COSMOGENIC RADIONUCLIDES IN ORDINARY CHONDRITE FALLS SELECTED FOR CALIBRATION OF THE ^{81}Kr-Kr METHOD. K. C. Welten[1], M. W. Caffee[2], K. Nishiizumi[1], I. Leya[3], N. Dalcher[3], N. Vogel[4] and R. Wieler[4], [1]Space Sciences Laboratory, University of California, Berkeley, CA 94720, USA (kcwelten@ssl.berkeley.edu), [2]Department of Physics, Purdue University, West Lafayette, IN 47907, USA. [3]Space Sciences and Planetology, University of Bern, Switzerland, [4]Department of Earth Sciences, ETH Zürich, CH-8092 Zürich, Switzerland.

Introduction: The ^{81}Kr-Kr exposure age method is based on the combination of stable Kr isotopes, mainly ^{78}Kr, ^{80}Kr, ^{82}Kr, ^{83}Kr, which increase linearly with increasing exposure age, and the radioactive isotope ^{81}Kr (half life = 0.23 Myr), which reaches saturation after 1.5 Myr and thus serves as an internal shielding parameter. The method has mainly been used for eucrites and lunar samples [e.g., 1-3], which have relatively high abundances of the targets elements for Kr production (Rb, Sr, Y, Zr, Nb), but has also been applied to chondrites [e.g. 4-7]. The advantage of the ^{81}Kr-Kr method over other CRE age methods is that a single measurement of the Kr-isotope composition is sufficient to determine the shielding-corrected CRE age of a sample. However, the method depends on accurate knowledge of the ^{81}Kr/^{83}Kr production ratio, which is generally estimated from the shielding sensitive ^{78}Kr/^{83}Kr ratio. The correlation between the two ratios derived from lunar samples [2] works well for average size chondrites [5-7], but has not been tested thoroughly for meteorites of different sizes. A previous calibration of the ^{81}Kr/Kr method for H-chondrites using independent ^{36}Cl-^{36}Ar ages in the metal fraction [8] suffered from large uncertainties in the cosmogenic Kr concentrations due to short CRE ages (4-10 Myr).

In this work, we selected 14 equilibrated H and L-chondrite falls with CRE ages >15 Myr to minimize the amount of trapped Kr relative to the cosmogenic Kr concentrations. The main purpose of this work is to use shielding independent ^{36}Cl-^{36}Ar ages in the metal fraction to calibrate the ^{81}Kr-Kr method. To determine the ^{36}Cl-^{36}Ar ages, we separated the metal fractions and measured cosmogenic ^{36}Cl (half-life =0.301 Myr), while aliquots of the same metal fraction as well as bulk samples were analyzed for noble gases [9]. We also measured concentrations of the radionuclides ^{10}Be (half-life = 1.36 Myr) and ^{26}Al (0.705 Myr) in the metal fraction, as well as ^{10}Be, ^{26}Al and ^{36}Cl concentrations in the stone fraction of the same meteorites. Concentrations of radionuclides were measured by accelerator mass spectrometry (AMS) at PRIME Lab, Purdue University [10] and are given in Table 1. The radionuclide concentrations provide information on the shielding conditions of the selected chondrites during the last few Myr of the CRE and are used to verify that the selected chondrites had simple CRE histories.

We calculated the shielding independent ^{36}Cl-^{36}Ar ages [14]; these are shown in Table 1. We also calculated ^{10}Be-^{21}Ne and ^{26}Al-^{21}Ne ages using the semi-empirical model of Graf [15]. We assumed constant production rate ratios (atom/atom) of 0.14±0.01 for P(^{10}Be)/P(^{21}Ne) and 0.39±0.04 for P(^{26}Al)/P(^{21}Ne). For comparison, Table 1 also shows preliminary ^{81}Kr-Kr ages that were calculated using the correlation of P(^{81}Kr)/P(^{83}Kr) vs. P(^{78}Kr)/P(^{83}Kr) for lunar samples [2].

Results and discussion. For 13 of the 14 meteorites, we report ^{36}Cl-^{36}Ar ages. Most of the ages are in the range between 17 Myr (for Mocs) and 66 Myr (for Ausson), high enough for reliable cosmogenic Kr measurements. The exception is Aumale, which yields a ^{36}Cl-^{36}Ar age of 3.7 Myr. This age is much shorter than the age of ~20 Myr reported previously [16], suggesting that one of the Aumale samples may have been mislabeled. Due to the short CRE age, the cosmogenic Kr concentrations in this sample are very low, making the Aumale sample unsuitable for calibration of the ^{81}Kr-Kr method.

The radionuclide results in the metal phase of Kandahar indicate a CRE age of ~3 Myr. This age is much shorter than the ^{36}Cl-^{36}Ar age of 22 Myr, suggesting that Kandahar may have experienced a complex exposure history. However, this interpretation is not consistent with the radionuclide concentrations in the stone fraction, which are saturated. In addition, the ^{10}Be/^{21}Ne and ^{26}Al/^{21}Ne ages agree within 10-20% with the ^{36}Cl-^{36}Ar age of Kandahar.

The cosmogenic radionuclide concentrations in Hessle, Mount Browne, St. Germain du Pinel, Alfianello, Ausson, Bruderheim, Harleton, Mbale, Mocs, and Peace River are in the range of average saturation values for small to medium sized objects [17]. The ^{22}Ne/^{21}Ne and ^{78}Kr/^{83}Kr ratios in 9 of these 10 meteorites are consistent with irradiation in small to medium sized objects, suggesting they experienced simple exposure histories. The only exception is Harleton, for which the ^{22}Ne/^{21}Ne and ^{78}Kr/^{83}Kr ratios data suggest much higher shielding than the radionuclide results. We conclude that Harleton most likely experienced a two-stage exposure history, with a first stage at high shielding in which most of the cosmogenic noble gas inventory was produced and a second stage in an object less than 30 cm in radius, in which the cosmogenic radionuclides were produced.

The shielding corrected ^{10}Be/^{21}Ne and ^{26}Al/^{21}Ne ages of Harleton are ~50% higher than the ^{36}Cl-^{36}Ar age of 36 Myr, providing additional evidence of a complex exposure history, which makes Harleton unsuitable for calibration of the ^{81}Kr-Kr method.

The radionuclide concentrations in two meteorites, Richardton and La Criolla, show evidence of high shielding conditions. These meteorites have low ^{36}Cl concentrations (<20 dpm/kg) in the metal fraction combined with high ^{36}Cl concentrations (14-17 dpm/kg) in the stone fraction that indicate production of ^{36}Cl by neutron-capture on ^{35}Cl. The large neutron-capture ^{36}Cl contributions in La Criolla (~8 dpm/kg) and Richardton (12 dpm/kg) are also consistent with the elevated ^{80}Kr/^{83}Kr and ^{82}Kr/^{83}Kr ratios in these two samples, which indicate contributions of ^{80}Kr and ^{82}Kr from neutron-capture reactions on ^{79}Br and ^{81}Br. Based on the presence of neutron-capture products and the low ^{22}Ne/^{21}Ne ratios of Richardton and La Criolla, we conclude that these meteorites had a radius >50 cm throughout their exposure history. Although this seems to suggest that these two meteorites had simple exposure histories, their ^{10}Be/^{21}Ne and ^{26}Al/^{21}Ne ages are significantly higher than the ^{36}Cl-^{36}Ar ages. It is not clear if this is due to a change in shielding conditions or to variations in the ^{10}Be/^{21}Ne and ^{26}Al/^{21}Ne production ratios as a function of shielding. The high ^{26}Al/^{10}Be ratio (3.9 ± 0.2) in Richardton may indicate a recent change in shielding conditions. Measurements of ^{10}Be and ^{26}Al in the metal phase of Richardton are in progress to verify its exposure history.

The preliminary ^{81}Kr-Kr ages, based on the correlation of P(^{81}Kr)/P(^{83}Kr) vs. P(^{78}Kr)/P(^{83}Kr) for lunar samples, show good agreement with the ^{36}Cl-^{36}Ar ages for some chondrites, but show significant discrepancies for other chondrites. The largest discrepancy between the two ages is found for Harleton, which is clearly due to the complex exposure history of this meteorite. However, the discrepancies found for some of the other chondrites with simple exposure ages suggest that the shielding dependence of the P(^{81}Kr)/P(^{83}Kr) ratio derived from lunar samples is not necessarily valid for chondrites.

Conclusions: Based on the cosmogenic radionuclide concentrations in combination with noble gases, we conclude that 10 of the selected 14 chondrites have one-stage CRE ages of >15 Myr, suitable for the calibration of the ^{81}Kr-Kr method. At least two of the selected chondrites are unsuitable, either because of a short CRE age (Aumale) or a two-stage exposure history (Harleton). These meteorites will be excluded from the ^{81}K-Kr calibration study. La Criolla and Richardton experienced high shielding throughout their exposure history, which makes these two meteorites good candidates to test the ^{81}Kr-Kr exposure age method for different shielding conditions.

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References: [1] Marti K. (1967) Phys. Rev. Lett. 18, 264-266. [2] Marti K. and Lugmair G. (1971) LPSC 2, 1591-1605. [3] Shukolyukov A. and Begemann F. (1996) MAPS 31, 60-72. [4] Eugster O. et al. (1967) EPSL 2, 77-82. [5] Finkel R. C. et al. (1978) GCA 42, 241-250. [6] Eugster O. (1988) GCA 52, 1649-1662. [7] Lavielle B. et al. (1997) MAPS 32, 97-107. [8] Leya I. et al. (2004) Ant. Met. Res. 17, 185-199. [9] Dalcher N. et al. (2010) MAPS 45, A41. [10] Sharma P. et al. (2000) NIM B172, 112-123. [11] Nishiizumi K. et al. (2007) NIM B258, 403-413. [12] Nishiizumi K. (2004) NIM B223-224, 388-392. [13] Sharma P. et al. (1990) NIM B52, 410-415. [14] Lavielle B. et al. (1999) EPSL 170, 93-104. [15] Graf T. et al. (1990) GCA 54, 2521-2534. [16] Mueller H. and Zahringer J. (1969) In: Meteorite Research, Reidel Publ., pp. 940. [17] Leya I. and Masarik J. (2009) MAPS 44, 1061-1086.

Table 1. Cosmogenic ^{22}Ne/^{21}Ne and ^{78}Kr/^{83}Kr ratios and preliminary ^{81}Kr-Kr ages (T81, in Myr) for bulk ordinary chondrites, as well as ^{36}Cl-^{36}Ar ages (T36, in Myr) for the metal fraction, and cosmogenic radionuclide concentrations* (in dpm/kg) in metal and stone fractions.

Meteorite	Type	^{22}Ne/^{21}Ne	^{78}Kr/^{83}Kr	T81	T36	^{10}Be(m)	^{26}Al(m)	^{36}Cl(m)	^{10}Be(s)	^{26}Al(s)	^{36}Cl(s)
Hessle	H5	1.14±0.02	0.166±0.010	72±3	61±3	4.64 ± 0.15	4.21 ± 0.15	24.5 ± 0.6	24.8 ± 0.7	70.8 ± 1.5	7.6 ± 0.3
Mt. Browne	H6	1.11±0.02	0.155±0.017	37±3	25±1	5.66 ± 0.16	4.33 ± 0.28	24.0 ± 0.5	24.7 ± 0.7	70.0 ± 1.6	7.7 ± 0.3
St. Germain	H6	1.14±0.02	0.185±0.018	45±2	-	-	-	-	19.7 ± 0.4	55.5 ± 1.3	6.4 ± 0.3
Richardton	H6	1.08±0.02	0.152±0.040	24±2	25±2	-	-	15.5 ± 0.3	16.6 ± 0.3	64.0 ± 1.4	16.9 ± 0.6
Alfianello	L6	1.13±0.02	0.147±0.006	34±1	25±2	5.49 ± 0.12	4.01 ± 0.15	24.4 ± 0.5	24.0 ± 0.5	68.5 ± 1.6	8.4 ± 0.2
Aumale	L6	1.07±0.06	(0.13±0.11)	-	3.7±0.2	2.79 ± 0.08	2.86 ± 0.08	21.0 ± 0.6	17.3 ± 0.3	69.3 ± 1.7	11.6 ± 0.3
Ausson	L5	1.13±0.02	0.157±0.025	76±3	66±4	5.48 ± 0.16	4.16 ± 0.14	22.6 ± 0.7	23.6 ± 0.5	68.5 ± 1.4	8.8 ± 0.2
Bruderheim	L5	1.10±0.02	0.149±0.015	33±2	25±2	4.58 ± 0.17	3.98 ± 0.14	22.3 ± 0.5	23.1 ± 0.5	69.1 ± 1.9	8.7 ± 0.2
Harleton	L6	1.06±0.02	0.127±0.004	62±2	36±2	5.10 ± 0.16	4.30 ± 0.18	23.4 ± 0.7	20.6 ± 0.4	59.3 ± 1.7	6.5 ± 0.3
Kandahar	L5	1.09±0.02	0.140±0.010	29±1	22±2	3.86 ± 0.14	3.49 ± 0.17	23.1 ± 0.9	25.4 ± 0.5	74.9 ± 2.1	9.1 ± 0.4
La Criolla	L6	1.09±0.02	0.119±0.007	34±1	24±2	3.40 ± 0.10	2.47 ± 0.16	18.4 ± 0.5	20.8 ± 0.5	69.7 ± 1.9	14.6 ± 0.3
Mbale	L5/6	1.10±0.02	0.136±0.009	31±1	26±2	4.95 ± 0.10	3.61 ± 0.24	23.1 ± 0.6	23.0 ± 0.5	65.6 ± 1.9	8.5 ± 0.2
Mocs	L6	1.11±0.02	0.150±0.015	19±1	17±1	4.89 ± 0.13	3.3 ± 0.3	21.4 ± 0.5	23.5 ± 0.5	74.9 ± 2.4	10.6 ± 0.3
Peace River	L6	1.13±0.02	0.152±0.016	32±1	30±2	5.06 ± 0.13	3.68 ± 0.21	23.0 ± 0.7	22.6 ± 0.5	66.9 ± 7.5	8.2 ± 0.2

*Radionuclide concentrations were normalized to ^{10}Be, ^{26}Al, and ^{36}Cl AMS standards [11-13].