88894	0.00005	17.510	0.008	15.570	0.007	38.226	0.018
6	AKF						many inclusions	2.18289	0.00017	0.88900	0.00011	17.526	0.015	15.585	0.014	38.246	0.033
7	AKF	clean, next to 120 µm pit	60	46.5	<0.004	<0.002	clean	2.18270	0.00009	0.88910	0.00005	17.530	0.006	15.586	0.006	38.270	0.014
8	Plag An24	clean	90	22.5	<0.002	<0.002	clean	2.17229	0.00010	0.88387	0.00007	17.663	0.014	15.613	0.013	38.374	0.032
9	Plag						clean	2.17038	0.00008	0.88302	0.00013	17.692	0.026	15.620	0.023	38.403	0.058
10	Plag An24	clean	60	21.2	<0.005	<0.005	clean	2.16742	0.00010	0.88163	0.00010	17.683	0.015	15.595	0.014	38.339	0.034
11	Plag An23	clean	60	22.0	<0.005	<0.002	clean	2.17792	0.00009	0.88687	0.00012	17.565	0.029	15.578	0.026	38.248	0.063
12	Plag						clean	2.17714	0.00014	0.88592	0.00011	17.606	0.018	15.601	0.017	38.335	0.041
<b>D195-1006 (Mine grids: -1438.73 E, +447.51 N): Biotite Porphyry</b>																	
1	Plag An24	clean, near pit	60	20.5	<0.005	<0.005	clean	2.17537	0.00011	0.88540	0.00010	17.585	0.016	15.567	0.015	38.267	0.037
2	Plag An23	clean	90	21.4	<0.002	0.002	clean	2.17521	0.00009	0.88516	0.00008	17.569	0.015	15.554	0.013	38.202	0.032
3	Plag						few inclusions	2.17621	0.00012	0.88577	0.00011	17.539	0.019	15.540	0.017	38.174	0.043
4	Plag						few inclusions	2.17869	0.00014	0.88657	0.00030	17.567	0.022	15.577	0.021	38.237	0.056
5	Plag An22	clean, between 2 pits	90	23.9	<0.002	<0.002	clean	2.17529	0.00009	0.88594	0.00029	17.542	0.032	15.548	0.029	38.197	0.069
12	Plag						clean	2.17327	0.00010	0.88453	0.00008	17.601	0.016	15.569	0.014	38.263	0.034
6	Plag						clean	2.17459	0.00010	0.88486	0.00008	17.606	0.015	15.576	0.014	38.303	0.035
7	Plag An23	clean, near pit	90	25.0	<0.002	<0.002	clean	2.17077	0.00011	0.88335	0.00010	17.621	0.017	15.568	0.015	38.264	0.037
8	Plag An24	clean	90	25.0	0.002	<0.005	clean	2.17135	0.00011	0.88364	0.00011	17.620	0.018	15.575	0.016	38.265	0.040
8	Plag An24						wider interval	2.17166	0.00009	0.88408	0.00010	17.596	0.014	15.561	0.012	38.217	0.031
9	Plag						clean	2.17540	0.00011	0.88509	0.00009	17.577	0.018	15.559	0.016	38.239	0.039
10	Plag						clean	2.17185	0.00011	0.88383	0.00009	17.583	0.017	15.542	0.015	38.201	0.037
11	Plag						clean	2.17421	0.00010	0.88479	0.00009	17.585	0.016	15.568	0.014	38.250	0.035
<b>D195-1181 (Mine grids: -1438.73 E, +447.51 N): Quartz Latite Porphyry</b>																	
1	AKF	near pits	60	44.1	<0.004	<0.004	clean	2.17460	0.00016	0.88469	0.00012	17.608	0.019	15.579	0.017	38.283	0.043
2	AKF						clean	2.17517	0.00018	0.88530	0.00014	17.620	0.030	15.601	0.027	38.353	0.070
3	AKF	clean	60	50.4	<0.004	<0.004	clean	2.17002	0.00013	0.88280	0.00009	17.659	0.016	15.590	0.014	38.323	0.034
4	AKF	clean, next to 120 µm pit	90	57.1	<0.004	<0.002	clean	2.17802	0.00010	0.88646	0.00005	17.591	0.008	15.595	0.007	38.317	0.018
5	AKF						clean	2.17080	0.00014	0.88303	0.00007	17.668	0.010	15.602	0.009	38.360	0.022
6	AKF	clean	90	51.8	<0.002	<0.002	clean	2.17255	0.00011	0.88375	0.00008	17.652	0.012	15.604	0.011	38.352	0.027
7	AKF	clean, next to 120 µm pit	60	39.3	<0.004	<0.004	clean	2.17289	0.00011	0.88393	0.00005	17.642	0.006	15.596	0.005	38.337	0.014

Table ES 2: continued

Spot number	Phase	Element conc. analysis Comments	crater size (µm)	Pb (µg g <sup>-1</sup> )	Th (µg g <sup>-1</sup> )	U (µg g <sup>-1</sup> )	Pb isot. analysis Comments	208Pb/206Pb final	1 s.e.m. (meas)	207Pb/206Pb final	1 s.e.m. (meas)	206Pb/204Pb (meas)	1 s.e.m. (meas)	207Pb/204Pb final	1 s.e.m. (meas)	208Pb/204Pb final	1 s.e.m. (meas)
<b>D450-1676 (Mine grids: -1652.83 E, -47.44 N): Quartz Monzonite Porphyry</b>																	
1	AKF	few small inclusions	60	35.2	<0.002	<0.002	inclusions	2.17624	0.00015	0.88561	0.00011	17.609	0.016	15.605	0.014	38.330	0.035
1							Inclusion signal	2.17868	0.00012	0.88674	0.00010	17.560	0.015	15.572	0.012	38.261	0.029
2	AKF	several small inclusions	60	30.4	0.014	0.008	inclusions	2.16851	0.00015	0.88227	0.00007	17.669	0.011	15.591	0.009	38.320	0.024
3	AKF	no inclusions	44	29.9	<0.006	0.008	clean	2.17300	0.00021	0.88431	0.00010	17.621	0.015	15.584	0.013	38.316	0.034
4	AKF						inclusions	2.18149	0.00019	0.88826	0.00029	17.524	0.066	15.565	0.058	38.229	0.147
4							Inclusion signal	2.18008	0.00015	0.88741	0.00024	17.523	0.058	15.552	0.054	38.206	0.122
5	AKF	many and big inclusions	60	30.5	<0.004	0.084 <sup>‡</sup>	inclusions	2.16330	0.00024	0.87984	0.00010	17.711	0.016	15.578	0.013	38.309	0.034
5							inclusions	2.16184	0.00023	0.87904	0.00009	17.765	0.013	15.618	0.012	38.410	0.032
6	AKF	many and big inclusions	60	51.7	<0.004	0.012	inclusions	2.16734	0.00026	0.88173	0.00012	17.680	0.019	15.591	0.017	38.312	0.041
6							Inclusion signal	2.17262	0.00018	0.88421	0.00008	17.617	0.130	15.579	0.110	38.279	0.280
<b>D550-594 (Mine grids: -5070.40 E +545.080 N): Quartz Latite Porphyry</b>																	
1	Plag An23	clean, next to 120 µm pit	60	35.2	<0.005	<0.005	clean	2.15896	0.00010	0.87783	0.00007	17.759	0.011	15.589	0.010	38.345	0.025
2	Plag						clean	2.16039	0.00008	0.87861	0.00006	17.739	0.010	15.586	0.009	38.326	0.023
3	AKF	full of inclusions, next to pit	90	79.9	0.094	0.023	full of inclusions	2.16090	0.00010	0.87868	0.00004	17.751	0.004	15.600	0.004	38.364	0.010
3	AKF	several inclusions	44	68.8	<0.012	<0.006											
4	Plag An24	clean, next to 120 µm pit	90	33.3	0.005	<0.002	clean	2.15741	0.00009	0.87729	0.00006	17.746	0.009	15.570	0.008	38.302	0.021
5	Plag						clean	2.15483	0.00009	0.87647	0.00009	17.798	0.016	15.598	0.014	38.355	0.035
6	Plag An22	clean, next to 80 µm pit	44	34.5	<0.007	<0.004	clean	2.15696	0.00010	0.87695	0.00011	17.741	0.020	15.560	0.017	38.262	0.044
<b>4890-2080 (surface sample, near Mine grids: -134 E, -1353 N): Quartz Latite Porphyry</b>																	
1	AKF	clean	120	53.1	<0.002	0.002	clean	2.18437	0.00009	0.88946	0.00005	17.495	0.008	15.561	0.007	38.213	0.017
2	AKF	clean	120	54.7	<0.002	0.002	clean	2.18399	0.00011	0.88930	0.00006	17.493	0.008	15.556	0.007	38.205	0.018
3	AKF	clean					clean	2.18426	0.00010	0.88940	0.00005	17.512	0.008	15.578	0.007	38.254	0.017
4	AKF	clean					clean	2.18417	0.00010	0.88944	0.00005	17.500	0.008	15.566	0.007	38.226	0.017
5		clean					clean	2.18497	0.00012	0.88967	0.00005	17.506	0.007	15.574	0.006	38.255	0.016
6		clean					clean	2.18437	0.00012	0.88930	0.00006	17.518	0.009	15.581	0.008	38.269	0.019
7		clean					clean	2.18440	0.00011	0.88933	0.00005	17.498	0.007	15.564	0.006	38.228	0.015
8		clean	120	58.2	<0.002	0.000	clean	2.18409	0.00018	0.88927	0.00007	17.489	0.011	15.554	0.009	38.217	0.024
9		clean	90	57.1	<0.004	<0.002	clean	2.18477	0.00012	0.88958	0.00005	17.491	0.008	15.557	0.007	38.213	0.017
<b>5090-980 (surface sample, near Mine grids: -134 E, -1353 N): Clast of Biotite Porphyry</b>																	
1	AKF	clean, near pit	60	48.7	<0.004	<0.008	clean	2.17386	0.00011	0.88441	0.00008	17.585	0.014	15.552	0.013	38.228	0.032
2	AKF	clean, near pit	60	38.2	<0.006	<0.004	a few inclusions	2.17786	0.00013	0.88636	0.00010	17.584	0.018	15.582	0.016	38.303	0.040
3	AKF	clean, near pit	60	42.2	<0.008	<0.004	clean	2.18074	0.00012	0.88757	0.00016	17.501	0.034	15.529	0.031	38.165	0.077
3	AKF						high-Pb peak	2.18137	0.00022	0.88854	0.00051	17.483	0.090	15.542	0.079	38.139	0.197
3	AKF						high-Pb peak	2.18159	0.00021	0.88886	0.00038	17.540	0.075	15.592	0.070	38.270	0.125
4	Plag An23	clean, near pit	90	23.2	<0.002	<0.002											
5	Plag An23	clean, near pit	90	23.2	<0.002	<0.002											
<b>Bulk rock †</b>																	
	Melanepherinite	TICK 103, fresh						2.13029	0.00003	0.85502	0.00002	18.291	0.001	15.640	0.001	38.965	0.003
	Melanepherinite	TICK 120, fresh						2.13035	0.00005	0.85505	0.00001	18.287	0.001	15.636	0.001	38.957	0.002
	Shoshonite	TICK 116, fresh						2.18642	0.00003	0.87879	0.00001	17.827	0.001	15.666	0.001	38.978	0.002
	Shoshonite	TICK 117, fresh						2.19637	0.00002	0.88229	0.00001	17.748	0.000	15.659	0.000	38.981	0.001
	Equigran. Monz.	WR 8a, propyl. altered						2.16574	0.00003	0.88076	0.00001	17.697	0.000	15.587	0.000	38.328	0.001

**Notes:** For sample identifiers beginning with a letter followed by XXX-YYY, XXX is the drill core number and YYY is the depth (in ft) of core (Mine Grids are given in brackets)

For sample identifiers of the form XXXX-YYY, XXXX is the bench elevation and YYY is the distance along the bench (Mine Grids are given in brackets)

Lithologies are field determinations (Landtwing, 2004)

‡ Elevated U content due to ablation of inclusions

clean Indicates no visible inclusions

<value Uncertainties are reported as the one standard error of the mean of measurement (1 s.e.m.)

† Element concentration below the limit of detection (3σ criterion)

† Coordinates for bulk rocks are not available, except Mine Grids for P 8a (-1150 E / -4850 N)