

## AMS $^{14}\text{C}$ AGES OF YAMATO ACHONDRITIC METEORITES

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**Abstract:** Terrestrial  $^{14}\text{C}$  ages of some Yamato achondrites and other meteorites, have been determined by accelerator mass spectrometry (AMS). Samples of meteorites previously studied for  $^{14}\text{C}$  are reported, as are new measurements on Yamato achondrites. Results on a number of meteorites from the Yamato-79 series shows longer terrestrial ages than expected for this site, where young ages predominate. A new  $^{14}\text{C}$  age on Y-74037 confirms the younger age for this diogenite similar to Y-74097, whereas Y-74010 is older (15 kyr) and these two data suggest there may be more than one diogenite fall in the Yamato-74 group.

### 1. Introduction

The terrestrial age of a meteorite is an important parameter in the study of fall times, meteorite distributions, and meteorite concentration mechanisms. The radio-nuclide  $^{14}\text{C}$  ( $t_{1/2} = 5730$  years) is very useful in most meteorite collection areas of the earth for determining these ages. The first work on  $^{14}\text{C}$  in Antarctic meteorites, using counter techniques, was reported by FIREMAN (1978, 1979). Since 1984, nearly all  $^{14}\text{C}$  measurements on meteorites have been done by accelerator mass spectrometry (AMS). The literature on  $^{14}\text{C}$  terrestrial age measurements has been summarized by JULL and DONAHUE (1988), BEUKENS *et al.* (1988) and JULL *et al.* (1989). Longer ages such as are often observed in the Allan Hills Main Icefield region of Antarctica can be determined using the longer-lived nuclide  $^{36}\text{Cl}$  (NISHIIZUMI *et al.*, 1989). As has already been observed by NISHIIZUMI *et al.* (1989) and BEUKENS *et al.* (1988), the Yamato site appears to have a much younger distribution of meteorite falls than the Allan Hills Main Icefield.

### 2. Experimental

Samples of meteorites of 0.2–1.0 g were crushed, mixed with 3–5 g of iron chips, used to enhance combustion, and preheated in air at 500°C for one hour. As has been shown by JULL *et al.* (1989), this step is important to remove many contaminants. The samples are then loaded into an RF furnace, and heated in a flow of oxygen up to about 1700°C. The rock-iron mix fuses completely. The gases evolved are passed over

$\text{MnO}_2$  and  $\text{CuO/Pt}$ , collected at  $-196^\circ\text{C}$ , and then excess oxygen is removed. Carbon dioxide is separated from any water at  $-78^\circ\text{C}$ . The  $\text{CO}_2$  volume is measured in a known volume using a capacitance manometer, and diluted to about  $1\text{ cm}^3$  STP with  $^{14}\text{C}$ -free  $\text{CO}_2$ . The gas is then reduced to graphite as described by SLOTA *et al.* (1987). The graphite powder is pressed into an accelerator target, and the targets are mounted in the accelerator ion source. Two standards are used in each run, and the isotope ratio of  $^{14}\text{C}/^{13}\text{C}$  is measured by AMS, as discussed by LINICK *et al.* (1986). Compared to our earlier paper (JULL *et al.*, 1989), we found a systematic error of 7% during a recalibration of the standard volume used in the measurement. The saturated activities for different meteorites are thus 7% higher than reported by JULL *et al.* (1989), as is discussed elsewhere (JULL *et al.*, 1993). The saturated activity for a given class of meteorite is calculated by normalization of the  $^{14}\text{C}$  content of Bruderheim (L6; mean value:  $51.1 \pm 1.4$  dpm/kg) to the oxygen content of the meteorite.

### 3. Results and Discussion

The results of the  $^{14}\text{C}$  measurements are presented in Table 1. A sample of a recent fall, Kokubunji (L6), gave an activity of  $45.0 \pm 0.4$  dpm  $^{14}\text{C}/\text{kg}$ . This value is consistent with a sample taken from the top 10 cm of a meteorite of typical size ( $< 45$  cm), and can be compared to the depth profile we have measured in Knyahinya (REEDY *et al.*, 1993). High  $^{22}\text{Ne}/^{21}\text{Ne}$  values in Kokubunji also support this conclusion (LOEKEN *et al.*, 1992). The scatter in the  $^{14}\text{C}$  measurements of different fragments of Bruderheim is also consistent with these results. We compared our results on three chondrites, Yamato (Y)-75271, Y-791717 and Y-791630 to the results obtained previously by BEUKENS *et al.* (1988). Good agreement in  $^{14}\text{C}$  age was obtained, as indicated in Table 1. Our  $^{14}\text{C}$  results in dpm/kg also indicate analytical errors and the terrestrial age estimate additionally includes the uncertainties associated with lack of knowledge of the sample position and the pre-atmospheric size of the meteorite (REEDY *et al.*, 1993), which is typically about  $\pm 15\%$ . Also listed are the results of FIREMAN (1978) on ALHA 77256. Our measurements show excellent agreement with these results.

The  $^{14}\text{C}$  results on six eucrites (Y-790260, -791186, -791960, -791962, -792510 and -82082) show a range from 20.1 to 34 kyr age. It is unlikely that this range, which is a factor of 5 in  $^{14}\text{C}$  content could be explained by a common fall for all these meteorites. Samples Y-791960 and -791962 were found in the same location, and differ by about 4 kyr in apparent  $^{14}\text{C}$  age. NAGAO and OGATA (1989) proposed that these meteorites are paired on the basis of  $^{81}\text{Kr}$  and  $^{21}\text{Ne}$ , although they do differ significantly in  $^{38}\text{Ar}$ . The difference in  $^{14}\text{C}$  is most likely due to either some amount of weathering  $^{14}\text{C}$  contamination, rather than a difference in location in a common fall. Thus, there is insufficient evidence to indicate whether these two are separate falls. In order to assess the importance of weathering, we removed the weathering carbonates using the phosphoric-acid procedure (JULL *et al.*, 1993). Our results are given in Table 2. For the three older meteorites (Y-791960, Y-791962 and Y-792510), weathering could contribute some  $^{14}\text{C}$  and could measurably shift the  $^{14}\text{C}$  age for Y-791960 to 29 kyr. Y-791962 would be only slightly corrected, and would remain about 26 kyr. These effects underline the importance of assessing the degree of weathering in older falls, and removing the

Table 1. Terrestrial  $^{14}\text{C}$  age measurements.

Sample	Type	Weight (g)	$^{14}\text{C}$ dpm/kg	Saturated activity <sup>1</sup>	Age (kyr) <sup>2</sup>	Literature values
Kokubunji 10	L-6	0.232	45.0 $\pm$ 0.4	51.1	—	
ALHA77256	Dio	1.00	17.7 $\pm$ 0.4	61.1	10.2 $\pm$ 1.3	11.1 $\pm$ 1.0 <sup>3</sup>
Y-75271,51	L-5	0.177	40.5 $\pm$ 0.6	51.1	1.9 $\pm$ 1.3	1.9 $\pm$ 0.1 <sup>3</sup>
Acid residue		0.159	40.5 $\pm$ 0.3	51.1	1.9 $\pm$ 1.3	
Y-791717,55	CO <sub>3</sub>	0.226	28.0 $\pm$ 0.3	50.6	4.9 $\pm$ 1.3	5.5 $\pm$ 0.1 <sup>3</sup>
Acid residue		0.120	27.2 $\pm$ 0.4	50.6	5.1 $\pm$ 1.3	
Y-791630,64	L-4	0.300	44.3 $\pm$ 2.5 <sup>3</sup>	51.1	1.2 $\pm$ 1.4	0.5 $\pm$ 0.1 <sup>3</sup>
Y-74010,81	Dio	0.502	9.9 $\pm$ 0.1	61.1	15.0 $\pm$ 1.3	
Y-74037,88	Dio	0.229	14.8 $\pm$ 0.2	61.1	11.7 $\pm$ 1.3	
Y-790007,72	Euc	0.559	37.4 $\pm$ 0.3	60.3	3.6 $\pm$ 1.3	29 $\pm$ 34 <sup>4</sup>
Y-790260,90	Euc	0.180	3.47 $\pm$ 0.16	60.3	23.6 $\pm$ 1.4	140 $\pm$ 32 <sup>4</sup>
Y-791186,51	Euc	0.251	0.98 $\pm$ 0.11	60.3	> 32 <sup>6</sup>	240 $\pm$ 40 <sup>5</sup>
Y-791960,52	Euc	0.113	2.48 $\pm$ 0.22	60.3	> 25 <sup>6</sup>	270 $\pm$ 40 <sup>5</sup>
Y-791962,50	Euc	0.214	1.47 $\pm$ 0.12	60.3	> 29 <sup>6</sup>	270 $\pm$ 40 <sup>5</sup>
Y-792510,95	Euc	0.126	2.84 $\pm$ 0.2	60.3	25.3 $\pm$ 1.4	230 $\pm$ 40 <sup>5</sup>
Y-82082,64	Euc	0.360	5.29 $\pm$ 0.09	60.3	20.1 $\pm$ 1.3	160 $\pm$ 50 <sup>5</sup>

<sup>1</sup> The saturated activity assumed for this type of meteorite, based on oxygen content and the measurements of JULL *et al.* (1989, 1993) and REEDY *et al.* (1993).

<sup>2</sup> Note that the calculated error of typically  $\pm 1.3$  kyr includes errors associated with lack of knowledge of location of the sample in the meteoroid. This error is much larger than analytical errors, listed in column 4 for  $^{14}\text{C}$  content.

<sup>3</sup> Literature  $^{14}\text{C}$  values include counting errors only. Data compiled from BEUKENS *et al.* (1988), FIREMAN and NORRIS (1981).

<sup>4</sup>  $^{81}\text{Kr}$  data of SCHULTZ (1986).

<sup>5</sup>  $^{81}\text{Kr}$  data of NAGAO and OGATA (1989) and MIURA *et al.* (1991).

<sup>6</sup>  $^{14}\text{C}$  ages quoted as limits due to small sample size and uncertainty in removal of all weathering carbonates.

Table 2. Acid-hydrolysis experiments.

Sample	Type	Weight (g)	Amount of carbonate (cm <sup>3</sup> CO <sub>2</sub> )	$^{14}\text{C}$ dpm/kg	Percent modern $^{14}\text{C}$ <sup>1</sup>	$^{14}\text{C}$ age (BP)
Y-791717	CO <sub>3</sub>	0.271	0.282	9.45 $\pm$ 0.01	138 $\pm$ 1	post-bomb
Y-75271	L5	0.295	0.0281	1.13 $\pm$ 0.01	79 $\pm$ 2	1950 $\pm$ 200
Y-791960	Euc	0.083	0.0286	0.69 $\pm$ 0.25	30 $\pm$ 11	9800 $\pm$ 3000
Y-791186	Euc	0.219	0.0127	0.21 $\pm$ 0.01	54 $\pm$ 2	5100 $\pm$ 300
Y-792510	Euc	0.355	0.0144	0.23 $\pm$ 0.01	87 $\pm$ 4	1100 $\pm$ 400

<sup>1</sup> Amount of  $^{14}\text{C}$  compared to modern (1950AD) atmospheric  $^{14}\text{C}$ .

weathering carbonates from these older samples.

In contrast to these six older eucrites, Y-790007 has also been studied. This meteorite has a very young terrestrial age of 3.6 kyr, and this measurement confirms the short  $^{81}\text{Kr}$  terrestrial age of  $20 \pm 20$  kyr of SCHULTZ (1986). Our measurements on the other

euclrites appear inconsistent with long  $^{81}\text{Kr}$  age indicated in Table 1. The two youngest ages of 20.1 kyr (Y-82082) and 23.6 kyr (Y-790260) seems too young to be explicable in terms of weathering  $^{14}\text{C}$  contamination. Table 2 shows results of  $^{14}\text{C}$  in dpm/kg from terrestrial weathering products from three euclrites, which range from 0.21 to 0.69 dpm/kg, and which are much less than the several dpm/kg needed to explain the euclrite  $^{14}\text{C}$  data if the  $^{81}\text{Kr}$  results are correct. The results for Y-82082 are definitely indicative of cosmogenic  $^{14}\text{C}$  as the undiluted  $\text{CO}_2$  activity was 446% modern terrestrial  $^{14}\text{C}$ , well above terrestrial levels. The undiluted  $\text{CO}_2$  for Y-790260 is also 110% modern terrestrial  $^{14}\text{C}$  and, judging by the fraction of modern  $^{14}\text{C}$  observed in weathering products in Table 2 and the results of JULL *et al.* (1993), is also cosmogenic  $^{14}\text{C}$ . Any cosmogenic  $^{14}\text{C}$  would have decayed long ago for samples of the ages suggested by the  $^{81}\text{Kr}$  results (NAGAO and OGATA, 1989; MIURA *et al.*, 1991; SCHULTZ, 1986). The inconsistency in the other euclrites in Table 1, compared to the data of NAGAO and OGATA (1989) and MIURA *et al.* (1991), may be explicable in terms of weathering carbonates which have not been completely removed. For these samples, the undiluted  $\text{CO}_2$  released on combustion gave 71% (Y-791186), 68% (Y-791960), 87% (Y-791962), 29% (Y-792510) of the value of modern terrestrial  $^{14}\text{C}$ . These levels can be compared to the  $^{14}\text{C}$  composition of the weathering products in Table 2. Weathering-produced terrestrial  $^{14}\text{C}$  could definitely explain a substantial amount of the  $^{14}\text{C}$  in Y-791186, Y-792510 and Y-791960. Thus, to be conservative, we quote all three as limit ages for  $^{14}\text{C}$ . If the  $^{14}\text{C}$  data are apparently too young, it is almost certainly due to incomplete removal of the weathering products from these samples. *In situ* production by cosmic rays at the earth's surface of  $^{14}\text{C}$  (JULL *et al.*, 1992) cannot account for an activity of over 1 dpm/kg in a rock exposed at the Yamato site. Previously, we have investigated several meteorites which had both long  $^{81}\text{Kr}$  and  $^{36}\text{Cl}$  ages and no  $^{14}\text{C}$  above what is expected from terrestrial *in situ* of  $^{14}\text{C}$  (JULL *et al.*, 1992), indicating agreement between different radionuclides. There are few discrepancies between  $^{14}\text{C}$  and  $^{36}\text{Cl}$  ages amongst ordinary chondrites (NISHIZUMI *et al.*, 1989). The differences between  $^{14}\text{C}$  and  $^{81}\text{Kr}$  for achondrites warrant further investigation.

The diogenites studied, Y-74037 and Y-74010 are interesting. Y-74037 has a  $^{14}\text{C}$  content twice the values observed in other diogenites from the Yamato 74 collection reported by KIGOSHI and MATSUDA (1985), and an estimated age of  $11.7 \pm 1.3$  kyr. Y-74010 has a terrestrial age of 15 kyr, similar to three other diogenites (Y-74013, Y-74097 and Y-74136) which have terrestrial ages of around 15–19 kyr (KIGOSHI and MATSUDA, 1986; JULL *et al.*, 1984). There is also another determination by BEUKENS *et al.* (1988) on Y-74097 of 9.9 kyr. On the latter basis, the  $^{14}\text{C}$  result for Y-74037 would be consistent with a pairing of Y-74037 and -74097. The other Yamato diogenites are clearly an older age group. The age variations could be ascribed to differences in extraction technique between the much larger samples used by KIGOSHI and MATSUDA (1986) for counting and JULL *et al.* (1984) for both counting and AMS, but this would not explain the measurement for Y-74010. Both these earlier papers reported good extraction yields for other meteorites, but the much better yield of BEUKENS *et al.* (1988) compared to KIGOSHI and MATSUDA (1986) appears to suggest their yield for Y-74097 might be low.

In Fig. 1, we compare the terrestrial-age distribution of the euclrites studied to

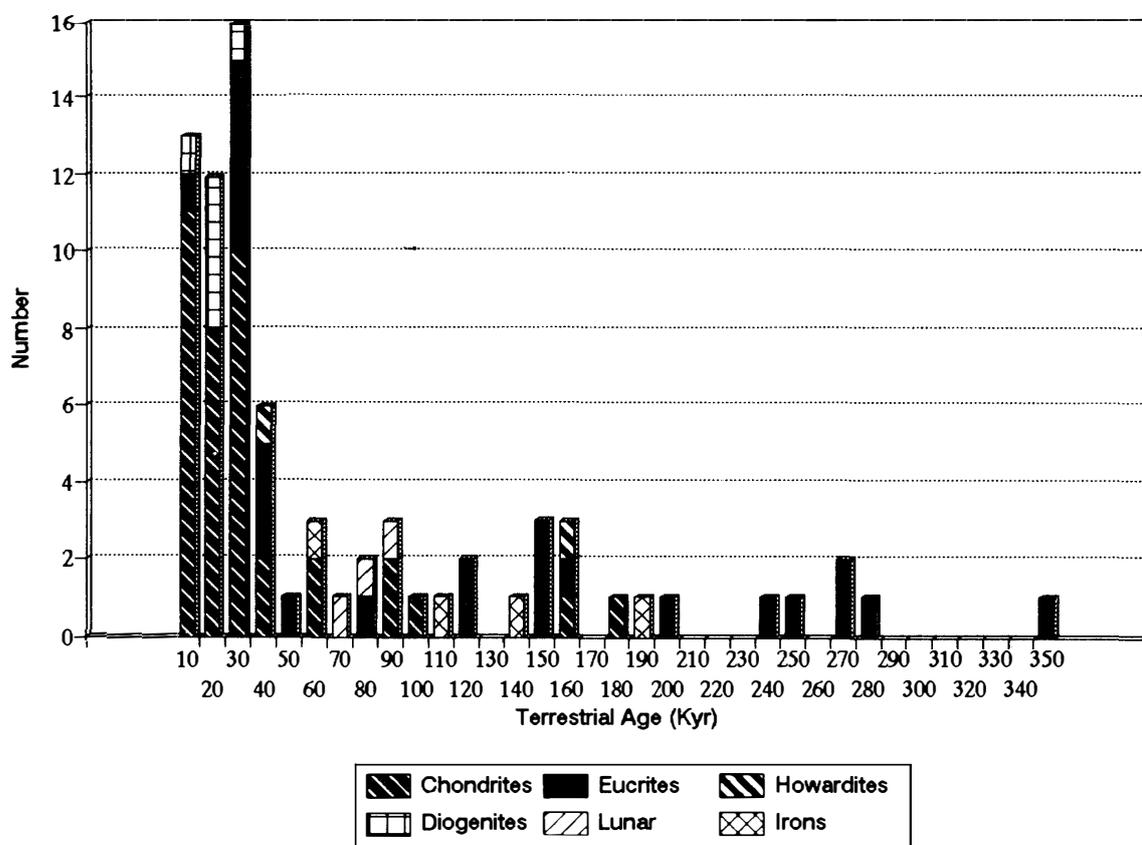


Fig. 1. Histogram of  $^{14}\text{C}$  terrestrial ages of Yamato eucrites compared to the terrestrial age distribution of different classes of Yamato meteorites. The histogram summarizes data of NISHIZUMI (1989), NAGAO and OGATA (1989), MIURA *et al.* (1991), SCHULTZ (1986) and this work.

other Yamato terrestrial ages. In general, the ages of this small group of achondrites from the Yamato site are similar in distribution to the generally young  $^{14}\text{C}$  ages of ordinary chondrites (JULL *et al.*, 1984; KIGOSHI and MATSUDA, 1986; BEUKENS *et al.*, 1988) from this region. The Yamato site in general appears to show fewer meteorites of long terrestrial ages, compared to the Allan Hills Main Icefield (NISHIZUMI, 1989). Figure 1 does point out the discrepancy for  $^{14}\text{C}$  ages *versus*  $^{81}\text{Kr}$  and perhaps  $^{36}\text{Cl}$  for Yamato achondrites which has been alluded to earlier. Some, but not all,  $^{14}\text{C}$  ages of about 30 kyr must be suspect for weathering contamination. However, there is a clear excess of  $^{81}\text{Kr}$  ages of  $>150$  kyr. If the age distribution at the Yamato site obeys the expected exponential distribution (JULL *et al.*, 1993), then most of the *ca.* 30 kyr  $^{14}\text{C}$  ages must reflect the ages of the meteorites. This emphasizes the need for removal of weathering products from older Antarctic meteorite samples.

#### 4. Conclusions

$^{14}\text{C}$  terrestrial-age measurements from several chondrites indicate good agreement with earlier literature values. Studies of  $^{14}\text{C}$  content of six Yamato-79 eucrites

indicate that these meteorites must have consisted of at least three discrete falls, with terrestrial ages of approximately 4, 20 and  $>25$  kyr. The problems of separating weathering products make the distinction of 25 and  $>30$  kyr difficult. The diogenites Y-74037 and Y-74097 have a  $^{14}\text{C}$  age distinct from three other Yamato-74 diogenites (Y-74010, Y-74013 and Y-74136), suggesting more than one diogenite fall in this collection. This is in contrast to petrological descriptions which suggest all are from one fall (TAKEDA *et al.*, 1981). The possibility that these age differences might be due to differences in techniques of several groups needs to be investigated. Possible discrepancies in terrestrial ages between  $^{81}\text{Kr}$  and  $^{14}\text{C}$  in achondrites of  $\sim 30$  kyr terrestrial age indicate that further study of this problem is needed, both in improvement of removal of weathering  $^{14}\text{C}$  contamination, and in cross-correlation of  $^{14}\text{C}$ ,  $^{36}\text{Cl}$  and  $^{81}\text{Kr}$ .

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