

The Noble Gas Record in Antarctic and Other Meteorites

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Dedicated to Professor Alfred Klemm on the occasion of his 70th birthday

Data are reported for the concentration and isotopic composition of He, Ne, and Ar in 11 Antarctic and 8 other stone meteorites. Cosmic ray exposure ages and whole rock gas retention model ages are given. The noble gas record suggests that all three ALLAN HILLS eucrites analysed so far belong to the same meteorite fall while the three eucrites from the ELEPHANT MORaine area appear to be three independent individual falls.

Introduction

Noble gases in stone meteorites are a mixture of different components which contain information pertaining to different events in the history of the meteorites. Radiogenic nuclides, like ^4He and ^{40}Ar , allow us to deduce gas retention ages, while from the spallogenic nuclides so-called exposure ages can be calculated which give the time during which the meteorites have been exposed to the cosmic radiation. Primordial noble gases, introduced in the early stage of meteoritic matter, and solar-trapped gases give information about the early environment and on surface processes on the parent bodies of the respective meteorites.

The discovery of about 6000 new meteorites in Antarctica [1] has increased the world inventory by more than a factor of three. This alone, as well as the fact that some of these meteorites fell almost a million years ago [2], makes it appear possible that hitherto unknown types of meteorites may be discovered among these deep-frozen extraterrestrial rocks. Furthermore, a number of unusually small meteorites, with linear dimensions of a centimeter or less, have been retrieved which open up the possibility to study the interaction with meteoritic matter of low-energy solar cosmic rays, although the possibility must be kept in mind that the small pieces recovered may be fragments of much larger ones.

We have analysed 11 meteorites from the Antarctic, mainly to study their irradiation history. In addition, we report new data for seven other stone meteorites and the results of a re-determination of the noble gases in Ambapur Nagla [3, 4].

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Experimental procedure and results

The meteorites investigated are listed in Table 1, together with their classification and other relevant information.

Samples of 100–200 mg, whenever possible as chunks, were wrapped in Ni foil (~20 mg) and

Table 1. Meteorites investigated.

1. Allan Hills 77278	LL3	Find	UNA
2. Allan Hills 77299	H3	Find	UNA
3. Allan Hills 78132	Eu	Find	JSC
4. Allan Hills 79017	Eu	Find	JSC
5. Ambapur Nagla	H5	5-27-1895	MNH
6. Bouvante-le-Haut	Eu	7-30-1978	UP
7. Chitenay	L6	2-21-1978	UP
8. Elephant Moraine 79004	Eu	Find	JSC
9. Elephant Moraine 79005	Eu	Find	JSC
10. Elephant Moraine 79006	Eu (How)	Find	JSC
11. Guenie	H4	4-1960	UP
12. Junan ^a	L6	5-15-1977	C
13. Waconda	L6	Find	MPIM
14. Yamato 74116	L	Find	NIPR
15. Yamato 74192	H6	Find	NIPR
16. Yamato 74418	H6	Find	NIPR
17. Yamato 74663	L6	Find	NIPR
18. Xingyang	H5	12-1-1977	C
19. Ybbsitz	H4	Find	NMW
20. Bruderheim	L6	3-4-1960	UCB
(Berkeley Standard)			
C:	Institute of Geochemistry Guiyang, Academia Sinica (Prof. Ouyang)		
JSC:	Johnson Space Center (NASA), Houston, Texas		
MNH:	Museum National d'Histoire Naturelle, Paris (Dr. P. Pellas)		
MPIM:	Max-Planck-Institut für Chemie, Mainz (Paneth-Collection)		
NIPR:	National Institute of Polar Research, Tokyo (Dr. K. Yanai)		
NMW:	Naturhistorisches Museum, Wien (Dr. G. Kurat)		
UCB:	University of California, Berkeley (Dr. J. H. Reynolds)		
UNA:	University of New Mexico, Albuquerque (Dr. E. Scott)		
UP:	University of Paris (Drs. M. Christophe Michel-Levy and J.-C. Lorin)		

^a The meteorite was made available under the name "Lounan".

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stored in the sample holder of the extraction system. They were pre-heated at 60 °C for 1 day and then degassed in one step in a Ta oven at a temperature of about 1700 °C. The gases were cleaned over hot Ti sponge and a SAES getter. Argon, krypton, and xenon were adsorbed on activated charcoal at the temperature of boiling nitrogen, while He and Ne were admitted into the all-metal 60° mass spectrometer. After analysis, He and Ne were pumped off, Ar was desorbed at ~ 300 °C and measured.

The mass fractionation (13.3%, 2.5%, and 1.8% per atomic mass unit for He, Ne, and Ar, respectively) and the sensitivity of the spectrometer were determined by measuring mixtures of atmospheric Ne

and Ar; in the case of He an artificial mixture with $^3\text{He}/^4\text{He} = 0.98$ was used.

Total extraction blanks (in units of cm^3 STP) were typically 5×10^{-11} , 1×10^{-9} , 1×10^{-10} , and 1×10^{-8} for ^3He , ^4He , ^{20}Ne , and ^{40}Ar , respectively.

The results are compiled in Table 2. Listed in Table 3 are some pertinent abundance ratios and the amounts of spallogenic ^{21}Ne and ^{38}Ar . They were calculated by assuming the spallogenic $^{20}\text{Ne}/^{21}\text{Ne}$ ratio to be 0.84 and any excess ^{20}Ne to be due to a neon component with the isotopic composition of atmospheric neon. Similarly, spallogenic ^{38}Ar has been derived with an abundance ratio in a trapped or adsorbed component of $^{36}\text{Ar}/^{38}\text{Ar} = 5.32$

Table 2. The gas retention ages in the last two columns are model ages based on the following assumed radionuclide concentrations. U: 11 ppb and 130 ppb in chondrites and eucrites, respectively; Th/U = 3.6; K: 830 ppm, 860 ppm, 830 ppm, and 400 ppm for H-, L-, LL-chondrites and eucrites, respectively [29]. Spallogenic ^4He has been removed by subtracting 5 times the amount of ^3He .

	³ He	⁴ He	²⁰ Ne	²¹ Ne	²² Ne	³⁶ Ar	³⁸ Ar	⁴⁰ Ar	U, Th- ⁴ He	K-Ar	Lit.
	in 10 ⁻⁸ cm ³ STP/g								in 10 ⁹ years		
<i>H-chondrites</i>											
ALH 77299	55.9	1710	10.2	9.67	11.2	27.7	6.65	6320	3.95	4.50	
Ambapur Nagla	7.13	172	3.89	3.89	4.28	1.98	0.82	2710	0.55	3.15	
	7.9	125	4.0	3.4	3.9	1.4	0.98	1850	—	—	[3]
	5.8	150	3.5	2.98	3.23	—	—	—	—	—	[4]
Guenie ^a	2.0	1250	0.90	0.67	0.74	1.22	0.37	5450	3.60	4.20	
Xingyang	29.3	1640	4.46	4.63	5.48	1.15	0.82	6050	4.05	4.40	
Y 74192	57.0	1700	11.3	12.5	13.7	1.96	1.80	5070	3.90	4.10	
Y 74418	11.1	1140	1.65	1.72	2.03	0.65	0.42	5300	3.30	4.20	
	17.6	1230	1.89	2.36	2.79	0.88	0.55	5990	3.2	4.55	[6]
Ybbsitz	1.63	910	0.61	0.61	0.66	2.04	0.46	3650	2.90	3.60	
<i>L-chondrites</i>											
Bruderheim	47.8	480	8.88	9.66	10.5	1.32	1.46	1190	0.90	2.00	
(Berkeley Standard)											
Chitenay ^a	12.9	593	2.96	3.24	3.52	2.1	0.71	5340	1.90	4.15	
Junan	64.9	890	10.8	11.5	13.1	1.73	1.78	6640	1.95	4.50	
Wacanda dark	25.9	1360	5.52	6.15	6.70	1.29	0.97	6000	3.55	4.30	
Wacanda light	28.0	1300	6.03	6.67	7.21	1.29	0.98	5940	3.35	4.30	
Y 74116	6.3	460	2.26	2.50	2.68	0.73	0.38	5300	1.60	4.10	
	11	820	3.1	3.3	3.6	0.9	0.9	3600	2.0	4.34	[6]
Y 74663	23.2	645	2.97	2.94	3.73	0.83	0.60	5350	1.90	4.15	
<i>LL-chondrite</i>											
ALH 77278	44.2	3040	30.2	6.38	9.69	21.0	4.88	6020	— ^b	4.40	
<i>Eucrites</i>											
ALH 76005 ^a	11.7	1380	1.87	2.07	2.40	1.56	2.00	1735	—	—	[7]
	9.1	1150	1.62	1.71	1.98	1.22	1.66	1400	—	—	[2]
ALH 78132	13.5	1350	2.58	2.31	2.66	2.19	2.23	1500	0.40	3.35	
ALH 79017	13.6	1770	3.25	2.38	2.84	3.26	3.37	2420	0.55	4.10	
Bouvante ^a	7.42	7590	1.41	1.46	1.78	0.71	1.02	2700	2.15	4.30	
EET 79004	16.1	4140	5.10	3.76	4.62	5.08	3.14	2020	1.25	3.80	
EET 79005	24.5	3400	4.55	4.49	5.32	2.97	3.52	1480	1.05	3.30	
EET 79006	14.5	1780	4.27	4.18	4.96	2.83	3.40	1790	0.55	3.60	

^aMean of two measurements. ^bContains trapped ^4He .

Table 3. Spallogenic gases and cosmic ray exposure ages.

	³ He	²¹ Ne	³⁸ Ar	³ He/ ²¹ Ne	²² Ne/ ²¹ Ne	T ₃	T ₂₁	T ₃₈
	in 10 ⁻⁸ cm ³ STP/g					in 10 ⁶ years		
<i>H-chondrites</i>								
ALH 77299	55.9	9.67	1.64	5.78	1.127	23.8	23.1	24.0
Ambapur Nagla	7.13	3.89	0.51	1.83	1.085	2.9	7.7	5.9
Guenie	2.0	0.67	0.16	3.0	1.10	0.8	1.4	1.9
Xingyang	29.3	4.63	0.69	6.33	1.171	13.3	12.5	12.0
Y 74192	57.0	12.5	1.63	4.56	1.088	23.1	25.0	19.3
Y 74418	11.1	1.72	0.34	6.45	1.169	5.0	4.6	5.9
Ybbsitz	1.63	0.61	0.09	2.66	1.065	0.6	1.2	0.9
<i>L-chondrites</i>								
Bruderheim	47.8	9.66	1.38	4.95	1.081	18.8	17.5	16.0
Chitenay	12.9	3.24	0.36	3.98	1.077	5.1	5.8	4.0
Junan	64.9	11.5	1.66	5.64	1.130	27.2	26.1	24.7
Waconda D	25.9	6.15	0.83	4.21	1.089	10.3	11.6	9.3
Waconda L	28.0	6.67	0.84	4.20	1.081	11.0	12.0	9.6
Y 74116	6.3	2.50	0.28	2.52	1.064	2.4	4.5	3.2
Y 74663	23.2	2.94	0.51	7.89	1.252	11.7	7.6	9.5
<i>LL-chondrite</i>								
ALH 77278	44	6.31	1.06	6.97	(1.12) ^a	20	16	20
<i>Eucrites</i>								
ALH 76005 ^b	11.7	2.07	1.90	5.65	1.159	4.4	7.1	8.9
ALH 78132	13.5	2.31	2.07	5.84	1.152	5.4	8.2	9.8
ALH 79017	13.6	2.38	2.00	5.71	1.177	5.2	8.5	9.0
EET 79004	16.1	3.76	2.49	4.28	1.178	6.4	13.0	12.8
EET 79005	24.5	4.49	3.37	5.46	1.183	9.7	15.2	17.5
EET 79006	14.5	4.18	3.27	3.47	1.184	5.8	14.7	14.7
Bouvante	7.42	1.46	1.01	5.08	1.121	2.9	5.6	4.4

^a This value is presumably too small since the corrections for trapped neon were made assuming the isotopic composition of atmospheric neon.

^b For reasons of consistency Mainz data only [7].

D, L: Dark and light phase, respectively.

and a spallogenic ratio of 0.65. Although the latter value may be too high for meteorites with short cosmic ray exposure ages [5] (Guenie, Ybbsitz) this does not affect the amount of spallogenic ^{38}Ar by more than 5% at most.

Included in Tables 2 and 3 are data for the Berkeley Bruderheim Standard as well as published results for Ambapur Nagla [3, 4], Y 74116 [6], Y 74418 [6], and ALH 76005 [2, 7]. For the spallogenic nuclides ^3He , ^{21}Ne , and ^{38}Ar in Bruderheim the results agree with our 1978 measurement [8] to within 1%. They, furthermore, agree to within 5% with most of the results reported from other laboratories [4]. For Ambapur Nagla the results are also in fair agreement with those reported before. Less satisfactory is the rather serious disagreement with the data of Takaoka *et al.* [6] which can only in part be explained by the fact that the Bruderheim results

of these authors are outside the range reported by other laboratories as well.

Discussion

Chondrites

Cosmic ray exposure ages have been calculated with the shielding-corrected ^3He and ^{21}Ne production rates of Cressy and Bogard [9], making the usual assumption of a single-stage exposure history. Our ^{38}Ar production rates for H-, L-, and LL-chondrites are 3%, 7.5% and 8% higher, respectively, than those of Cressy and Bogard (see Table 4); for the shielding correction we have adopted their procedure without modification.

The exposure ages cover the normal range encountered for stone meteorites. Note, that both H4-

Table 4. The chemical composition (weight %) used to calculate the production rate of ^{38}Ar in units of $10^{-10} \text{ cm}^3 \text{ STP/g} \cdot \text{Ma}$.

	Ca	Fe ^a	K	Ti + Cr + Mn	P (^{38}Ar)	Lit.
ALH 76005	6.81	14.6	0.05	1.18	21.3	[32, 33, 34] ^b
ALH 78132	6.82	13.8	0.04	1.05	21.1	[32, 33] ^b
ALH 79017	7.23	14.6	0.04	1.0	22.2	^c
Bouvante	7.34	15.4	0.05	1.1	22.8	[30]
EET 79004	6.05	14.0	0.07	1.0 ^c	19.5	[30]
EET 79005	6.2	13.5	0.04	1.0 ^c	19.3	[30]
EET 79006	7.23	14.6	0.04	1.1	22.2	^c
H-chondrites	1.19	29.3	0.080	0.63	7.54	^c
L-chondrites	1.28	23.1	0.087	0.69	7.41	^c
LL-chondrites	1.25	20.9	0.091	0.71	7.22	^c

^a For H-, L-, and LL-chondrites the entries include Ni.^b Mean values are listed.^c Average chemical composition assumed [31].

chondrites (Guenie and Ybbsitz) have very short exposure ages. They fall onto the short-age peak in the number frequency distribution histogram (cf. e.g. [10]) and tend to make this peak statistically more significant and, thus, the exposure age distribution of H4-chondrites distinct from that of H-chondrites of higher petrological type.

It is not quite clear whether Guenie blemishes the beauty of the statistics of Wood [11] in that H-chondrites with exposure ages shorter than 2 Ma "all fell during the 6 month period of October to March". Guenie fell during April 1960 [12]; perhaps it was April 1.

For Ambapur Nagla our results confirm that the ^3He exposure age is short compared to those derived for the same samples from ^{21}Ne and ^{38}Ar [3, 4] indicating losses by diffusion of ^3He . Since Ambapur Nagla is one of the rare meteorites where fission tracks are not even observed in the pyroxenes adjacent to uranium-rich phosphates [13] it is tempting to ascribe the extinction of the tracks and the loss of spallogenic ^3He to the same thermal event. This is at variance, however, with the presence of cosmic ray tracks in the pyroxene. Rather, the extinction of the fission tracks must have happened before, or at the time of, the onset of the exposure of the meteorite to the cosmic radiation.

Similarly low in the relative ^3He exposure ages are Yamato 74116, Ybbsitz, and Guenie. For the latter this is surprising in so far as the ^4He -U/Th gas retention age is fairly high; apparently the percentage loss of radiogenic ^4He is smaller than that of cosmogenic ^3He .

Yamato 74663 is peculiar in that its ^3He and ^{38}Ar exposure ages agree within the limits of error, but that the ^{21}Ne age is lower by 30%. Among the suite of chondrites analysed, this meteorite has by far the highest spallogenic $^{22}\text{Ne}/^{21}\text{Ne}$ ratio which necessitates the largest shielding correction of the ^{21}Ne production rate. It is conceivable that the correction applied was not sufficient. The same explanation may hold for the discrepancy between the noble gas exposure ages and that of 14 Ma [14] derived from spallogenic ^{40}K produced in the metal phase. On the other hand, the ^{40}K exposure ages of all four Antarctic meteorites from the Yamato Mts. are distinctly higher – by up to a factor of two (Y74116) – than the noble gas derived exposure ages [14], so that there may well be a systematic difference. If this should be the case the reason is not clear at present. It cannot be due to an early irradiation since this would make the ^{40}K exposure ages shorter than the ones deduced from stable nuclides like ^3He , ^{21}Ne , or ^{38}Ar .

The reason for selecting for noble gas analysis the four Yamato chondrites was their ^{53}Mn content [15]. Y 74192, with an activity of 578 dpm/kg metal is one of the two with the highest decay rates, while Y 74418 (275 dpm/kg) and Y 74663 (255 dpm/kg) are intermediate, and Y 74116 with 63 dpm/kg is on the extremely low side. With the exposure ages from Table 3 the ^{53}Mn saturation activities would be 578, 460, 300, and 125 dpm/kg, respectively, assuming that the decay since the time of fall is negligible (terrestrial age \ll mean life time of ^{53}Mn of about 5 Ma). The first three activities are in the

range expected for small to medium-size objects [16, 17]. For Y 74116, however, a rather large meteoroid with a pre-atmospheric radius of about 2 m or more is required. Although this would be in accord with the low spallogenic $^{22}\text{Ne}/^{21}\text{Ne}$ ratio of 1.064 the possibility should be kept in mind that the meteorite may have had a complex exposure history. To us this seems to be the more probable explanation.

The two type 3 chondrites – ALH77278 and ALH 77299 – are both outstanding in their high content of trapped noble gases. Nevertheless, there is a distinct difference: in the H3 ALH 77299 the ratio $^{20}\text{Ne}/^{36}\text{Ar}$ in the trapped gases is more than ten times lower than that in the LL3 ALH 77278. The latter actually shows a significant excess of ^4He of $\gtrsim 1000 \times 10^{-8} \text{ cm}^3 \text{ STP/g}$ as well which is indicative of solar wind implanted gases. Waconda, on the other hand, although light-dark structured shows no evidence for solar wind implanted gases in either phase.

Eucrites

The different chemical composition of eucrites and chondrites is reflected in their noble gases:

- 1) Eucrites contain more radiogenic ^4He and less ^{40}Ar because they contain more U, Th and less K than chondrites (see legend to Table 2).
- 2) The ratio of spallogenic $^{38}\text{Ar}/^{21}\text{Ne}$ is approximately 5 times higher in eucrites because their Ca content is much higher and their Mg content is much lower than that of chondrites.
- 3) The ratio of spallogenic $^{22}\text{Ne}/^{21}\text{Ne}$ in eucrites is high as well. It is important to note that this is essentially due to a low Mg content; it does not necessarily reflect the hardness of the irradiation.

Until 1980 eight eucrites had been found at the Allan Hills site and six more in the Reckling Peak Moraine/Elephant Moraine area. It is not immediately apparent whether all these individual specimens represent different falls or, if not, how many of them belong to the same fall(s). Since meteorites from paired or multiple falls must have the same cosmic ray exposure age and, to a lesser degree, must contain the same amounts of radiogenic ^4He and ^{40}Ar it is of interest to compare these quantities.

In the case of eucrites this poses some problems, however, since, as a rule, the exposure ages calculated from the concentrations of ^3He , ^{21}Ne , and ^{38}Ar do not agree [18]. Although diffusive losses of

^3He and of ^{21}Ne are a possible explanation for the shorter ^3He and ^{21}Ne exposure ages, we are, nevertheless, left with the fact that, for a given meteorite, only a ^{38}Ar exposure age can be calculated. Consequently, it is not possible to check the data for internal consistency.

The ^{38}Ar production rate has been calculated for each meteorite from its chemical composition (Table 4) using

$$P(^{38}\text{Ar}) = 2.75 [\text{Ca}] + 0.086 [\text{Fe}] \\ + 0.33 [\text{Ti, Cr, Mn}] + 19.2 [\text{K}].$$

The element-specific production rates (in units of $10^{-8} \text{ cm}^3 \text{ STP/g} \cdot \text{Ma}$) are different from those of [18] and [9]. They are based on the following experimental data:

- 1) The average decay rate of ^{36}Cl in iron meteorites and the metal phase from stone meteorites and stony-iron meteorites of sufficiently short terrestrial age is 23 disintegrations/min · kg (cf. e.g. [19, 20]). This together with $P(^{36}\text{Cl})/P(^{36}\text{Cl}) + P(^{36}\text{Ar}) = 0.82$ [21] and $^{36}\text{Ar}/^{38}\text{Ar} = 0.63$ yields a production rate for FeNi of $8.6 \times 10^{-10} \text{ cm}^3 \text{ STP/g} \cdot \text{Ma}$. The different target composition in eucrites (almost no Ni) is not expected to affect this rate to any significant degree.
- 2) The production rate ratio $P(^{38}\text{Ar})_{\text{Ca}}/P(^{38}\text{Ar})_{\text{FeNi}}$ in *individual* stone meteorites and mesosiderites varies between 17.3 and 48 [20, 22, 23] while pooling the data from *different* meteorites yielded a value of 16.5 [24]. We assume the ratio to be independent of the bulk chemical composition of the meteorites and adopt a mean of 32, with an accuracy of a factor of 1.5 either way.
- 3) For K and Ti, Cr, Mn there exist no experimental data. We have adopted the ratios $P(^{38}\text{Ar})_{\text{K}}/P(^{38}\text{Ar})_{\text{Ca}} = 7.0$ and $P(^{38}\text{Ar})_{\text{Ti, Cr, Mn}}/P(^{38}\text{Ar})_{\text{FeNi}} = 3.8$ given by [9]. Since the combined production from these elements amounts to less than 10% of the total the exact values are not critical.

Since, for eucrites, the $^3\text{He}/^{21}\text{Ne}$ vs. $^{22}\text{Ne}/^{21}\text{Ne}$ correlation systematics is not sufficiently clear, no shielding-corrected production rates can be derived from the measured $^{22}\text{Ne}/^{21}\text{Ne}$ ratios. This is unfortunate because it leaves us with the large uncertainty in the production from Ca which, for eucrites, is the most important target element by far. (For normal chondrites Ca is less important. Consequently, for H- and L-chondrites the Ca-related

uncertainty in the production rate of ^{38}Ar is only about 15% or so.)

The noble gas evidence supports the suggestion [25], based on the mineralogy, that all three Allan Hills eucrites analysed so far may belong to the same meteorite fall: Their contents of spallation-produced noble gas nuclides agree to within $\pm 10\%$ or better, and the somewhat larger variation in radiogenic ^4He and ^{40}Ar can easily be accounted for by sample heterogeneity and/or by different losses due to diffusion. After all, there is unambiguous evidence now that different samples from definitely the same meteorite may vary in their radiogenic ^4He and ^{40}Ar by factors of 5 and 3, respectively [26].

The three specimens found in the Elephant Moraine area are again rather similar in their contents of spallogenic noble gases. The differences

which do exist are of the same order as have been found in St. Severin [27] and Keys [28]; they do not preclude a common origin from the same preatmospheric mass. The close agreement between the $^{22}\text{Ne}/^{21}\text{Ne}$ ratios argues against such an explanation, however. The differences required in the ^{21}Ne and ^{38}Ar production rates to make the exposure ages equal should be accompanied by variations of the $^{22}\text{Ne}/^{21}\text{Ne}$ ratio of the order of 5% or so [26, 27]. This is not observed.

Another, perhaps weaker argument, can be made from the radiogenic nuclides. The pattern observed in 79005 cannot be derived by diffusion losses from that of 79004 nor, for that matter, from that of 79006. Thus, 79005 cannot be a paired fall with either of the other two. For the latter two, on the other hand, the radiogenic gases are not of much use in settling the problem.

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