1 Constraining the timing and sources of volcanism at the Apollo 12 landing site using new Pb isotopic
2 compositions and crystallisation ages

3 **Authors:** Joshua F. Snape^{1*}, Bart Davids², Alexander A. Nemchin³, Martin J. Whitehouse¹, Jeremy J.
4 Bellucci¹

5 **Affiliations:**

6 ¹ Department of Geosciences, Swedish Museum of Natural History, SE-104 05 Stockholm, Sweden.

7 ² Department of Geological Sciences, Stockholm University, SE-106 91 Stockholm, Sweden.

8 ³ Department of Applied Geology, Curtin University, Perth, WA 6845, Australia.

9 *Corresponding author (joshua.snape@nrm.se)

10 **Abstract**

11 The basaltic suites collected at the Apollo 12 landing site have been interpreted as representing a
12 stratigraphic sequence of volcanic flows emplaced in the Oceanus Procellarum region between
13 approximately 3100-3300 Ma. This study presents Secondary Ion Mass Spectrometry (SIMS) Pb
14 isotopic analyses of samples from each of the basaltic suites, which have been used to constrain
15 precise crystallisation ages and initial Pb isotopic compositions. The new crystallisation ages are
16 consistent with the three main basaltic suites (olivine, pigeonite and ilmenite) being emplaced over a
17 period of approximately 60 million years, and the improved precision of these ages has made it
18 possible to reinterpret the stratigraphic sequence of basalt flows underlying the Apollo 12 landing site.
19 Contrary to previous studies, the three ilmenite basalts are determined as having the oldest ages (with
20 a weighted average of 3187±6 Ma; 2σ) and are, therefore, interpreted as representing the lowest unit in
21 the sequence, underlying the olivine and pigeonite basalts (with an age range constrained by the oldest
22 and youngest pigeonite basalts; 3176±6 Ma and 3129±10 Ma; 2σ). The initial Pb isotopic
23 compositions have been compared with recalculated initial Sr and Nd isotopic compositions, and are
24 consistent with the three main basaltic suites originating from magmatic sources that incorporated
25 different proportions of a common primitive mafic cumulate and the residual trapped liquid fraction

remaining after a majority of the lunar magma ocean had crystallised. Our data also demonstrate that the feldspathic basalt (12038) is unique, both in terms of its crystallisation age (3242 ± 13 Ma) and its derivation from a distinct mantle reservoir.

1. Introduction

The Apollo 12 mission provided 34.3 kg of mainly basaltic samples (43 of the 47 rocks collected) documenting volcanic activity in Oceanus Procellarum between approximately 3100-3200 Ma. All of the basalts returned by the mission are low-Ti mare basalts ($\text{TiO}_2 = 1\text{-}6$ wt%; following the scheme of Neal and Taylor 1992). These have been subdivided into three main basaltic suites: olivine, pigeonite and ilmenite basalts (e.g. Neal et al. 1994a). A fourth, feldspathic, suite has been proposed, which is represented by only one rock sample (12038), although several grains from Apollo 12 soil samples have been suggested as additional members (Korotev et al. 2011; Alexander et al. 2016). Crystallisation ages previously determined for the Apollo 12 basalts have not been sufficiently precise to constrain the ages of the basaltic suites relative to each other, given the narrow time interval (20-50 million years) over which the three main suites appear to have been emplaced. Based on the data available at that time, Snyder et al. (1997) calculated a series of weighted average ages demonstrating the similarity for all but the single anomalous feldspathic basalt (pigeonite = 3180 Ma; olivine = 3190 Ma; ilmenite = 3200 Ma; feldspathic = 3280 Ma).

Modelling of the Apollo 12 basalt sources initially relied on the sample mineralogies and major- and trace-element characteristics (Warner 1971; Rhodes et al. 1977). This work led to the hypothesis that the olivine and pigeonite basalts were co-magmatic, with the olivine basalts representing more primitive cumulates, while the pigeonite basalts were considered to be formed from more evolved parental magma compositions (Rhodes et al. 1977). Subsequent studies have also attempted to model the sources of the basalts using Sr and Nd isotopic data in addition to incompatible trace-element (ITE) chemistry (Nyquist et al. 1977; 1979; 1981; Neal et al. 1994a,b; Snyder et al. 1997). Neal et al. (1994a) examined the compositions for fine-grained and vitrophyric Apollo 12 basalts thought to be most representative of the parental magmas, focusing on particular ITE ratios (e.g. Co/Hf, Sc/Sm,

Rb/Sr and Sm/Eu) that would be indicative of fractionation or accumulation, as well as the presence of pyroxene and plagioclase in the mantle sources. The results of this work indicate that the olivine and pigeonite basalt suites are unrelated and were derived from distinct mantle sources.

Based on the Mg# (atomic Mg/[Fe+Mg]) and the systematics of Li vs. Rare Earth Elements (REE) in similarly fine-grained and vitrophyric Apollo 12 basalts, Snyder et al. (1997) concluded that the three main basaltic suites were derived from partial melts of cumulates formed after 82-94% crystallisation of the Lunar Magma Ocean (LMO). Furthermore, the significantly higher initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratios determined for the ilmenite basalts, when compared with those of the olivine and pigeonite basalts, were interpreted as definitive evidence of a distinct source for at least the ilmenite basalts. Combining the Sr and Nd isotopic data for the basalts, Snyder et al. (1997) presented a model in which the parent melts of the three main basaltic suites were formed by varying degrees of partial melting (5-6% for the ilmenite basalts and 7-9% for the olivine and pigeonite basalts) of Lunar Magma Ocean (LMO) cumulates that had incorporated different amounts of ITE-rich LMO residual liquid. In the case of the ilmenite basalts, the relatively high initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratios (initial $\epsilon^{143}\text{Nd} = 10.5-11.3$) and low initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.69920-0.69932) indicate that the ilmenite source was nearly devoid of this residual liquid (<0.15%), while the olivine and pigeonite sources (initial $\epsilon^{143}\text{Nd} = 4.3-4.7$; initial $^{87}\text{Sr}/^{86}\text{Sr} = 0.69954-0.69960$) contained more residual liquid (0.3-0.5%). Subsequent work by Hallis et al. (2014) recreated REE abundances measured in olivine, pigeonite and ilmenite basalts starting from a model composition of a low-Ti source with 1% LMO residual liquid and smaller amounts of partial melting (1-2% for the ilmenite basalts and 1-3% for the olivine and pigeonite basalts). The differing conclusions of Snyder et al. (1997) and Hallis et al. (2014) are a reflection of the different modelling approaches and assumptions, but both studies demonstrate the strong effect of very small amounts of LMO residual liquid on the ITE compositions of the basalt sources.

Recent work by Snape et al. (2016) has demonstrated the potential of Secondary Ion Mass Spectrometry (SIMS) Pb isotopic measurements to provide high precision dates for lunar basalts, interpreted to represent igneous crystallisation. The strength of this approach lies in its ability to analyse rapidly a range of phases, providing measurements of both radiogenic Pb isotopic

compositions formed through the radioactive decay of ^{235}U and ^{238}U in U-rich minerals since crystallisation of a basalt sample and hence indicating the crystallisation age of the sample, as well as compositions closer to the initial Pb component inherited by the basalt from its source. The aim of this study is to use this approach in an attempt to resolve the remaining ambiguity regarding the chronology of volcanic activity at the Apollo 12 landing site and provide new insights to the evolution of the mantle sources from which the Apollo 12 basalts originated.

2. Methods

2.1. Sample descriptions

In addition to the three Apollo 12 basalts investigated previously (feldspathic basalt 12038, pigeonite basalt 12039 and ilmenite basalt 12063; Snape et al. 2016), a further five samples have been analysed in the present study. These samples include two ilmenite basalts (12005 and 12016), two olivine basalts (12002 and 12012) and a pigeonite basalt (12021).

The two olivine basalts analysed in this study (12002,600 and 12012,34; Figs. 1a-b and A.1-A.2) have similar porphyritic-subophitic textures, characterised by olivine (13-23% by modal abundance) and pyroxene (50-56%) phenocrysts (typically 200-600 μm in size) surrounded by subophitic groundmasses. Both mafic phases are prominently zoned from more Mg-rich cores to more Fe-rich rims, with the cores of the pyroxene crystals also exhibiting more complex zoning between pigeonite and augite compositions. The subophitic groundmass surrounding the mafic phenocrysts comprises pyroxene intergrown with plagioclase crystals (typically 50-400 μm ; 23-27% by modal abundance; Table B.1), which are generally elongated, occasionally with hollow intrafasciculate morphologies, and in the case of 12002 are sometimes clustered to form acicular bundles. Ilmenite laths (up to approximately 150 μm long; 2% by modal abundance) and spinel crystals (50-100 μm ; 1%) are common throughout the groundmass of the two samples, with the spinels commonly exhibiting well defined chromite cores and ulvöspinel rims. Also present are grains of a silica polymorph (most likely cristobalite; 10-50 μm ; 1%; Table B.1), which tend to occur within or adjacent to the plagioclase crystals. Mesostasis areas, approximately 50 μm in size, are commonly found at the boundaries

105 between plagioclase and the most Fe-rich pyroxene. These typically contain K-rich glass, sulfide
106 grains and Ca-phosphate, often as fine-grained (1-5 μm) intergrowths.

107 The pigeonite basalt (12021,619; Figs. 1c and A.3) contains several very large (1-4 mm) pyroxene
108 phenocrysts in a variolitic groundmass primarily composed of pyroxene (total pyroxene modal
109 abundance of 64%), plagioclase (28%), ilmenite (4%) and grains of silica (4%; Table B.1). The silica
110 grains form either fine laths or less elongated subhedral-anhedral crystals (previous studies indicate
111 that the former is likely tridymite while the latter is cristobalite; Weill et al. 1971). Less common
112 phases in the groundmass include occasional grains of both chromite and ulvöspinel (<1%; Table B.1).
113 Relatively large (up to approximately 200 μm) symplectite assemblages and smaller (50-100 μm)
114 mesostasis areas typically occur at the boundaries between the Fe-rich pyroxene grains and the other
115 groundmass phases. The mesostasis includes K-rich glass, K-feldspar and Ca-phosphate crystals.

116 The ilmenite basalt 12005 has been previously identified as being an unique lunar basalt, due to its
117 high Mg# (62) and olivine content (30% by modal abundance), as well as its “cumulate texture”
118 (Rhodes et al. 1977). As is typical of the sample, the section analysed in this study (12005,78; Figs. 1d
119 and A.4.) contains rounded olivine grains (typically 50-300 μm in size; 37% by modal abundance;
120 Table B.1) poikilitically enclosed by pyroxene (typically 100-500 μm ; 40%), plagioclase (typically
121 50-200 μm ; 19%) and (less commonly) ilmenite (typically 100-200 μm ; 3%), while euhedral to
122 subhedral grains of ulvöspinel and chromite (typically 10-50 μm) are common throughout the sample
123 (1%), occasionally forming glomerophyric clusters, with the ulvöspinel often exhibiting ilmenite
124 exsolution. The EDS element map of the sample (Fig. A.4.) indicates that the major phases in the
125 section are generally equilibrated and homogenous in major element composition. Grains of FeNi-
126 metal and troilite (between 1-20 μm ; <1% by modal abundance; Table B.1) are also present within the
127 pyroxene and plagioclase grains, while several mesostasis areas (approximately 50 μm) of additional
128 accessory phases are present interstitially between the major phases. The accessory phases in the
129 mesostasis (typically between 1-10 μm) include Ca-phosphates, K-rich glass, K-feldspar, sulfides and
130 ilmenite grains.

Ilmenite basalt 12016 was described by Dungan and Brown (1977) as an equigranular, medium-grained microgabbro. The section analysed in this study (12016,39; Figs. 1e and A.5.) contains pyroxene (54 % by modal abundance; Table B.1), plagioclase (25 %), olivine (15 %) and ilmenite (4 %). As has been previously reported (Dungan and Brown 1977), the pyroxene grains (up to approximately 600 μm in size) are characterised by complex intergrowth of augite and pigeonite. The olivine grains (typically 50-500 μm) exhibit some zonation from more Mg-rich cores to Fe-rich rims, and typically have embayed grain boundaries with the pyroxene. Anhedral plagioclase grains (typically 50-400 μm) occur interstitially to the olivine and pyroxene and occasionally subpoikilitically enclosing the mafic phases. After ilmenite, spinel (both chromite and ulvöspinel) is the next most abundant phase (~1 %; Table B.1). Mesostasis areas (between approximately 20-100 μm in size) occasionally occur in interstitial sites between the Fe-rich rims of the pyroxene and typically contain 1-10 μm sized grains K-rich glass, Ca-phosphates and sulphides.

2.2. Analytical protocol

The ilmenite basalt sample, 12016,39, is a thin section, while the remaining four samples are rock chips embedded in epoxy blocks. All five samples were prepared at NASA Johnson Space Center. Each sample was cleaned with ethanol before being carbon coated. Backscattered electron (BSE) images and elemental maps of each section were acquired using a Quanta 650 FEGSEM and accompanying Oxford Instruments Energy Dispersive Spectroscopy (EDS) detector at Stockholm University, operating with an accelerating voltage of 20 kV at a working distance of 10 mm.

Following the SEM documentation of the samples and prior to the SIMS analyses, the samples were cleaned with a fine (1 μm) diamond paste and ethanol to remove the carbon coating before adding a 30 nm gold coating. The Pb isotopic compositions of the phases were determined over two analytical sessions using a CAMECA IMS 1280 ion microprobe at the NordSIMS facility, Swedish Museum of Natural History, Stockholm, using a methodology similar to that outlined in previous studies (Whitehouse et al. 2005; Bellucci et al. 2015). Apertures in the primary column were used to generate a slightly elliptical O_2^- sample probe with dimensions appropriate to the target. The smaller accessory

phases (including K-feldspar and phosphates) were analysed using a $\sim 10\ \mu\text{m}$ spot (beam current 2-3 nA), whereas larger grains (including plagioclase and K-rich glass) were analysed with a $\sim 30\ \mu\text{m}$ spot (beam current 14-16 nA). Prior to each measurement, an area of 20-35 μm around the spot location was rastered for 120 seconds in order to remove the gold coating and minimise possible surface contamination. The instrument was operated in high-transmission mode, corresponding to a transfer magnification of $160\times$. In this mode, the field aperture size was chosen to limit the field of view on the sample surface (i.e. the area from which ions will be admitted to the mass spectrometer) to be bigger than the unrastered spot but smaller than the rastered beam, further minimising the possibility of surface contamination. The mass spectrometer was operated with a nominal mass resolution of 4860 ($M/\Delta M$), sufficient to resolve Pb from known molecular interferences. A nuclear magnetic resonance (NMR) field sensor regulated the stability of the magnetic field to high precision. The Pb isotopes were measured simultaneously in multi-collector mode using four low-noise ion counting electron multipliers (Hamamatsu 416) with electronically-gated deadtimes of 65 ns. Background counts for each channel were measured at regular intervals during each session. The average background counts were used to correct the sample analyses in instances where statistically significant background noise levels were detected (i.e. where the standard deviation of the background measurements for a given session was lower than the average background values for that session). The average background values are reported in Table B.3. Individual analyses were filtered out of the final dataset if the count rates for any masses were lower than $3\times$ the background count rates during that session.

Analyses of the USGS basaltic glass reference material, BCR-2G, were used to generate a correction factor to account for mass fractionation and detector relative gain calibration in the unknown analyses, assuming the values of Woodhead and Hergt (2000). The reproducibility of the BCR-2G analyses for the two sessions were as follows: $^{208}\text{Pb}/^{206}\text{Pb} = 0.36\%$ and 0.57% ; $^{207}\text{Pb}/^{206}\text{Pb} = 0.33\%$ and 0.47% ; $^{208}\text{Pb}/^{204}\text{Pb} = 0.95\%$ and 0.95% ; $^{207}\text{Pb}/^{204}\text{Pb} = 0.68\%$ and 0.82% ; $^{206}\text{Pb}/^{204}\text{Pb} = 0.72\%$ and 1.06% (reported as 2σ standard deviations from the average values for each session).

2.3. Data processing

The data were processed using in-house SIMS data reduction spreadsheets and the Excel add-in Isoplot (version 4.15; Ludwig 2008), using the same approach as that outlined in Snape et al. (2016). Briefly, the Pb isotopic compositions measured in each sample (the complete dataset is presented Table B.2.) are interpreted as representing a mixture between three main components: (1) initial Pb present in the basaltic melt when it crystallised, (2) radiogenic Pb formed by the decay of U in the basalt after crystallisation, and (3) terrestrial contamination. These endmember components define a triangular array of points on a plot of $^{207}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$. Given the very radiogenic Pb isotopic compositions associated with the Moon, relative to those found on Earth, the terrestrial contamination end-member will have higher $^{204}\text{Pb}/^{206}\text{Pb}$ ratios. The bounding edge on the left side of the triangle, between the initial and radiogenic lunar Pb compositions, forms an isochron defining the time when the basalt crystallised (Fig. 2). The values lying on this isochron with the highest $^{207}\text{Pb}/^{206}\text{Pb}$ ratios will be the closest estimates to the initial Pb composition of the sample.

The uncertainties stated for each ratio in the individual measurements (Table B.2) are derived from the internal run error propagated together with the standard deviations of the BCR-2G analyses for the relevant analytical session and the uncertainty given for the published BCR-2G values (Woodhead and Hergt 2000). The errors stated for the Pb-Pb isochron dates in the following results and discussion sections are quoted at the 95% confidence level. Uncertainties associated with the calculated $^{238}\text{U}/^{204}\text{Pb}$ (μ -values), $^{87}\text{Rb}/^{86}\text{Sr}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios were determined using a Monte Carlo approach, by calculating standard deviations from repeat runs of the calculations (see Appendix C for further details).

3. Results

The two ilmenite basalts, 12005 and 12016, yield Pb-Pb isochron dates of 3182 ± 10 Ma and 3186 ± 11 Ma, respectively (Table 1; Fig. 2). A similar date of 3176 ± 6 Ma was determined for the 12021 pigeonite basalt (Table 1; Fig. 2). The olivine basalts, 12002 and 12012, yield Pb-Pb isochrons dates of 3167 ± 12 Ma and 3151 ± 21 Ma respectively (Table 1; Fig. 2).

Three K-feldspar analyses in the 12005 ilmenite basalt yield similar Pb isotopic compositions with the highest $^{207}\text{Pb}/^{206}\text{Pb}$ ratios and, as such, a weighted average of these points is taken as the best estimate of the initial Pb isotopic composition of the basalt (Table 1; Fig. 2). Similarly, two K-feldspar analyses in the 12021 pigeonite basalt are interpreted as providing the best estimate of the initial Pb isotopic composition in that sample (Table 1; Fig. 2). For the remaining three basalts the highest $^{207}\text{Pb}/^{206}\text{Pb}$ ratios are only represented by single analyses, which are taken as being the closest available determinations of the initial Pb isotopic compositions (Table 1; Fig. 2).

Plots of the $^{208}\text{Pb}/^{206}\text{Pb}$ ratios against $^{204}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ for the filtered datasets from each of the samples lie on a plane in the 3D coordinate space, with the initial Pb compositions converging at similar $^{208}\text{Pb}/^{206}\text{Pb}$ ratios (~ 0.9 - 1.0), while the measurements of phases containing more Pb from *in situ* radiogenic decay spread out, such that the purely radiogenic endmember compositions would be between $^{208}\text{Pb}/^{206}\text{Pb} \sim 0.2$ - 4.2 (Fig. 3). This range in radiogenic $^{208}\text{Pb}/^{206}\text{Pb}$ ratios is interpreted as variability in $^{232}\text{Th}/^{238}\text{U}$ ratios between different mineral phases within the samples. Taking the crystallisation ages of the samples into account, these $^{208}\text{Pb}/^{206}\text{Pb}$ values would correspond to $^{232}\text{Th}/^{238}\text{U}$ ratios of between ~ 0.0 - 4.0 , with the exception of two phosphate grains analysed in the 12016, which much higher $^{208}\text{Pb}/^{206}\text{Pb}$ ratios, equating to radiogenic $^{208}\text{Pb}/^{206}\text{Pb}$ ratios of up to 10.6 , and $^{232}\text{Th}/^{238}\text{U}$ ratios of 10.3 .

4. Discussion

4.1. Timing of volcanism at the Apollo 12 landing site

Given their pristine basaltic nature of the Apollo 12 basalts the Pb-Pb isochron dates in this study are interpreted as representing igneous crystallisation ages. When combined with the pigeonite and ilmenite basalts analysed previously (12039 and 12063; Snape et al. 2016), these ages define a period of volcanic activity at the Apollo 12 landing site lasting about 60 million years, while the feldspathic basalt 12038 is significantly older (3242 ± 13 Ma; Snape et al. 2016) than the other three basaltic suites (Nyquist et al. 1981; Snyder et al. 1997). This is consistent with the weighted average ages of the three main basalt suites reported by Snyder et al. (1997). Figure 4 provides more a detailed comparison

between the ages reported in this study and previous age determinations for the same samples (n.b. the ages from previous studies have been recalculated using Isoplot to account for updated decay constants; a summary of these recalculated ages is also presented in Table B.4). These new ages are broadly in agreement with these previous studies, although the ages for 12002 (olivine basalt) and 12021 (pigeonite) appear to be younger than two previous Rb-Sr ages (Papanastassiou and Wasserburg 1970; 1971). For 12021, an additional Rb-Sr age was reported by Cliff et al. (1971), which is within error of the new age determined from the Pb-Pb isochron. Two ^{40}Ar - ^{39}Ar ages have been reported for 12002 by Turner (1971) and Alexander et al. (1972), which have been recalculated to 3226 ± 49 Ma and 3294 ± 78 Ma, respectively. Updated ages were not available for the neutron flux monitor used during irradiation of the samples in order to calculate a J-value, so these recalculated values are based on the original monitor ages. Despite the age presented by Alexander et al. (1972) being slightly older than that inferred from the Pb-Pb isochron, it is noted that these data do not yield a plateau on the standard ^{40}Ar - ^{39}Ar age spectrum plot, as there are no adjacent temperature steps within 2 standard deviations of each other, and the highest apparent age is yielded from a single temperature step comprising less than 50% of the total released Ar. Instead, the ^{40}Ar - ^{39}Ar age presented by Alexander et al. (1972) is obtained by plotting the values from the individual heating steps on a plot of $^{37}\text{Ar}/^{39}\text{Ar}$ vs. $^{40}\text{Ar}/^{39}\text{Ar}$, and obtaining a cubic-least-squares fit for the steps $>800^\circ\text{C}$ (in this case constituting only a 3-point fit), where the intercept of this line with the $^{40}\text{Ar}/^{39}\text{Ar}$ axis was interpreted as the potassium-derived argon in the sample and used to determine a crystallisation age. For this reason, the age determined by Turner (1971) is considered to be more reliable.

The most detailed evaluation for the stratigraphic sequence of the basalt flows underlying the landing site was previously inferred on the basis of sample distribution at the landing site relative to the craters that likely excavated them, while the thicknesses of the flows were determined from the petrologic textures and inferred cooling rates of the basalts (Rhodes et al. 1977 and references therein). In this interpretation an ~ 80 m stratigraphic sequence is described, with the olivine basalts originating from the greatest depths and, therefore, representing the oldest of the three suites, due their ubiquity on the rim of the largest crater in the area (Middle Crescent). The ilmenite basaltic suite was interpreted as

representing the youngest flow, with the potential co-magmatic nature of the olivine and pigeonite suites suggesting that the pigeonite basalt flow was most likely located between the olivine and ilmenite basalt flows.

Contrary to that stratigraphic sequence, the three ilmenite basalts dated by the Pb-Pb isochron approach (this study and Snape et al. 2016) have the oldest ages of the three main basaltic suites, defining a particularly narrow window during which the basalts were emplaced, from 3182 ± 10 Ma to 3193 ± 11 Ma, with a weighted average of the three ages at 3187 ± 6 Ma (2σ). The ages of the two olivine basalts (3167 ± 12 Ma and 3151 ± 21 Ma) are bracketed by the two pigeonite basalt samples (3176 ± 6 Ma and 3129 ± 10 Ma). This indicates that the source of the pigeonite basalts was active and resulted in multiple eruptions over a more protracted period (30-60 Ma) than the ilmenite basalts. The overlap in the pigeonite and olivine basalt crystallisation ages could be explained by previous suggestions that the two suites were co-magmatic (Rhodes et al. 1977; Snyder et al. 1997). Alternatively, if the basalts represent two distinct mantle sources (Neal et al. 1994a; 1994b), then the ages imply either distinct periods of pigeonite basalt volcanism before and after the olivine basalts formed, or that both sources were active at the same time resulting in a more complicated interfingering stratigraphic relationship of the lava flows (Fig. 5). Finally, the older age of 12038 indicates either that the feldspathic basalt represents a poorly sampled older unit underlying the ilmenite basalts, or that it was introduced to the landing site as impact ejecta from a different location (Neal et al. 1994a).

4.2. Pb isotope systematics of Apollo 12 basalts

The initial Pb isotopic compositions of the Apollo 12 basalts form two groups when plotted on a diagram of $^{207}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$ (Fig. 6a). The feldspathic basalt, ilmenite basalts and one pigeonite basalt (12039) are characterised by higher $^{207}\text{Pb}/^{206}\text{Pb}$ ratios (and generally slightly higher $^{204}\text{Pb}/^{206}\text{Pb}$ ratios), while the two olivine basalts and remaining pigeonite basalt (12021) have lower $^{207}\text{Pb}/^{206}\text{Pb}$ ratios. This variability between the Apollo 12 basalts can be further investigated by combining the ages and initial Pb isotopic compositions with the multiple stage Pb isotopic evolution

model described by Snape et al. (2016). The model implies a major differentiation event at 4376 ± 18 Ma (consistent with model ages derived from the Sm-Nd and Lu-Hf isotopic systems: e.g. Borg et al. 2011; Carlson et al. 2014; McLeod et al. 2014) most likely linked to the crystallisation of the LMO, resulting in the separation of an isotopically homogenous lunar mantle into distinct silicate reservoirs, which formed the sources of the chemically diverse basaltic rocks. The model also provides an estimate of the Pb isotopic composition for the homogenous lunar mantle prior to differentiation, which, when combined with the time of differentiation, allows calculation of model source μ -values for the analysed basalt samples. These sources are broadly divided into two main categories: (1) high- μ (~ 2600 - 3700) reservoirs for the KREEP-rich basalts, and (2) a relatively low- μ (370 - 640) group of high-Ti and low-Ti mare basalt reservoirs (Fig. 6b). Despite the variation in $^{207}\text{Pb}/^{206}\text{Pb}$ ratios between the Apollo 12 initial Pb isotopic compositions, all 8 of the basalts are approximately positioned along the trajectory of the low-Ti basalt growth curve.

Provided that the initial Pb isotopic compositions are accurate and that the model is a suitable approximation for the evolution of the basalt sources, paleo-isochrons constructed from the model composition at the time of differentiation and defined by the range of basalt crystallisation ages should intersect the compositions of the Apollo 12 basalts (Fig. 6b). This is the case for some of the samples, but the feldspathic basalt, olivine basalts and the 12021 pigeonite basalt have significantly lower $^{207}\text{Pb}/^{206}\text{Pb}$ ratios than would be predicted by the model. This either suggests that the initial Pb isotopic compositions of these four samples are underestimates of the true values, or indicates a problem in the Pb isotopic evolution model, making it unsuitable to explain the origin of the Apollo 12 basalt sources. The most confident determinations of initial Pb, both in this dataset and that presented previously (Snape et al. 2016), originate from samples where these compositions are defined by multiple analyses, particularly in K-feldspar and plagioclase phases, where there are lower concentrations of U (i.e. consistent with the Pb representing the initial component rather than that derived from radiogenic decay of U after the basalt crystallised) than there are in many of the analysed K-rich glass areas (Snape et al. 2016). As such, the Pb isotopic model was originally constructed using the values from three samples (10044, 12039 and 14072) with the most confidently defined initial Pb isotopic

compositions (Snape et al. 2016). By comparison, in this new dataset, it was only possible to place a lower estimate on the initial Pb isotopic composition for the majority of the samples (the three K-feldspar analyses in 12005 being an exception) using one or two analyses. In the case of the two olivine basalts (12002 and 12012), the initial Pb isotopic compositions are defined by single measurements K-rich glass, which commonly yields more radiogenic Pb isotopic compositions.

The lower initial $^{207}\text{Pb}/^{206}\text{Pb}$ values for some of the samples can potentially be explained by having a relatively low μ -value (~ 50 -100) for the homogenous lunar mantle (compared with ~ 460 in the original model), with the sources of the basalts originating from a younger differentiation event at ~ 4300 Ma (Fig. 6c), but the higher initial $^{207}\text{Pb}/^{206}\text{Pb}$ values in the remaining samples cannot be explained with this model, as they would lie above the paleo-isochrons corresponding to their respective crystallisation ages. An alternative way to explain the compositions for all of the Apollo 12 basalts is to have them originating from sources that separated from the homogenous lunar mantle much later (~ 4100 Ma; Fig. 6d). This would then require that the 12021 pigeonite basalt had a μ -value >750 (compared to that of about 640 for the other pigeonite basalt, 12039), while the olivine basalt compositions could only be explained with μ -values >1000 , which are significantly higher than those proposed for any other mare basalt. More importantly, these alternative model ages for the differentiation event would no longer be in agreement with the Sm-Nd and Lu-Hf isotopic model ages, and would not be consistent with even very slow (~ 220 Ma) LMO crystallisation described in recent models for early lunar evolution (Meyer et al. 2010; Elkins-Tanton et al. 2011). This discrepancy between the compositions of the two pigeonite basalts, and the difficulty in correlating these alternative model ages with other models of LMO crystallisation, indicate that an underestimation of initial Pb isotopic compositions in the two olivine basalts, the 12021 pigeonite basalt and the 12038 feldspathic basalt is the most likely explanation for the range of compositions observed between the basalts.

Taking the model from Snape et al. (2016), the degree to which the initial Pb isotopic compositions have been underestimated can be predicted by calculating the intercept between the paleo-isochrons from the model differentiation point and the primary sample isochrons (Figs. 2 and 6a; Table 1).

Combining either these predicted initial compositions or the measured initial compositions with the Snape et al. (2016) model values for the time of the magmatic differentiation and composition of the homogenous lunar mantle, it is then possible to determine model μ -values of the basalt sources (Fig. 7a; Table 1). These source μ -values are particularly useful, as they provide a time-independent value for characterising the basaltic suites. Furthermore, the μ -values should be accurate provided the $^{204}\text{Pb}/^{206}\text{Pb}$ initial ratios of the basalts are not significantly underestimated, which appears to be the case for all but the two olivine basalts (Figs. 2 and 7), where the predicted initial Pb isotopic compositions are outside of the uncertainties on the measured values. The lowest μ -values (440 ± 30) are estimated for the ilmenite basalts, which is consistent with smaller proportions of late stage LMO residual liquid/KREEP-rich material in the source, in agreement with the radiogenic Nd isotopic compositions of the ilmenite basalt sources, and the prediction from Snyder et al. (1997). The μ -value of the feldspathic basalt is slightly higher at either 531 ± 61 or 499 ± 58 , depending on whether the measured or predicted initial Pb isotopic composition of the sample is used. Based on the measured initial Pb isotopic compositions of the pigeonite and olivine basalts, the two suites appear to have distinct μ -values (640 ± 27 and 738 ± 24 , respectively; Fig. 7b), but if an underestimation of the initial $^{204}\text{Pb}/^{206}\text{Pb}$ ratios in the olivine basalts is factored in, the predicted μ -value for the olivine basalt suite is more similar to that of the pigeonite basalts (~ 650). Distinct μ -values for these two basaltic suites would suggest that they were the products of distinct magmatic sources as was indicated by Neal et al. (1994a; 1994b), and not co-magmatic as was previously proposed (Rhodes et al. 1977), but the uncertainty in the μ -values of the olivine basalts makes it difficult to confirm either scenario.

4.3. Evolution of the Apollo 12 basalt sources

Several models for the magmatic evolution of the Apollo 12 basaltic suites were considered to explain the range of observed initial Pb isotopic compositions and source μ -values, with the simplest being the evolution of a single magmatic reservoir. While there is some correlation between the initial Pb compositions and the crystallisation ages of the three main basaltic suites (excluding the feldspathic basalt) that might support such a hypothesis, this trend is broken by the youngest sample (pigeonite basalt 12039; Fig. 7b). More importantly, a single source model can be ruled out as it is not possible to

explain the range in compositions of the different basaltic suites by evolution of a single magmatic reservoir within the 60 million year period that the basalts were formed (Fig. 6a).

In order to further assess potential models for the evolution of the basalt sources, the crystallisation ages produced in this study have been used to recalculate the initial $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of the basalts, based on previous Rb-Sr and Sm-Nd analyses (Table B.3; Papanastassiou and Wasserburg 1970; Cliff et al. 1971; Nyquist et al. 1977; 1979; 1981; Unruh et al. 1984; Sprung et al. 2013; McLeod et al. 2014). As with the Pb isotope systematics and source μ -values, it is also possible to calculate time-independent values defining the source $^{87}\text{Rb}/^{86}\text{Sr}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios (Table B.4; details of these calculations are included in Appendix C). In the case of the olivine, pigeonite and ilmenite basalt suites, a positive correlation exists between $^{87}\text{Rb}/^{86}\text{Sr}$ ratios for the basalt sources and the source μ -values (Fig. 8a), whereas the source $^{147}\text{Sm}/^{144}\text{Nd}$ ratios and μ -values are negatively correlated (Fig. 8b). The feldspathic basalt 12038 falls away from both of these trends, particularly in the case of the $^{147}\text{Sm}/^{144}\text{Nd}$ ratios (Fig. 8b). Irrespective of any more detailed modelling, these correlations between the different isotopic systems provide additional confidence in the new crystallisation ages and Pb isotopic compositions, but also suggest that it might be possible to explain all three of the main basalt suites as originating from a common low-Ti LMO cumulate source, by adding different proportions of a high- μ LMO residual component. This corresponds with previous modelling of the Apollo 12 basalt sources, which indicated that they contained some proportion of LMO residual/KREEP-rich liquid (Snyder et al. 1997; Hallis et al. 2014).

It is not clear what the $^{87}\text{Rb}/^{86}\text{Sr}$, $^{147}\text{Sm}/^{144}\text{Nd}$ or μ -values were for the low-Ti cumulate source, but for this study, measurements of an Apollo 15 KREEP basalt (15386: Nyquist et al. 1975; McLeod et al. 2014; Snape et al. 2016), were considered to provide the best constrained estimates for the $^{87}\text{Rb}/^{86}\text{Sr}$, $^{147}\text{Sm}/^{144}\text{Nd}$ and μ -values in this KREEP-rich component. These 15386 source values form a distant end member composition on the high- μ side of the trends in Figure 8. Mixing between element and isotopic ratios has been demonstrated to form hyperbolic relationships (Vollmer 1976; Langmuir et al. 1978), described, in general, by the equation:

$$Ax + Bxy + Cy + D = 0 \quad [1]$$

where x represents the μ -values and y represent either the $^{87}\text{Rb}/^{86}\text{Sr}$ or $^{147}\text{Sm}/^{144}\text{Nd}$ ratios. Using the MATLAB nonlinear regression model fitting function it is possible to determine the coefficients in the specific equations defining such mixing trends between the Apollo 12 basalt and 15386 source values (for $^{87}\text{Rb}/^{86}\text{Sr}$ vs. μ : $A = 0.00118 \pm 0.00005$; $B = -0.0010 \pm 0.0009$; $C = -23 \pm 2$; $D = -0.12 \pm 0.06$; for $^{147}\text{Sm}/^{144}\text{Nd}$ vs. μ : $A = 0.00098 \pm 0.00004$; $B = -0.0070 \pm 0.0002$; $C = -1.9 \pm 0.2$; $D = 0.84 \pm 0.06$). Based on the source $^{87}\text{Rb}/^{86}\text{Sr}$, $^{147}\text{Sm}/^{144}\text{Nd}$ and μ -values for the Apollo 12 basalts and 15386, it is theoretically possible to begin placing constraints on the equivalent values for the low-Ti cumulate source, using the amounts of KREEP-rich material in the basalt sources and the proportions of partial melting suggested in previous models (Snyder et al. 1997; Hallis et al. 2014). However, the non-linear nature of the mixing relationship makes such extrapolations extremely sensitive to the uncertainties in these values. In addition to the uncertainties on the Apollo 12 basalt and 15386 source values, the final values determined for the low-Ti cumulate source are also sensitive to the large ranges in the amount of partial melting of the cumulate source in the models of Snyder et al. (5-6 % for the ilmenite basalts, 7-9 % for the olivine and pigeonite basalts) and Hallis et al. (1-2 % for the ilmenite basalts, 1-3 % for the olivine and pigeonite basalts), as well as amount the trapped late-stage LMO residual liquid in the sources (< 0.15 % for the ilmenite basalts, 0.3-0.5 % for the olivine and pigeonite basalts – Snyder et al. 1997; < 1 % for both sources – Hallis et al. 2014).

Unlike the other three basaltic suites, the feldspathic basalt source does not fit with the mixing hyperbola described above, implying that the basalt must have originated from a source that evolved along a distinct path. The source of the 12038 basalt is predicted to have an intermediate μ -value relative to those of the other Apollo 12 basalts, but lower $^{87}\text{Rb}/^{86}\text{Sr}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ ratios. In the case of Sr isotope systematics, this would place it further away from the KREEP source, while the Nd isotopic characteristics are closer to those of the KREEP source. As such, the feldspathic basalt source was not only distinct from the other Apollo 12 basalts, but it also cannot be explained by a similar LMO cumulate/KREEP-rich residual mixing model.

5. Conclusions

Precise Pb-Pb isochron crystallisation ages for the Apollo 12 basalts indicate that the three main basaltic suites were emplaced over a period of 60 million years from ~3190-3130 Ma, and confirm that the feldspathic basalt 12038 was formed significantly earlier (3242 ± 13 Ma). The ilmenite basalts appear to be the oldest of the three main basaltic suites, and were emplaced around 3187 ± 6 Ma (2σ). Crystallisation ages of the olivine and pigeonite basalts overlap, with the two analysed pigeonite basalts falling at either end of this range (3176 ± 6 Ma and 3129 ± 10 Ma). This indicates a more complicated eruptive history for these basalts, with the pigeonite basaltic suite either being emplaced in at least two distinct periods, or with a more protracted period of overlapping activity of the two suites.

The Pb isotopic compositions of the samples are consistent with all four basaltic suites representing separate parental melts, and indicate that the olivine and pigeonite basalts were not co-magmatic. Nonetheless, when combined with Sr and Nd data, these new Pb data are consistent with a model in which the three main basaltic suites were generated by mixing different proportions of a KREEP-rich late stage LMO component into a single low-Ti cumulate source. The isotopic characteristics of the feldspathic basalt, meanwhile, cannot be explained by such a mixing model and require a separate source to the low-Ti cumulate, which was isolated from mixing with the KREEP-rich component.

438 Acknowledgements

The authors thank the astronauts of Apollo 12 for collecting the samples. This work was funded by grants from the Knut and Alice Wallenberg Foundation (2012.0097) and the Swedish Research Council (VR 621-2012-4370) to MJW and AAN and (VR 2016-03371) to JJB. Additionally, BD acknowledges funding from the Department of Geological Sciences at Stockholm University. The NordSIMS facility is operated under a joint Nordic agreement; this is NordSIMS publication ***. The research has made use of NASA's Astrophysics Data System.

References

- Alexander E. C., Davis P. K. and Reynolds J. H. (1972) Rare-gas analyses on neutron irradiated Apollo 12 samples. Lunar Planet. Sci. III. Lunar Planet. Inst., Houston. pp. 1787-1795.
- Alexander L., Snape J. F., Joy K. H., Downes H. and Crawford I. A. (2016) An analysis of Apollo lunar soil samples 12070,889, 12030,187, and 12070,891: Basaltic diversity at the Apollo 12 landing site and implications for classification of small-sized lunar samples. Meteorit. Planet. Sci. 51, 1654-1677. <http://dx.doi.org/10.1111/maps.12689>.
- Bellucci J. J., Nemchin A. A., Whitehouse M. J., Humayun M., Hewins R. and Zanda B. (2015) Pb-isotopic evidence for an early, enriched crust on Mars. Earth Planet. Sci. Lett. 410, 34-41. <http://dx.doi.org/10.1016/j.epsl.2014.11.018>.
- Borg L. E., Connelly J. N., Boyet M. and Carlson R. W. (2011) Chronological evidence that the Moon is either young or did not have a global magma ocean. Nature 477, 70-72. <http://dx.doi.org/10.1038/nature10328>.
- Carlson R. W., Borg L. E., Gaffney A. M. and Boyet M. (2014) Rb-Sr, Sm-Nd and Lu-Hf isotope systematics of the lunar Mg-suite: the age of the lunar crust and its relation to the time of Moon formation. Phil. Trans. R. Soc. A. 372, 20130246. <http://dx.doi.org/10.1098/rsta.2013.0246>.
- Cliff R. A., Lee-Hu C. and Wetherill G. W. (1971) Rb-Sr and U, Th-Pb measurements on Apollo 12 material. Lunar Planet. Sci. II. Lunar Planet. Inst., Houston. pp. 1493-1502.
- Compston W., Berry H., Vernon M. J., Chappell B. W. and Kaye M. J. (1971) Rubidium-strontium chronology and chemistry of lunar material from the Ocean of Storms. Lunar Planet. Sci. II. Lunar Planet. Inst., Houston. pp. 1471-1485.
- Dungan M. A. and Brown R. W. (1977) The petrology of the Apollo 12 ilmenite basalt suite. Lunar Planet. Sci. VIII. Lunar Planet. Inst., Houston. pp. 1339-1381.

Elkins-Tanton, L. T., Burgess, S. & Yin, Q. -Z. The lunar magma ocean: Reconciling the solidification process with lunar petrology and geochronology. *Earth Planet. Sci. Lett.* 304, 326-336 (2011). <http://dx.doi.org/10.1016/j.epsl.2011.02.004>.

Göpel C., Manhès G. and Allegre C. J. (1985) U-Pb systematics in iron meteorites – Uniformity of primordial lead. *Geochim. Cosmochim. Acta* 49, 1681-1695. [http://dx.doi.org/10.1016/0016-7037\(85\)90139-5](http://dx.doi.org/10.1016/0016-7037(85)90139-5).

Hallis L., Anand M. and Strekopytov S. (2014) Trace-element modelling of mare basalt parental melts: Implications for a heterogeneous lunar mantle. *Geochim. Cosmochim. Acta* 134, 289-316. <http://dx.doi.org/10.1016/j.gca.2014.01.012>.

Korotev R. L., Jolliff B. L., Zeigler R. A., Seddio S. M. and Haskin L. A. (2011) Apollo 12 revisited. *Geochim. Cosmochim. Acta* 75, 1540-1573. <http://dx.doi.org/10.1016/j.gca.2010.12.018>.

Langmuir C. H., Vocke R. D., Hanson G. N. and Hart S. R. (1978) A general mixing equation with applications to Icelandic basalts. *Earth Planet. Sci. Lett.* 37, 380-392. [http://dx.doi.org/10.1016/0012-821X\(78\)90053-5](http://dx.doi.org/10.1016/0012-821X(78)90053-5).

Ludwig K. R. (2008) User's Manual for Isoplot 3.60, A geochronological toolkit for Microsoft Excel. Berkeley Geochronological Center Special Publication 4. Berkeley Geochronological Center, Berkeley, California.

McLeod C. L., Brandon A. D. and Armytage R. M. G. (2014) Constraints on the formation age and evolution of the Moon from ^{142}Nd - ^{143}Nd systematics of Apollo 12 basalts. *Earth Planet. Sci. Lett.* 396, 179-189. <http://dx.doi.org/10.1016/j.epsl.2014.04.007>.

Meyer J., Elkins Tanton L. T. and Wisdom J. (2010) Coupled thermal-orbital evolution of the early Moon. *Icarus* 208, 1-10. <http://dx.doi.org/10.1016/j.icarus.2010.01.029>.

Murthy V., Evensen N., Jahn B.-M. and Coscio M. R. (1971) Rb-Sr ages and elemental abundances of K, Rb, Sr, and Ba in samples from the Ocean of Storms. *Geochim. Cosmochim. Acta* 35, 1139-1153. [http://dx.doi.org/10.1016/0016-7037\(71\)90030-5](http://dx.doi.org/10.1016/0016-7037(71)90030-5).

Neal C. R. and Taylor L. A. (1992) Petrogenesis of mare basalts - A record of lunar volcanism. *Geochim. Cosmochim. Acta* 56, 2177-2211. [http://dx.doi.org/10.1016/0016-7037\(92\)90184-K](http://dx.doi.org/10.1016/0016-7037(92)90184-K).

Neal C. R., Hacker M. D., Snyder G. A., Taylor L. A., Liu Y. G. and Schmitt R. A. (1994a) Basalt generation at the Apollo 12 site, Part 1: New data, classification, and re-evaluation. *Meteoritics* 29, 334-348.

Neal C. R., Hacker M. D., Snyder G. A., Taylor L. A., Liu Y. G. and Schmitt R. A. (1994b) Basalt generation at the Apollo 12 site, Part 2: Source heterogeneity, multiple melts, and crustal contamination. *Meteoritics* 29, 349-361.

Nyquist L. E., Bansal B. M. and Wiesmann H. (1975) Rb-Sr ages and initial $^{87}\text{Sr}/^{86}\text{Sr}$ for Apollo 17 basalts and KREEP basalt 15386. *Lunar Planet. Sci. VI. Lunar Planet. Inst., Houston*. pp. 1445-1465.

Nyquist L. E., Bansal B. M., Wooden J. L. and Wiesmann H. (1977) Sr-isotopic constraints on the petrogenesis of Apollo 12 mare basalts. *Lunar Planet. Sci. VIII. Lunar Planet. Inst., Houston*. pp. 1383-1415.

Nyquist L. E., Shih C.-Y., Wooden J. L., Bansal B. M. and Wiesmann H. (1979) *Lunar Planet. Sci. X. Lunar Planet. Inst., Houston*. pp. 77-114.

Nyquist L. E., Wooden J. L., Shih C.-Y., Wiesmann H. and Bansal B. M. (1981) Isotopic and REE studies of lunar basalt 12038: implications for petrogenesis of aluminous mare basalts. *Earth Planet. Sci. Lett.* 55, 335-355. [http://dx.doi.org/10.1016/0012-821X\(81\)90162-X](http://dx.doi.org/10.1016/0012-821X(81)90162-X).

Papanastassiou D. A. and Wasserburg G. J. (1970) Rb-Sr ages from the Ocean of Storms. *Earth Planet. Sci. Lett.* 8, 269-278. [http://dx.doi.org/10.1016/0012-821X\(70\)90111-1](http://dx.doi.org/10.1016/0012-821X(70)90111-1).

Papanastassiou D. A. and Wasserburg G. J. (1971) Lunar chronology and evolution from Rb-Sr studies of Apollo 11 and 12 samples. *Earth Planet. Sci. Lett.* 11, 37-62. [http://dx.doi.org/10.1016/0012-821X\(71\)90139-7](http://dx.doi.org/10.1016/0012-821X(71)90139-7).

Rhodes J. M., Brannon J. C., Rodgers K. V., Blanchard D. P. and Dungan M. A. (1977) Chemistry of
 Apollo 12 mare basalts - Magma types and fractionation processes. Lunar Planet. Sci. VIII. Lunar
 Planet. Inst., Houston. pp. 1305-1338.

Snape J. F., Nemchin A. A., Bellucci J. J., Whitehouse M. J., Tartèse R., Barnes J. J., Anand M.,
 Crawford I. A. and Joy K. H. (2016) Lunar basalt chronology, mantle differentiation and implications
 for determining the age of the Moon. Earth Planet. Sci. Lett. 451, 149-158.
<http://dx.doi.org/10.1016/j.epsl.2016.07.026>.

Snyder G. A., Neal C. R., Taylor L. A. and Halliday A. N. (1997) Anatexis of lunar cumulate mantle
 in time and space: Clues from trace-element, strontium, and neodymium isotopic chemistry of parental
 Apollo 12 basalts. Geochim. Cosmochim. Acta 61, 2731-2747. [http://dx.doi.org/10.1016/S0016-7037\(97\)00082-3](http://dx.doi.org/10.1016/S0016-7037(97)00082-3).

Sprung P., Kleine T. and Scherer E. E. (2013) Isotopic evidence for chondritic Lu/Hf and Sm/Nd of
 the Moon. Earth Planet. Sci. Lett. 380, 77-87. <http://dx.doi.org/10.1016/j.epsl.2013.08.018>.

Turner G. (1971) ^{40}Ar - ^{39}Ar ages from the lunar maria. Earth Planet. Sci. Lett. 11, 169-191.
[http://dx.doi.org/10.1016/0012-821X\(71\)90161-0](http://dx.doi.org/10.1016/0012-821X(71)90161-0).

Unruh D. M., Stille P., Patchett P. J. and Tatsumoto M. (1984) Lu-Hf and Sm-Nd evolution in lunar
 mare basalts. J. Geophys. Res. 89, B459-B477. <http://dx.doi.org/10.1029/JB089iS02p0B459>.

Vollmer R. (1976) Rb-Sr and U-Th-Pb systematics of alkaline rocks: the alkaline rocks from Italy.
 Geochim. Cosmochim. Acta 40, 283-295. [http://dx.doi.org/10.1016/0016-7037\(76\)90205-2](http://dx.doi.org/10.1016/0016-7037(76)90205-2).

Warner J. L. (1971) Lunar crystalline rocks: Petrology and geology. Lunar Planet. Sci. II. Lunar
 Planet. Inst., Houston. pp. 469-480.

Weill D. F., Grieve R. A., McCallum I. S. and Bottinga Y. (1971) Lunar Planet. Sci. II. Lunar Planet.
 Inst., Houston. pp. 413-430.

- 539 Whitehouse M. J., Kamber B. S., Fedo C. M. and Lepland A. (2005) Integrated Pb- and S-isotope
1 investigation of sulphide minerals from the early Archaean of southwest Greenland. Chem. Geol. 222,
2 112-131. <http://dx.doi.org/10.1016/j.chemgeo.2005.06.004>.
3
4
5
6
7 542 Woodhead J. D. and Hergt J. M. (2000) Pb-Isotope Analyses of USGS Reference Materials. Geostand.
8
9 543 Geoanal. Res. 24, 33-38. <http://dx.doi.org/10.1111/j.1751-908X.2000.tb00584.x>.
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

Figure and Table captions

Figure 1. – Back Scattered Electron (BSE) images of representative regions in: (a) 12002,600; (b) 12012,34; (c) 12021,619; (d) 12005,78; (e) 12016,39, with and typical analytical areas highlighted for each sample. Examples of the main mineral phases present in each image have been labelled (Pl = plagioclase; Px = pyroxene; Ol = olivine; Sil = silica; Ilm = ilmenite). Complete BSE sample maps and false colour element maps are included in the Electronic Appendix (Figs. A.1-A.5).

Figure 2. – $^{207}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$ plots of the filtered data sets for the five Apollo 12 basalts in this study, plus three samples (12039, 12063 and 12038) presented in Snape et al. (2016). In addition to the primary sample isochrons, paleo-isochrons (in red) have been constructed using the time and composition of magmatic differentiation specified in the multiple stage Pb evolution model of Snape et al. (2016). In each case, the point at which the sample isochron and paleo-isochron intersect provides an independent model prediction for the initial Pb composition of each sample. Error bars represent 2σ uncertainties.

Figure 3. – $^{208}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$ plots of the filtered data sets for the five Apollo 12 basalts in this study, plus three samples (12039, 12063 and 12038) presented in Snape et al. (2016). Grey triangular fields mark the range of compositions in each sample. The points at which these fields intersect the y-axis represents the radiogenic endmember compositions for each samples, and the approximate Th/U ratios that these $^{208}\text{Pb}/^{206}\text{Pb}$ ratios equate to have been indicated. Error bars represent 2σ uncertainties.

Figure 4. – Comparison of the Pb-Pb isochron ages for the Apollo 12 basalts with previous age determinations by other methods (Papanastassiou and Wasserburg 1970; 1971; Compston et al. 1971; Turner 1971; Murthy et al. 1971; Alexander et al. 1972; Nyquist et al. 1977; 1979; 1981). Where possible the literature ages have been recalculated to account for updated decay constants and other factors (an exception being the Ar-Ar ages of 12002, where there was not sufficient information to recalculate the monitor ages). Error bars represent 2σ uncertainties.

Figure 5. – Hypothetical cross-section of the Apollo 12 landing site and underlying basalt flows, taking into account the new crystallisation ages from this study. The vertical scale has been exaggerated for clarity.

Figure 6. – (a) $^{207}\text{Pb}/^{206}\text{Pb}$ vs. $^{204}\text{Pb}/^{206}\text{Pb}$ plot of the initial Pb compositions for the Apollo 12 basalts analysed in this study and those presented by Snape et al. (2016). The square symbols (without error bars) indicate the initial compositions predicted by the Snape et al. (2016) Pb isotopic evolution model. The schematic orange arrow indicates a hypothetical Pb growth curve in a relatively high- μ (1000) source reservoir from an average ilmenite basalt composition between 3190-3130 Ma. Note, even with such an unrealistically high μ -value, there would not be sufficient growth to explain the range of compositions in the different basaltic suites by evolution of a single magmatic source. (b) Comparison of the initial Pb compositions for the Apollo 12 basalts with the multiple stage Pb evolution model of Snape et al. (2016). Several of the initial Pb isotopic compositions of the Apollo 12 basalts (particularly those of the olivine basalts) lie below the appropriate paleo-isochrons constructed from the model differentiation point, indicating either an underestimation of these initial Pb compositions or a problem with the model. (c) A significantly lower μ -value (50) for the homogeneous lunar mantle results in a better fit between the olivine basalts and the paleo-isochrons, but cannot then explain the initial Pb compositions of the pigeonite and ilmenite basalts. (d) A much later differentiation event (at ~3100 Ma) can potentially explain the initial Pb compositions of all the Apollo 12 basalts, but would require that they evolved in sources with very high μ -values (>1000). The growth curves are all calculated using Canyon Diablo Troilite (CDT; Göpel et al. 1985) to represent a primitive Pb isotopic starting composition.

Figure 7. – (a) Age vs. the measured initial $^{206}\text{Pb}/^{204}\text{Pb}$ ratios for each sample compared with the age and composition of the magmatic differentiation event described by the model of Snape et al. (2016). The model predicted initial $^{206}\text{Pb}/^{204}\text{Pb}$ ratios for each sample have also been indicated (square symbols). Focusing on the ages and initial compositions of the Apollo 12 samples (b), it is clear that while the measured values indicate that the three main basaltic suites (olivine, pigeonite and ilmenite)

would have evolved with distinct μ -values, the model predicted values indicate an overlap between the olivine and pigeonite basalts.

Figure 8. – Comparison of estimated source μ -values for the Apollo 12 basalts with equivalent (a) $^{87}\text{Rb}/^{86}\text{Sr}$ and (b) $^{147}\text{Sm}/^{144}\text{Nd}$ ratios. The dashed line (and surrounding grey field) in each plot indicates the hypothetical mixing trend between a low- μ mafic cumulate and a high- μ KREEP-rich residual composition. The red zones within these fields indicate the range of likely μ , Rb/Sr and Sm/Nd values of the mafic cumulate. Error bars represent 2σ uncertainties.

Table 1. – Summary of the crystallisation ages, initial Pb isotopic compositions and source μ -values for the Apollo 12 basalts analysed in this study. The values for the three Apollo 12 samples published in Snape et al. (2016) are included for comparison.

Supplementary material

Appendix A - Figures A.1-A.5 – (a) Back Scattered Electron (BSE) and (b) false colour element maps of each sample analysed in this study.

Appendix B - Table B.1. – Modal abundances for the basalts analysed in this study. Table B.2. – Complete datasets for the five basalts analysed in this study. Table B.3. – Average background values for the electron multipliers during the two analytical sessions during which the data were collected. Table B.4. – Extended summary table for the Apollo 12 basalts, including previous age determinations (recalculated for updated decay constants). Also included are the recalculated initial $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, as well the source $^{87}\text{Rb}/^{86}\text{Sr}$ or $^{147}\text{Sm}/^{144}\text{Nd}$ ratios used in the modelling calculations.

Appendix C – Supplementary text outlining the recalculation of initial $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios and source $^{87}\text{Rb}/^{86}\text{Sr}$ or $^{147}\text{Sm}/^{144}\text{Nd}$ ratios, as well as the propagation of analytical uncertainties in the calculations.

Figure 1 (cont.)

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Table 1

Sample	Basalt type	Age	±2σ	Source μ _m ±2σ			Initial Pb isotope composition (measured - lowest estimate)						Initial Pb isotope composition (predicted)	
							²⁰⁴ Pb/ ²⁰⁶ Pb	±2σ	²⁰⁷ Pb/ ²⁰⁶ Pb	±2σ	²⁰⁸ Pb/ ²⁰⁶ Pb	±2σ	²⁰⁴ Pb/ ²⁰⁶ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb
12038*	Feldspathic	3242	13	531	61	499	0.00509	0.00047	1.166	0.011	1.014	0.010	0.00536	1.208
12002	Olivine	3167	12	733	29	655	0.00364	0.00010	1.083	0.006	0.851	0.005	0.00402	1.167
12012	Olivine	3151	21	749	43	645	0.00353	0.00016	1.064	0.007	0.894	0.006	0.00403	1.162
12039*	Pigeonite	3129	10	639	36	630	0.00401	0.00019	1.152	0.005	0.890	0.004	0.00406	1.156
12021	Pigeonite	3176	6	641	41	591	0.00413	0.00023	1.106	0.007	0.970	0.330	0.00443	1.176
12005	Ilmenite	3182	10	471	89	459	0.00541	0.00085	1.190	0.008	0.933	0.038	0.00552	1.192
12016	Ilmenite	3186	11	460	47	427	0.00553	0.00046	1.169	0.010	0.910	0.009	0.00588	1.198
12063*	Ilmenite	3193	11	415	43	424	0.00606	0.00050	1.146	0.010	0.915	0.009	0.00595	1.201

Source μ_m values calculated from measured estimates of initial Pb composition. Predicted initial Pb compositions and μ_p values calculated based on Snape et al. (2016) model.

*Originally reported in Snape et al. (2016) - summary data included here for completeness.

Appendix A

[Click here to download Background dataset for online publication only: a12_basalt_sources_appendixA_figs.pdf](#)

Appendix C

[Click here to download Background dataset for online publication only: a12_basalt_sources_appendix_c.docx](#)