

Pb isotope systematics of the achondrite Asuka 881394 revisited.

Y. Amelin¹, P. Koekoed¹ and Q.-Z. Yin, ¹Research School of Earth Sciences, The Australian National University, Canberra ACT, Australia, ³Department of Earth and Planetary Sciences, University of California at Davis, One Shields Avenue, Davis, CA 95616, USA.

Introduction:

Asuka 881394 is an achondrite that is similar in mineralogy to coarse-grained cumulate eucrites [1], but differs in oxygen and ⁵⁴Cr isotopic composition [2-4] and is therefore thought to have originated from a different parent body than the eucrites and other HED meteorites. Wadhwa et al. 2009 [4] reported U-Pb, Al-Mg and Mn-Cr data showing that Asuka 881394 is the oldest achondrite studied so far.

The age intervals between Asuka 881394 and quenched angrites (e.g. D'Orbigny) are, however, inconsistent between the chronometer systems. The difference between the ages of Asuka 881394 and D'Orbigny measured with Pb isotopes is 2.1±0.3 Ma, whereas the age difference measured with the ²⁶Al-²⁶Mg and ⁵³Mn-⁵³Cr systems is 1.0±0.2 Ma or 0.90±0.4 Ma, respectively [4,5]. These age discrepancies hamper using Asuka 881394 as one of the benchmarks of the early Solar System chronology

Possible causes of these age discrepancies, including closure of isotopic clocks at different temperatures, disturbance of isotopic systems by secondary processes, and possibly heterogeneous distribution of ²⁶Al/²⁷Al, ⁵³Mn/⁵⁵Mn and ²³⁸U/²³⁵U in the accreting Solar System were discussed by [4, 5], but the culprit remains to be identified. Considering importance of Asuka 881394 as the oldest known non-chondritic material, we are undertaking a systematic re-examination of its chronometry in order to understand the reason of the age discrepancy. As the first step, Wimpenny et al. [5] reported a new ²⁶Al-²⁶Mg isochron date based on analysis of multiple plagioclase and pyroxene fractions of variable mineralogical purity, which is more precise than the previously published date [4] but consistent within uncertainty.

Our initial study of the U-Th-Pb systematics of Asuka 881394 [4] yielded a precise Pb-Pb isochron of 4566.51±0.21 Ma (ages are reported relative to ²³⁸U/²³⁵U=137.88, unless indicated otherwise) from 16 analyses of acid-washed pyroxene fractions and 2 analyses of acid-washed bulk rock fractions. The isochron included 16 analyses with highly radiogenic Pb isotope composition (blank-corrected ²⁰⁶Pb/²⁰⁴Pb between 423 and 2675) but two less radiogenic data points (blank-corrected ²⁰⁶Pb/²⁰⁴Pb of 87 and 135). Because of the small fraction sizes and hence large blank correction, the more radiogenic analyses had large (although strongly correlated) uncertainties and produced overlapping error ellipses (Fig. 1). An isochron plot for all data (Fig.1) clearly shows that the isochron is effectively a three-point line, and its slope and intercept are largely controlled by the two least radiogenic analyses:

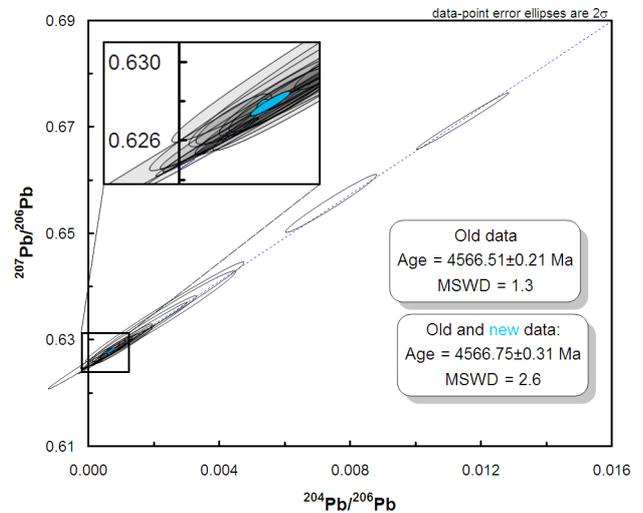


Fig. 1. All Pb isotope data for acid-washed pyroxene and bulk rock fractions. Grey ellipses – data from [4], blue ellipses – this study.

If the isotopic composition of non-radiogenic Pb differs between the more radiogenic group and less radiogenic fractions (which is possible considering the presence of both primordial and “terrestrial” Pb components in the acid leachates [4]), then the isochron age may be biased by up to 0.5 Ma or possibly more. Here we re-examine the Pb isotopic systematics of Asuka 881394 using an approach that differs in several ways from our first study of this meteorite [4] and is designed to evaluate the possible systematic uncertainty in the Pb isochron age, and to better understand the discrepancy between the Pb isotopic and ²⁶Al-²⁶Mg and ⁵³Mn-⁵³Cr ages.

Methods:

We have analysed five mineral fractions hand-picked from the mineral concentrates prepared by magnetic separation: 2.56 mg silicon oxide (tridymite), 28.03 mg (pure) and 39.66 mg (less pure) plagioclase fractions, 30.03 mg pyroxene fraction, and 25.86 mg bulk rock fraction. These fractions are larger than that in the previous study, and the procedure blanks (between 0.35 and 1.2 pg Pb for various leaching steps) are lower, hence the data are less dependent on blank corrections. Each fraction was leached in five steps, the first three being similar to our conventional procedure using dilute HBr and HNO₃, 7M HNO₃ and 6M HCl, and the two final steps using dilute HF (see Fig. 2 caption for details), before complete dissolution of the residue in concentrated HF. All materials were spiked with a ²⁰²Pb-²⁰⁵Pb-²²⁹Th-²³³U-²³⁶U tracer. Separated Pb was analysed on a modified MAT-261 mass spectrometer at the ANU using secondary electron multiplier.

Results:

The data are shown in Fig. 2:

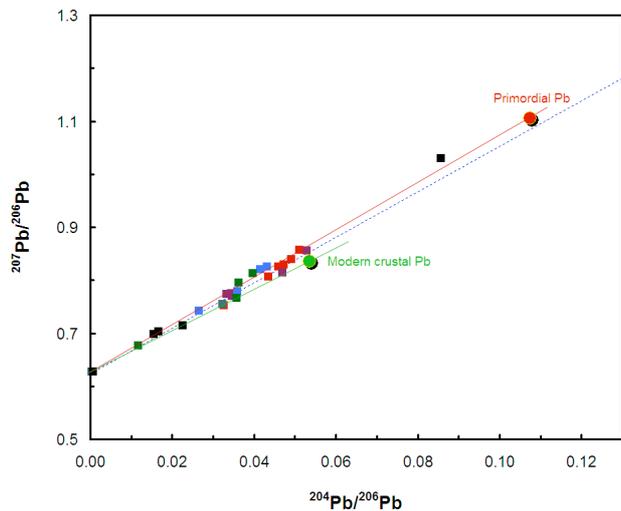


Fig. 2. Pb isotope data for all leaching steps and all mineral fractions analysed in this study. Red – 0.3M HBr (W1a), brown – 0.5M HNO₃ (W1b), blue – combined steps with 7M HNO₃ and 6M HCl (W2), dark green – 0.2M HF (W3), black – 1M HF (W4) and complete dissolution of the residue (R).

The data lie (within uncertainty) between the tie lines connecting the most radiogenic data with primordial Pb and modern terrestrial crustal Pb (shown in Fig. 2 with red and green lines, respectively), and can be thus interpreted as mixtures between these three components. There is no direct indication of presence of Pb that underwent multi-stage evolution in the Pb isotopic data. The 1M HF washes and residues of pyroxene and bulk fractions contain highly radiogenic Pb with measured $^{206}\text{Pb}/^{204}\text{Pb}$ between 900-1400.

Leaching steps for each mineral show a general trend of increasing the ratio of radiogenic to non-radiogenic Pb as the leaching progresses. No radiogenic Pb in the early leaching steps, such as found in angrites and some eucrite-like meteorites (e.g. [6]) attributable to the presence of U-rich phosphate minerals (apatite or merrillite), has been detected in Asuka 881394. If these minerals are present, they have low U and Th low content, and play little part in the balance of these elements.

The distribution of Pb components varies significantly between the minerals. Tridymite and pure plagioclase leaching steps plot along the line between radiogenic Pb and primordial Pb, with almost complete absence of “terrestrial”-like component. The arrays of bulk rock and impure plagioclase leaching steps are more scattered and show “terrestrial” Pb as the main non-radiogenic component. The pyroxene array (excluding W2) passes close to primordial Pb and yields the apparent age of 4567.7 ± 1.2 Ma (MSWD=7.3).

A strange, and still unexplained, observation in this study is that the 1M HF washes of pyroxene and bulk fractions contain radiogenic Pb with substantially higher $^{207}\text{Pb}/^{206}\text{Pb}$ than similarly radiogenic residues of the same fractions (Fig. 3):

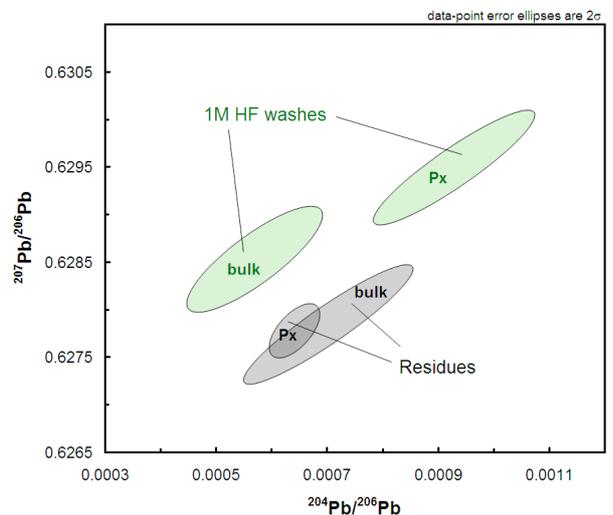


Fig. 3. Pb isotope data for 1M HF leachates and residues of pyroxene and bulk rock fractions.

This discrepancy cannot be explained by the presence of non-radiogenic Pb or by a natural open-system behavior, but, rather, looks like an evidence of isotopic fractionation in radiogenic Pb induced by leaching, similar to that reported in [7] for leaching with concentrated HBr. The application of multi-stage leaching involving HF steps to achondrites requires, therefore, rigorous testing, before it can be considered a safe and reliable technique in support of Pb-isotopic dating.

Discussion:

Addition of the new, more precise pyroxene and bulk rock residue analyses to the original residue data set from [4] increases the age from 4566.51 ± 0.21 Ma to 4566.75 ± 0.31 Ma, suggesting that the originally reported value should be considered a minimum age. This change does not reduce the Asuka – D’Orbigny age difference measured with U-Pb and the extinct radionuclide systems, but, rather, confirms and slightly increases it. Using the measured $^{238}\text{U}/^{235}\text{U} = 137.768 \pm 0.038$ [5] makes the new age value 4565.57 ± 0.55 Ma. Since the $^{238}\text{U}/^{235}\text{U}$ measured in Asuka 881394 is the same, within uncertainty, as in the angrites [8,9], correction for U isotope composition also has no appreciable effect on the age interval. These observations, together with recent report of refined ^{26}Al - ^{26}Mg systematics [5] that confirmed the earlier reports, suggest that the age discrepancies are likely to be caused by natural factors such as heterogeneous distribution of ^{26}Al and ^{53}Mn , rather than by artifacts of isotopic dating.

References:

- [1] Nyquist et al (2003) *EPSL*, 214, 11.
- [2] Scott et al. (2009), *GCA*, 73, 5835.
- [3] Sanborn and Yin (2014) *LPSC* 45, #2016.
- [4] Wadhwa et al. (2009) *GCA*, 73, 5189.
- [5] Wimpenny et al. (2013) *LPSC* 44, #2303.
- [6] Iizuka et al (2014) *GCA* 132, 259-273.
- [7] Amelin et al. (2010) *EPSL*, 300, 343-350.
- [8] Brennecka and Wadhwa (2012) *PNAS* 109, 9299-9303.
- [9] Amelin et al. (2011) *LPSC* 42, #1682.