Nitrogen in stone meteorites and terrestrial standards

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Nitrogen contents have been determined by neutron activation method in stone meteorites and silicate standards. Leaching and sieving experiments on standards indicate that contamination from atmospheric nitrogen is negligible. For BCR-1 nitrogen contents show a range from 15 to 62ppm. The study includes 32 chondrites belonging to various classes and six achondrites. Nitrogen is high in C1 and C2 chondrites (500 to 1,000ppm) and in E chondrites (100 - 500ppm). C3 group shows variable amounts of nitrogen. Ordinary chondrites have similar nitrogen contents (~30ppm) irrespective of their petrologic group. For all classes of meteorites, chondrules have lower nitrogen compared to the matrix. Dark phases of some gas-rich meteorites are enriched in nitrogen as compared to their light counterparts. Non-magnetic portions have slightly excess N compared to the magnetic portions. Correlation of N with C, In, Bi, Tl, Zn and Cd indicates the presence of two host phases for N.

INTRODUCTION

Nitrogen, one of the most abundant elements in the solar nebula has accreted only to a small extent in meteorites (GIBSON and MOORE, 1971a, b; MOORE, 1971; GIBSON et al., 1971a; KOTHARI and GOEL, 1974; SHUKLA and GOEL, 1981; KUNG and CLAYTON, 1978). Even in C1 and C2 chondrites the C/N ratio is 30 to 50 compared to the solar value of 8. If these elements condensed in near cosmic proportions a mechanism of losing N in a secondary process is needed. However, most likely N is only partially accreted, as compared to carbon (GEISS and BOCHSLER, 1982). Recent studies of N and its isotopic composition in the Allende meteorite have clearly identified at least two components for nitrogen (THIEMENS and CLAYTON, 1981; FRICK and PEPIN, 1981). The association of the light nitrogen component with the deuterium-rich carbonaceous matter (KERRIDGE, 1982; ROBERT and EPSTEIN, 1982; BECKER and EPSTEIN, 1982) and the recent finding of YANG and EPSTEIN (1982) that such a carbonaceous phase is ubiquitous in all chondrites, suggest that, in general, N in chondrites might be a two component mixture.

Earlier data on N have been obtained by classical methods like the Kjeldahl distillation (MÜLLER, 1974; MÜLLER et al., 1976), carrier-gas fusion (GIBSON and MOORE, 1970; hereafter referred to as the Arizona Group) and the vacuum pyrolysis (KUNG and CLAYTON, 1978; hereafter referred to as the Chicago Group) which suffer from either of the twin problems of incomplete extraction (Kjeldahl distillation and vacuum pyrolysis) and atmospheric contamination (carrier-gas fusion). The neutron activation method used in the present work (SHUKLA et al., 1978) is expected to minimize the above problems. However, it is instructive to compare the N data obtained on homogenized samples by various techniques and rationalise the discrepancies. Recently NORRIS and SCHAEFFER (1982) did such an excercise and concluded that the discrepancies in the N contents obtained by various methods on terrestrial standards are partly due to atmospheric contamination and partly due to heterogeneity in the powdered samples. Before

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presenting the results of our extensive measurements of N in stone meteorites, we discuss the experiments that demonstrate a negligible contamination from atmospheric nitrogen in our technique.

Experimental

The experimental technique involves radio-chemical neutron activation analysis, employing the reaction $^{14}N(n, p)^{14}C$. The detailed procedure has been described by Shukla *et al.* (1978). Contribution from the interfering reaction $^{17}O(n, \alpha)^{14}C$, is estimated to be equivalent to 6 ppm N for a sample containing 40 wt. percent oxygen. Our data for the standards are corrected for an assumed 40 weight percent oxygen in all cases. Data for meteorites are uncorrected for the ^{17}O contribution in view of the different mineralogy of various meteorite samples and the consequent uncertainty about their oxygen contents. The reproducibility, as assessed by measurements on replicate samples of NBS-standard '33d' (N = 110 ppm) and other homogeneized samples is put at ±10%.

Nitrogen in Terrestrial Standards

Contamination problem   Any technique for the trace element determination is fraught with contamination from the reagents used. The special advantage with the neutron activation method is that contamination is serious only prior to irradiation, while it is a serious problem all through the procedure in other techniques. In spite of minimum sample handling, contamination is not ruled out for nitrogen. Atmosphere being its vast reservoir, the very exposure of the sample to the atmosphere may cause enough adsorption of nitrogen on to the sample surface. The problem is more serious in the case of powdered samples (all the standards) because of the larger surface/volume ratio. Although we have sealed our samples after overnight degassing at $\leqslant 40\mu$m Hg pressure, absence of appreciable adsorption needs to be demonstrated. We have performed the following three types of

experiments that show an absence of surface adsorption.

a) *Sieving experiment*   Adsorption is a surface correlated property and as such the amount of adsorbed nitrogen should increase with the decrease in grain size. In lunar soils, where most of the nitrogen is due to solar wind implantation, its concentration varies inversely with the grain size (Kothari and Goel, 1973; Goel *et al.*, 1975). The sieve analysis results on G-2 are given in Table 1. No excess nitrogen is observed in the finest size fraction (the increase of N in the $75-151\mu$m fraction most probably may be due to mineralogical differences). These results clearly indicate an absence of atmospheric nitrogen.

b) *Leaching experiment*   Surface adsorbed species are held by weak bonds and should be easily leached out by mild alkali or acid treatment. Aliquots of irradiated BCR-1 were leached (for 2 hours) with NaOH (0.1%); HNO_3(3 M); H_2SO_4(1:10) and HF(1%). In each case the residue was washed free of the leachent, dried and analysed for nitrogen. The results are presented in Table 2. The N contents of the residues are not much lower than the untreated sample, except for the HF attack. But in the case of HF leaching, about 80% material is leached out presumably dissolving most of the N bearing phases.

c) *Step-wise leaching experiment*   Goel and Kothari (1972) successfully pealed off the solar wind nitrogen from lunar soils, by a step-wise leaching scheme illustrated in Fig. 1.

Table 1. Nitrogen in sieve fractions of G-2
(Error = ±10%)

Grain size (μm)	Mass (mg)	N* (ppm)
<38	23.5	27.6
38-75	18.5	28.6
>75	8.5	49.3
Bulk	8.5	25.8

* Corrected for ^{17}O contribution.

Table 2. Nitrogen in leached residues of BCR-1

Leachant	Mass before leaching (mg)	Mass of recovered residue (mg)	N* (ppm) Bulk	Residue
0.1% NaOH	23.5	20.0	19.2	15.9
3M HNO$_3$	11.0	8.0	19.2	17.5
1:10 H$_2$SO$_4$	6.5	4.5	20.0	12.2
1.0% HF	20.0	3.0	22.5	5.7

* Corrected for ^{17}O contribution.

The powdered sample is sequentially leached with 1% NaOH and 3M HNO$_3$ and the radiocarbon fractions, liberated in the leaching process and the subsequent drying of the filtrate and residue, are collected and their activities assayed. The final residue and the dry cake from the filtrates are also analysed for their ^{14}C contents. All the leaching and drying operations are carired out in a closed system. The data on nitrogen liberated in various steps mentioned in the scheme (Fig. 1), are presented in Table 3, for the standards BCR-1 and G-2 along with the lunar soil 14163 (from GOEL and KOTHARI, 1972). The residue of the lunar soil had 15 ppm N as against 80 ppm in the virgin sample (GOEL and KOTHARI, 1972), demonstrating that ~80% of the N, which is surface implanted, is leached out. Adsorbed species must be more easily leachable in the same scheme as compared to the 'implanted' nitrogen. The con-

trast between the standards and 14163 is very clear. Most of the N in BCR-1 and G-2 is in the final residue.

The above three sets of experiments clearly demonstrate that in our technique a contamination due to the atmospheric N is small or negligible.

Comparison with other techniques The N data on the standards are presented in Table 4, and also compared with the literature values. In general our values are lower compared to the Arizona Group and higher compared to MÜLLER's numbers. To understand these discrepancies we have analysed portions of the splits used by MOORE (BCR-1, DTS-1 and BVHO-1) and MÜLLER (G-2 and W-1) (Table 4). Our BCR-1 value agrees with MOORE's number. But for DTS-1, our value is lower. Both our G-2 and W-1 numbers are higher compared to the numbers of MÜLLER. Incomplete outgassing of the atmospheric nitrogen in the Arizona technique (MOORE *et al.,* 1972; MOORE and LEWIS, 1976) and the selectivity for the chemically bound nitrogen in the Kjeldahl distillation (MÜLLER *et al.,* 1976) could be responsible for the observed trends. Labelling experiments have shown that in Kjeldahl distillation the nitrates present in a soil are only partially decomposed (GUIRAUD and FARDEAU, 1977).

NORRIS and SCHAEFFER's (1982) value for G-2 is close to our number. But their BCR-1 value, by far the highest reported, is a factor of 2 higher compared to our number, presumably owing to contamination as shown by stepwise heating experiments (NORRIS and SCHAEFFER, 1982). BCR-1 is, anyway, very heterogeneous in N.
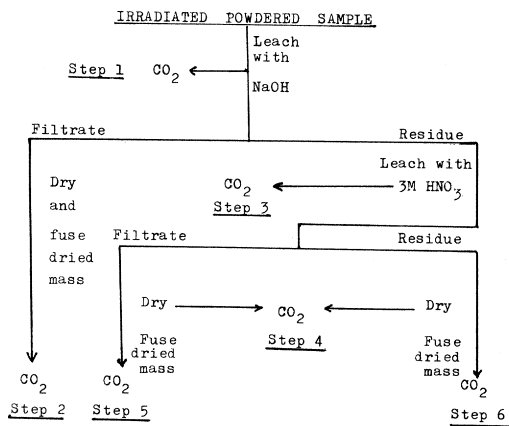

Fig. 1. Scheme for the step-wise leaching process. Data presented in Table 3 refer to the steps marked here. All the oeprations are carried out in a closed system (from: GOEL and KOTHARI, 1972).

Table 3. Nitrogen in various steps of the step wise leaching process*

Sample	BCR-1	G-2	Ambapur[+] Nagla	Dhajala[+]	14163 (**)
Starting mass (mg)	29.0	29.5	41.0	44.5	77.0
N(ppm) in steps***					
1	0.6	1.4	0.4	0.04	1.3
2	–	2.8	2.8	1.3	4.0
3	2.1 (24.5)	1.6 (29.0)	12.4 (38.5)	21.2 (43.0)	22.0
4	0	0	1.0	1.1	–
5	0	–	1.4	3.0	6.0
6	18.7 (22.0)	23.4 (27.0)	8.2 (22.0)	10.2 (22.0)	15.0
Sum of all steps	21.0	29.2	26.2	36.6	48.3
N(ppm) in bulk	26.0	39.0	40	18	80.0

* Uncorrected for ^{17}O contribution.
[+] See Table 5.
** From GOEL and KOTHARI (1972).
    Numbers in parantheses indicate the mass of the residue (mg) at this stage (see GOEL and KOTHARI, 1972).
*** See Fig. 1.

Table 4. Comparison of N (ppm) in terrestrial standards
(Error = ±10%)

Sample	This laboratory* replicate	Mean	Other laboratories 1	2	3	4
BCR-1-56 (CBM)	20.6, 24.4, 20.7 26.0, 15.9, 19.5	21.2	30.0			
BCR-1[a]		31.1		1.3	71	
DTS-1 (CBM)	8.4, 5.2, 12.0[b]	8.5	27.0			
BVHO-1 (CBM)	22.5, 22.7	22.6				
G-2 (OM)	30.4, 29.5, 38.0 32.5, 25.8 32.0, 35.0, 27.0	31.1	56.0	2.3	38	8±2
W-1 (OM)	28.9, 30.7, 26.6 32.2	29.6	52.0	10.8		14±0.6
AGV-1	32.0, 33.0	32.5	41.0	3.9		
GSP-1	25.0, 29.0, 33.0	26.0	48.0			
PCC-1[b]	41.0	41.0	43.0			
NIM-P	36.6, 35.6	36.1				
NIM-S	35.3, 35.9	35.6				
NIM-L	45.9, 42.7	44.3				
NIM-N	43.2, 42.6	42.9				
NIM-D	27.7, 26.6	27.1				
Russian standards						
Diabase	33.4, 23.5	28.5				
Granodiorite	32.2, 36.9	34.5				
Peridotite	27.8, 27.8	27.8				
Mayeskite	27.6, 22.1	24.8				

* Corrected for ^{17}O contribution.
    (CBM): Sample from C.B. MOORE; (OM): Sample from O. MÜLLER.
1. GIBSON and MOORE (1970); 2. BECKER and CLAYTON (1977); 3. NORRIS and SCHAEFFER (1982);
4. MÜLLER et al. (1976).
a. Mean value from Fig. 2; b. KOTHARI (1974).

The most serious disagreement is with the Chicago numbers which are lower by as much as a factor of 20 in some cases. NORRIS and SCHAEFFER (1982) have compared the N values of USGS standards from various laboratories (using different techniques) and concluded that there might be large scale heterogeneity in the USGS standards. The N contents of BCR-1, analysed in our laboratory over the past 10 years are represented by the histogram in Fig. 2a. Wide variations, from 15 to 62ppm, are seen. These data clearly indicate heterogeneity in the sample. Although there are fewer measurements, the same trend is also reflected in the sample G-2 (Fig. 2b). Replicate measurements R_1/R_2 (represents maximum value of N/minimum value of N) in each vial, for the standards NBS-33d and BCR-1 show a dispersion of up to 60% for BCR-1, whereas it is within 20% for NBS-33d. This clearly shows the validity of NBS-33d as a N-standard and at the same time points out the heterogeneity in BCR-1. Probably similar extensive N measurements may reveal heterogeneity for the other silicate standards as well. In fact G-2 and DTS-1 do show variations. But it would be highly improbable that the heterogeneity would always result in giving lower values for a laboratory, for different samples. On the other hand a consistent trend shown by a given technique must reflect its inherent virtues or shortcomings. It is worth-
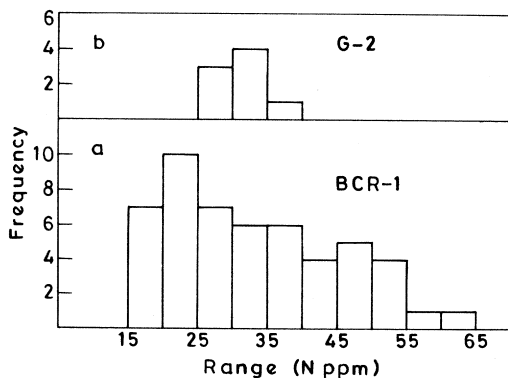
while to examine the vacuum pyrolysis method detail, in view of the serious disagreement.

Problems with the vacuum pyrolysis IHIDA (1959) found comparatively large amounts of nitrogen remaining in the melt of nitrides even in a dynamic vacuum of 10^{-4} to 10^{-5} torr. Some of the residual nitrogen was expelled from the melt upon cooling and some of the 'missing' nitrogen was recoverable by subsequent analysis of the spent vacuum fusion melt. The BISRA (1958) study has shown that the last traces of nitrogen become progressively more and more difficult to extract, in the vacuum fusion method, particularly in the presence of strong nitride forming elements (Al, Si, Zr and Ti) in the melt.

A comparison of nitrogen values by the vacuum pyrolysis method with those determined by other methods (Table 2 of BECKER and CLAYTON, 1975) in the lunar samples, brings out the following points: 1) A reasonably good agreement exists in the case of lunar soils. This is due to the fact that most of the nitrogen in lunar soils is surface implanted and easily extracted by the vacuum pyrolysis method. 2) In the case of breccias and rocks, the agreement is very bad, presumably due to the incomplete recovery of the more tightly bound indigenous nitrogen in these samples. 3) In the case of meteorites, there is an agreement between our numbers and the Chicago numbers for carbonaceous chondrites, where most of the N is present in the form of organic compounds that decompose very easily in pyrolysis. But for ordinary chondrites, the Chicago numbers are always low, presumably due to the refractory nature of most of the N in them.

The Chicago Group contends that their method ensures complete extraction of nitrogen from the samples as evidenced by the recovery of the most tightly bound spallogenic nitrogen upon melting the sample. This conclusion has been arrived by computing spallogenic ages from the highest temperature fraction of nitrogen (which is attributed to spallation production) and its $\delta^{15}N$ value (BECKER *et al.*, 1976).

Fig. 2. Histogram of the replicate measurements of nitrogen (a) (BCR-1 (52 measurements); (b) G-2 (8 measurements).

The equation $t_{sp} = \delta^{15}N \times N$, where both $\delta^{15}N$ and N correspond to the highest temperature fraction (BECKER *et al.,* 1976), has a self regulating feature. Contribution from low temperature fractions to the high temperature fraction can increase N; but $\delta^{15}N$ decreases because the low temperature fraction is isotopically lighter. Still the product $\delta^{15}N \times N$ can give an age that may fortuitously much with the spallogenic age. According to REEDY (1981) the ^{15}N production rate estimated by BECKER *et al.* (1976) might have been underestimated. Also the lunar regolith is known to have 1 to 1.5% of meteoritic component of the C1 type (ANDERS *et al.,* 1973) which should contribute 10 - 15 ppm N to the lunar soil. The meteoritic nitrogen has a complex isotopic pattern as revealed by step-wise heating experiments (FRICK and PEPIN, 1981). This mixes up with the isotopic pattern of the lunar soil and the resultant isotopic pattern cannot be taken too seriously as an index of any process.

NITROGEN IN STONE METEORITES

The nitrogen values for various meteorites are presented in Table 5 and are compared with the literature values for common samples in Table 6. With few exceptions, the values reported by the Arizona Group are higher than ours. We measured N in three powdered samples of carbonaceous chondrites (Murchison, Murray and Allende) from Arizona and found lower values. The most serious disagreement is for Orgueil, where the Arizona value is more than 3 times our value. This might be due to incomplete outgassing of the atmospheric nitrogen from the powdered samples (MOORE *et al.,* 1972, MOORE and LEWIS, 1976). For the C2 chondrites, Cold Bokkeveld, Murchison and Murray, the values reported by KUNG and CLAYTON (1978) agree with our results. However, for other samples their values are low by varying factors. Similarly values reported by MÜLLER *et al.* (1976) are also low. From controlled experiments it has been observed (KOTHARI and GOEL, 1973) that the laboratory

handling of the samples prior to irradiation does not introduce any contamination. It is possible that MÜLLER *et al.* (1976) and KUNG and CLAYTON (1978) have not been able to extract all the nitrogen. In the case of Abee, our value is a factor of 2 higher, and for Atlanta, it is a factor of 2 lower. Such differences might be due to the inherent heterogeneities of samples. KALLEMEYN *et al.* (1980) and RUBIN and KEIL (1980) have reported chemical heterogeneities for Abee.

For a given meteorite, replicate measurements indicate as much as a factor of two variation. This might be due to the variable amount of matrix material (rich in volatiles) in different pieces. FRICK and PEPIN (1981) found a N content of 42 ppm in the Allende $<4\mu$m fraction, as against 20 ppm N in a piece of Allende, reported by THIEMENS and CLAYTON (1981), and attributed it to different amounts of matrix material in the two samples. Our Allende N contents, ranging from 19 to 52 ppm, may reflect the differing matrix amounts in various pieces. Volatile-rich ultrafine matrix is apparently a common constituent of all chondrites (RAMBALDI *et al.,* 1981).

Carbonaceous chondrites of the type C1 and C2 show the highest nitrogen contents 500 to 1,000 ppm) whereas, C3 chondrites show quite variable amounts of nitrogen (42 to 252 ppm). Several nitrogen bearing organic compounds have been identified in these meteorites (HAYATSU, 1964; HAYATSU *et al.,* 1975). Taking average N values for C1, C2 and C3 chondrites as 990, 781 and 178 ppm, respectively, we find the corresponding ratio as 1 : 0.8 : 0.2 which is different from the ratio 1 : 0.6 : 0.3 suggested for volatile elements (LARIMER and ANDERS, 1967), and determined for Bi and Tl (LAUL *et al.,* 1970a, b). Nitrogen is apparently more strongly depleted in C3 chondrites as compared to the other volatiles (see GIBSON *et al.,* 1971b).

The average nitrogen values for H, L and LL chondrites from this work are 29, 34 and 31 ppm, respectively. Inclusion of the earlier data from this laboratory (KOTHARI and GOEL,

Table 5. Nitrogen contents of stone meteorites

Sample source	Class	Description	N(ppm) Error = ±10%	
			Replicate	Mean
1	2	3	4	5
C Chondrites				
Orgueil PP, 1297	C1	Matrix	1035, 1020, 915, 844	953
		Fragments	823, 972	898
		Tiny particles with white incl.	1260, 1220	1240
Al Rais NMNH, 1791	C2V	Matrix (may be some small chondrules)	965, 740,1000	902
Cold Bokkeveld GSI	C2M	One piece	516, 516	571
		Another piece	626, 626	
Murchison DL	C2M	One piece	1125, 1100, 1030	852
		Powder (CBM)	710, 736, 650, 825	
Murray CBM, 635.2	C2M	One piece	1020, 880, 960	797
		Powder (CBM)	712, 623, 793, 745	
			Av. C2 = 802	
Allende NMNH, 3496	C3V	Powder (CBM)	30, 48, 51, 52	42
		Pieces	49, 45, 52, 26, 29, 44, 19	39
		White particles	52, 33	
		Small pieces	38, 37	
		White incl.	13	
Vigarano DL	C3V	Coarse powder	236, 277, 248	260
Warrenton DL	C30	Fine powder	268, 196	232
			Av. C3 = 100	
Coolidge CBM, 397.4	C4V	Pieces	27, 21, 23	24
H Chondrites				
Dhajala NB	H3	Pieces	19, 17, 19	18
		Dark lithic frag.	6.3	
Fayetteville NMNH, 1731	H4	Pieces	39, 39	39
		Metallic chip	16	
Forest Vale AMS	H4	Pieces	29, 26, 20, 27	26
Ambapur Nagla GSI, 201	H5	Pieces	49, 54, 25, 46, 31, 33	40
Beardsley RD	H5	Pieces	35, 33	34
Pultusk RD	H5	Pieces	45, 47, 36, 38	40
		Fusion crust	60	
Richardton RD	H5	Pieces	31, 27, 29	29
Ucera DL	H5	Pieces	17, 16	17
Plantersville NMNH, 1228	H6	Pieces	22, 20	21
Phoum Sambo PP, 40/1654	H	Pieces	21	21
			Av. H = 29	
L Chondrites				
Bjurböle RD	L4	Pieces	46, 47	47
		Light particles	24	

Sample source	Class	Description	N(ppm) Error = ±10%	
			Replicate	Mean
1	2	3	4	5
Saratov ELK, 317	L4	Pieces	20, 17, 15	17
Assam GSI	L5	Pieces Fusion crust	35, 89 140	62
Bruderheim RD	L6	Pieces	22, 19	21
New Concord RD	L6	Pieces	36, 40	38
Peace River DL	L6	Pieces	35, 21	28
Rangala GSI, 320	L6	Pieces	24, 25, 36, 34	30
Montferre* YG	L	Pieces	28, 26, 20	25
			Av. L =	34
LL Chondrites Chainpur DL	LL3	Av. from sieve fractions	—	55
Paragould NMNH, 921	LL5	Pieces	12, 14, .12	13
St. Mesmin GP, 368	LL6	Av. from light and dark portions	—	24
			Av. LL =	31
E Chondrites Abee CBM, 701	E4	Pieces	506, 496	501
Atlanta* CBM	E6	Pieces	108, 85	97
Achondrites Norton County CBM, 523.3	Aub.	Av. from light dark fractions	—	70
		Glassy particle	92	
Pesyanoe NMNH, 1425	Aub.	Tiny particles	—	50
Shalka GSI, 174	Diog.	Pieces	15, 15, 10	13
Bereba PP, 1297	Euc.	Pieces	11, 13	12
Millibillille RAB, 13198D	Euc.	Pieces	25, 25	25
Sioux County CBM, 198.3	Euc.	Pieces	25, 25	25

The N data are not corrected for ^{17}O contribution. The meteorites marked with asterisk (*) are 'finds'. The cources of meteorites are abbreviated as follows:
AMS- The Australian Museum Sydney; CBM- C. B. MOORE, Arizona; DL- D. LAL, Ahmedabad; ELK- E. L. KRINOV, Moscow; GP- G. POUPEAU, Paris Museum; GSI- Geological Survey of India, Calcutta; NMNH- National Museum of Natural History, Washington, D.C.; NB- N. BHANDARI, Ahmedabad; PP- P. PELLAS, Paris Museum; RD- Ray DAVIS, Brookhaven; RAB- R. A. BINNS, Western Australian Museum; YG- Y. GILLET, Commission on Atomic Energy France.

Table 6. Comparison of nitrogen values of meteorites with literature data

Meteorite	N (ppm)		
	This work (Error = ±10%)	Arizona+ group	Others
Orgueil (fragments)	990	3185	2400,[a] 1476[b]
Cold Bokkeveld	571	1300	420,[c] 545,[b] 580[d]
Murchison	730*, 852	1500	845,[b] 828,[d] 950,[d] 226[e]
Murray	718*, 797	1905	1018,[b] 302[e]
Allende	47, 42*	62	19,[b] 18,[d] 15[e]
Vigarano	260	128	100[f]
Warrenton	232	92	–
Coolidge	24	51	15[d]
Forest Vale	26	40	–
Beardsley	34	57	–
Richardton	29	51	6,[d] 33[d]
Bjurböle	47	28	39,[c] 12,[d] 13[d]
Saratov	17	42, 107	27[i]
Bruderheim	21	32	14,[c] 31[g]
New Concord	38	21	45,[c] 10[h]
Chainpur	55	55	–
St. Mesmin	24	77	–
Norton County	70	–	10[d]
Abee	501	270, 262	277[d]
Atlanta	97	247	
Sioux County	25	29	

[+] GIBSON and MOORE (1971a) and GIBSON et al. (1971b).
[*] Powder samples from C. B. MOORE, Arizona.
(a) WIIK (1969); (b) INJERD and KAPLAN (1974); (c) KOTHARI and GOEL (1974); (d) KUNG and CLAYTON (1978); (e) MÜLLER (1974); (f) MASON (1963), (g) BUDDHUE (1966), (h) KÖNIG et al. (1961); (i) VINOGRADOV et al. (1963).

1974) gives the average values for H, L and LL chondrites as 25, 30 and 27ppm, respectively. GIBSON and MOORE (1971a) have observed a similar trend, although their values are higher than ours (H, 43ppm; L, 48ppm). Further, among the various petrologic types 4, 5 and 6, the nitrogen values do not show any regular variation indicating that the metamorphic events which produced these classes did not affect the nitrogen contents in ordinary chondrites (GIBSON and MOORE, 1971a). Perhaps most of the N in these meteorites is in the form of refractory nitrides (Si-N-Si bonds) and is tightly bound.

Even among the achondrites N contents are of the same range as in ordinary chondrites, indicating that N is not lost in the process of differentiation. Nitrogen might have been held in the silicate matrix by strong Si-N-Si bonds, so that it could not be driven off at the temperatures developed during the differentiation process. The two aubrites, Norton County and Pesyanoe, have 70 and 50ppm N, respectively, much higher compared to the rest of the achondrites analysed here. These two meteorites are polymict brecciated and might have acquired a volatile rich component during the impact events that resulted in their brecciation.

Nitrogen in chondrules Nitrogen contents of the chondrules analysed in this work, are given in Table 7. Most of the chondrules are lower in N as compared to the matrix material. In all the cases the ratio N (matrix)/N (chondrules) is greater than unity. Further, chondrules from the same meteorite display quite diverse nitrogen contents (Allende and Bjurböle). Taking the view that chondrules are formed by the impact melting process of chondritic masses (KERRIDGE and KIEFFER, 1977, DODD, 1978a, b), we presume that the loss of some mobile N might be responsible for their lower N contents.

Nitrogen in magnetic and non-magnetic frac-

Table 7. Nitrogen contents of chondrules((Error = ±10%)

Meteorites	Description	N (ppm) Replicate	N (ppm) Average	N-matrix N-chondrule
Al Rais	2 chondrules	–	312	2.9
Allende	1 chondrule	26, 30	28	1.5
	chondrules	15, 15	15	2.8
	1 big condrule with white spot	11, 11	11	3.8
Dhajala	dark chondrules	13		1.4
	light chondrules	8		2.3
Forest Vale	chondrules	–	13	2.2
Ambapur Nagla		22		1.7
Richardton	chondrules	23, 23	23	1.3
Bjurböle	chondrules	–	13	3.6, 1.0[+]
Saratov		13		1.3
Chainpur	chondrules	–	28	2.0

* *Uncorrected for* [17]*O interference.*
[+] *From KOTHARI and GOEL (1974).*

tions   Nitrogen data on magnetic and non-magnetic fractions separated with a hand magnet are presented in Table 8. Generally the ratio N (non-magnetic)/N (magnetic) > 1. Abee and Atlanta contain obsornite (TiN) (RAMDOHR, 1973) which might be present in the non-magnetic portion. Or there may be other N-rich carrier enhanced in the non-magnetic portion.

Nitrogen in light and dark phases   Some of the meteorites having light and dark phases in their matrices have been found to contain excessive amounts of rare gases in the dark phases (gas-rich meteorites). The dark portions also show

an excess of some other volatiles, e.g., Tl, Bi, In (LAUL *et al.,* 1970a, b, 1972, RIEDER and WÄNKE, 1969, MÜLLER and ZÄHRINGER, 1966). However, in the gas-rich meteorite Fayetteville the concentrations of volatile elements Zn and Cd are similar in both light and dark phases (CASE *et al.,* 1973). Earlier analyses of light and dark phases from Kapoeta and Assam (KOTHARI and GOEL, 1974) showed no enrichment of nitrogen in Kapoeta and a very nominal enrichment of nitrogen in the dark phase of Assam. In this work we have studied three meteorites showing light and dark structure (St. Mesmin, Pesyanoe and Norton County). The N data for these meteorites along with the data for Kapoeta

Table 8. Nitrogen in magnetic and non-magnetic fractions*

Sample	N (ppm) (Error = ± 10%) Bulk**	N (ppm) (Error = ± 10%) Magnetic	N (ppm) (Error = ± 10%) Non-magnetic	(N) non-magnetic (N) magnetic
Allende	42	19.8	19.5	0.9
Dhajala	18	13.2	13.3	1.0
Fayetteville	39	26.8	31.1	1.1
Forest Vale	26	22.7	24.4	1.1
Ambapur Nagla	40	36.5	45.8	1.3
Beardsley	34	28.7	42.8	1.5
Pultusk	40	26.0	34.5	1.3
Phoum Sambo	21	11.0	33.0	3.0
Saratov	17	18.6	15.5	0.8
Paragould	13	8.1	9.3	1.2
St. Mesmin	24	20.3	21.0	1.0
Abee	501	478	592	1.2
Atlanta	97	57.4	193	3.4

*   *Uncorrected for* [17]*O interference.*
** *Average value from Table 5.*

Table 9. Nitrogen in light and dark phases*

Meteorite	Phase	N (ppm) (Error = ± 10%)	
		Replicate	Average
St. Mesmin	Light	33, 22, 31, 25	22
	Dark	19, 24, 20	21
Pesyanoe	Light	—	33
	Dark	—	57
	Dark, very small light portion	—	73
Norton County	Light	—	54
	Dark	—	79
	Dark + Light	65, 70	68
Assam[1]	Light	—	53
Kapoeta[1]	Light	—	36
	Dark	31, 27, 21	26
	Dark + Light	—	30
	Dark + Light	33, 26	

* Uncorrected for contribution from ^{17}O.

1. From KOTHARI and GOEL, 1974.
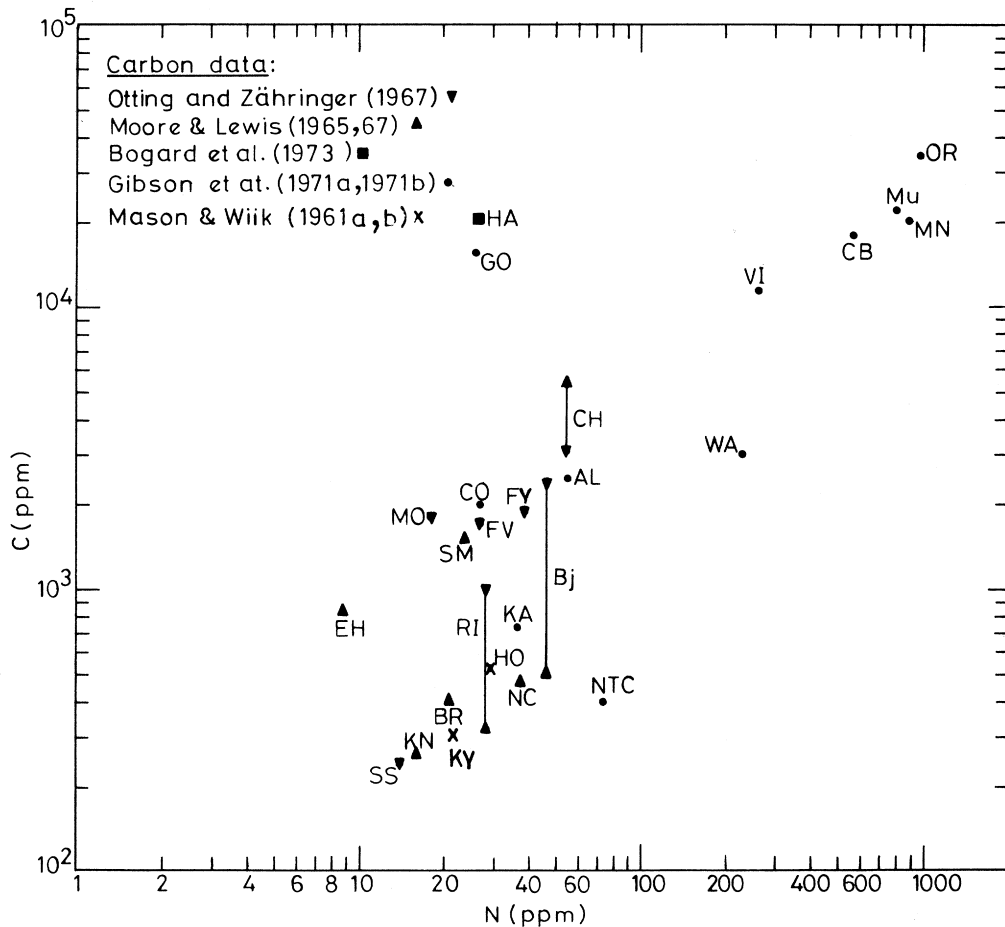
Fig. 3. Correlation of N vs. C. The nitrogen data are from this laboratory (present work; KOTHARI and GOEL, 1974). Meteorite names are abbreviated as follows: OR (Orgueil); MU (Murray); MN (Murchison); CB (Cold Bokkeveld); VI (Vigarano); WA (Warrenton); HA (Havero); GO (Goalpara); CH (Chainpur); AL (Allende); BJ (Bjurböle); NTC (Norton County); FY (Fayetteville); CO (Coolidge); FV (Forest Vale); MO (Modoc); SM (St. Mesmin); EH (Ehole); BR (Bruderheim); KN (Kunashak); SS (St. Severin) and KY (Kyushu, Satsuma).

and Assam (from KOTHARI and GOEL, 1974) are presented in Table 9. The dark phases of Pesyanoe and Norton County show an excess of N compared to their lighter counterparts. This excess cannot be accounted for by solar wind implantation (KOTHARI and GOEL, 1974). Admixture of 1-2 percent of C1 or C2 type material, which can explain excess amounts of some volatile elements in the dark phases (LAUL *et al.,* 1972) can account for the excess N observed in the dark phases of Pesyanoe and Norton County. However, such a component

can neither account for normal N nor the excess Li (MURTY *et al.,* 1983) in the dark phases of Kapoeta. A matrix material of the type, recently found in all classes of meteorites (RAMBALDI *et al.,* 1981) might account for the observed enrichments in the dark phases of gas-rich meteorites.

Elemental correlations The volatile elements Ar, Kr, Xe, C, In, Tl, Bi and Pb show a correlation with the degree of metamorphism in ordinary chondrites (ZÄHRINGER, 1966,
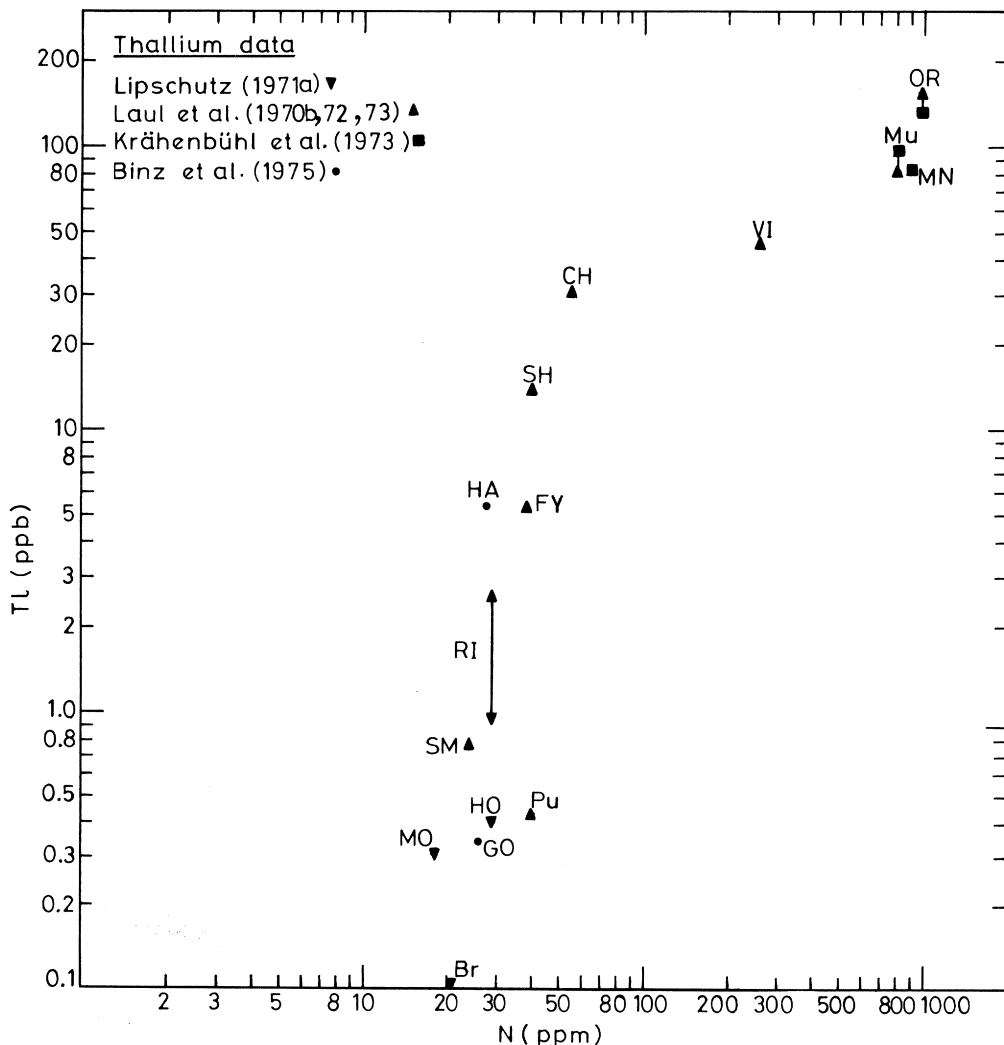

Fig. 4. Correlation of N vs. Tl. Meteorite names are abbreviated as: SH (Shergotty) and PU (Pultusk); Other abbreviations as in Fig. 3. Nitrogen data from this laboratory.

MARTI, 1967, HEYMANN and MAZOR, 1968, OTTING and ZÄHRINGER, 1967, TANDON and WASSON, 1968, HUEY and KOHMAN, 1973). The correlation fails for Ag, Cd, Br, Cs, Zn and N (LAUL et al., 1970a, b; KEAYS et al., 1971; ROSMAN and DE LAETER, 1974; GIBSON and MOORE, 1971a). We have tried to reveal the correlation of N with C, In, Tl, Bi, Cd and Zn.

1. *N vs. C*   Nitrogen data for various meteorites from this laboratory are plotted against the carbon data in Fig. 3. N values show posi-

tive correlation with C among carbonaceous chondrites. Most of the H chondrites also fall along the same line. The L chondrites with a lower C/N ratio form a group of their own. From Fig. 3 it is clear that for higher values of N, there is a positive correlation with C, whereas for the lower values of N, C varies by a factor of 100 while N varies only by a factor of 2. However, our technique fails to give correct N values when its concentration is low, owing to intereference from $^{17}O(n, \alpha)^{14}C$ reaction.

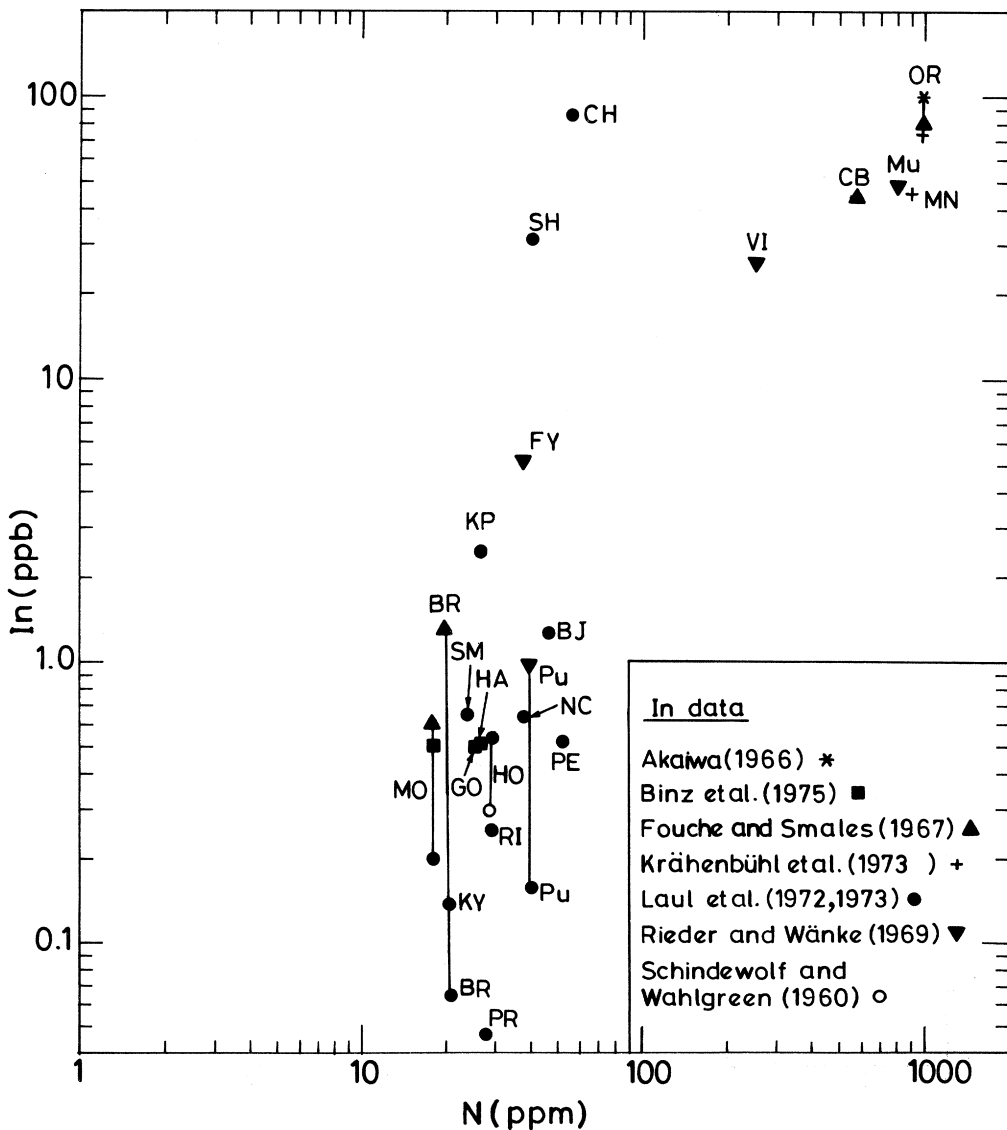In the models of LARIMER and ANDERS

Fig. 5. Correlation of N vs. In. Meteorite names are abbreviated as: KA (Kapoeta); PE (Pesyanoe) and PR (Peace River); Other abbreviations as in Figs. 3 and 4. Nitrogen data from this laboratory.

(1967) the accretion temperature of C1 and C2 chondrites is about 315°K. Perhaps, in these chondrites nitrogen condensed with carbon at low temperatures. Orgueil (C1) has a polymer-like organic material, which when subjected to stepwise pyrolysis, yields breakdown products containing nitrogen and carbon (BANDURSKI and NAGY, 1976 and references therein). Nitrogen and carbon apparently condensed in the form of compounds hydrolysable to purines, pyrimidines, amino-acids, etc. Other chemical forms like the refractory nitrides, which are important at higher temperatures create complications in the chemistry of nitrogen.

ii) *N vs. Tl, In and Bi*   These correlations are shown in Fig. 4 (N *vs.* Tl), Fig. 5 (N *vs.* In) and Fig. 6 (N *vs.* Bi). A behaviour similar to that of C is observed for these elements also. For high concentrations there is a positive correlation between N and these elements. However, for lower values of nitrogen (200-40 ppm), these elements vary independent of nitrogen.

iii) *N vs. Zn and Cd*   All these three elements do not show a correlation with the degree of metamorphism in ordinary chondrites. N shows a positive correlation with Zn for most of the stones as shown in Fig. 7, whereas N *vs.*
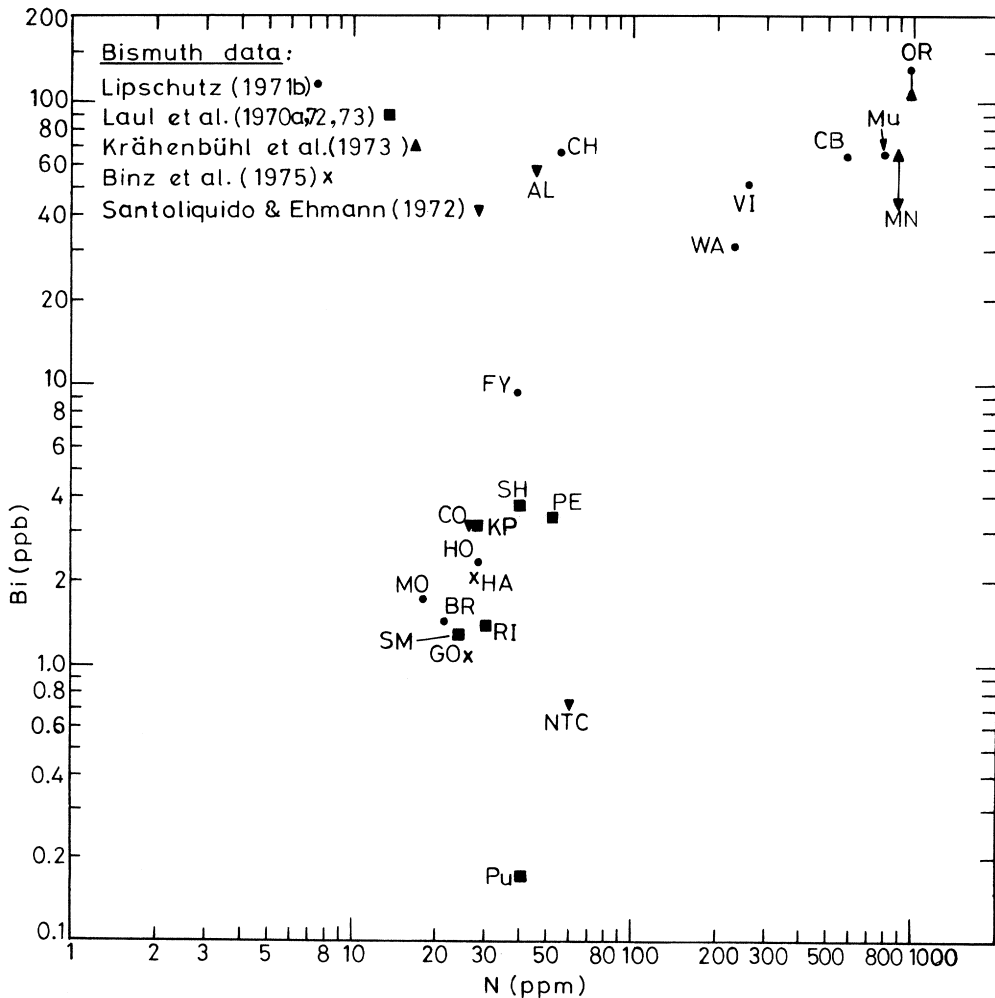

Fig. 6. Correlation of N vs. Bi. For meteorite names see Figs. 3, 4 and 5. Nitrogen data from this laboratory.

Cd correlation (Fig. 8) is noted among the carbonaceous chondrites alone. It is interesting to note that both Zn and N show almost the same concentration for H and L chondrites which does not seem to depend on the petrologic group types (ROSMAN and DE LAETER, 1974). Similar to N vs. Cd correlation, the correlation of Zn vs. Cd is also unsystematic (ROSMAN and DE LAETER, 1974).

All these correlation can be understood, if there are two carrier phases for N in ordinary chondrites. In one of the carrier phases N is present in a volatile form. The other carrier phase has N in a form unaffected by metamorphic changes. For the rest of the volatiles (C, Tl, In and Bi) there might be no refractory compounds present. In the case of carbona-

ceous chondrites N shows a positive correlation with the rest of volatiles, becuase of their low temperature accretion, wherein the volatile phases are retained. Most of the N in the ordinary chondrites (20 - 40 ppm) might then be contributed by the refractory N phase. Most probably N is held in the silicate matrix by strong Si-N-Si bonds. The laboratory heating experiments of LIPSCHUTZ and coworkers on primitive meteorites (NGO and LIPSCHUTZ, 1980; and references therein) show release patterns indicative of a single carrier phase for most of the volatile elements. The identification of "Holy Smoke" in all classes of chondrites (RAMBALDI et al., 1981) coupled with the fact that N is enriched in the fine grained low density matrix in chondrites (MURTY and GOEL,
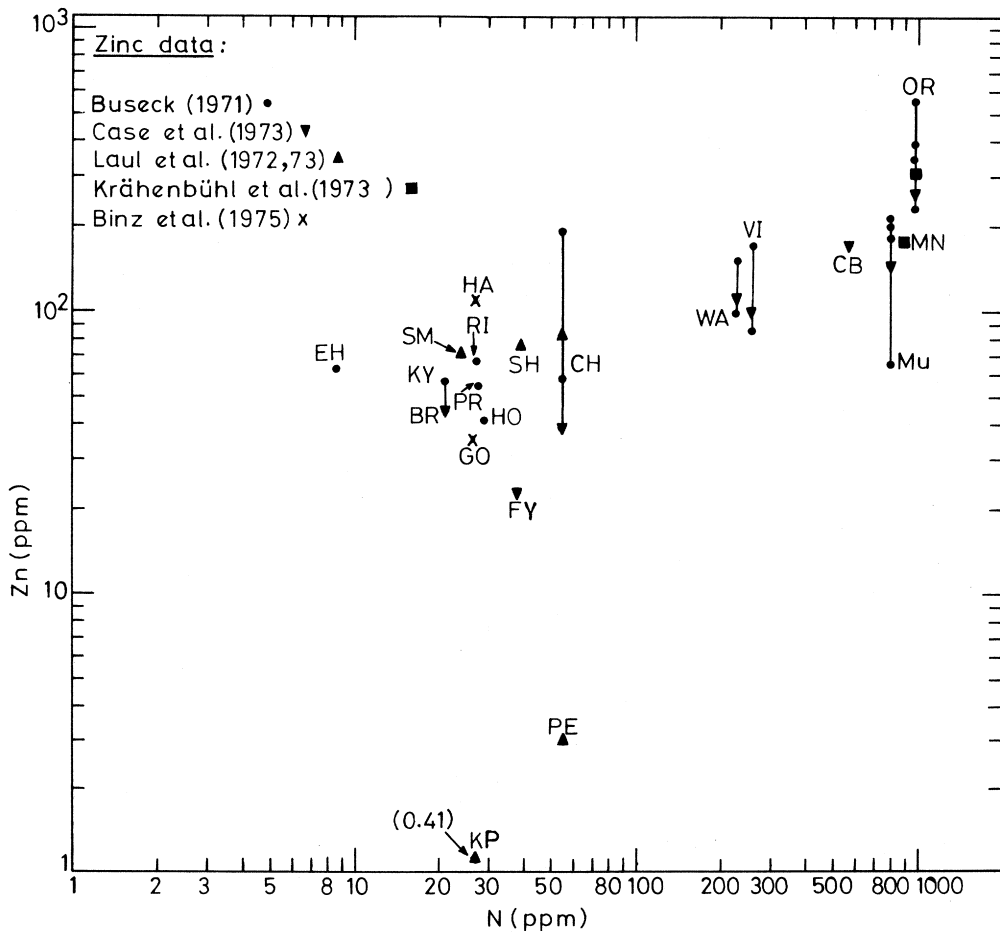

Fig. 7. Correlation of N vs. Cd. For abbreviations see Figs. 3, 4, 5. Nitrogen data from this laboratory.

1983) suggests a phase rich in N and other volatiles in all chondrites. The refractory N phase could contain a nitride. Recently FEGLEY (1983) analysed the stability fields of the probable refractory nitrides in meteorites and concluded that none of the metal nitrides are stable under the pressure and temperature conditions in the solar nebula. It has been suggested (LARIMER *et al.,* 1981) that sinoite ($Si_2 N_2O$) found in enstatite chondrites is also formed during metamorphism. The volatile elements trends in chondrites might have been established in the nebular condensation process and least affected in the parent body as suggested by TAKAHASHI *et al.* (1978) except for C and N. These two elements are mostly accreted in the form of volatile organic compounds at low tem-

peratures. Under the metamorphic conditions in the parent body part of the organic matter decomposes and the nitrogen is incorporated into the silicate frame work in the form of nitride ion N^{3-}. The similarity in the sizes of the N^{3-} and O^{2-} (1.46 and 1.37Å respectively) might facilitate the substitution (BAUR, 1972). This process may not be efficient and a partial loss of nitrogen might be expected, leading to a higher C/N ratio as compared to the carbonaceous chondrites. The C/N ratio of 30-50 for C1 and C2 chondrites as compared to C/N value of 200 for C3 and C4 chondrites (THIEMENS and CLAYTON, 1981) is clearly suggestive of partial N loss.
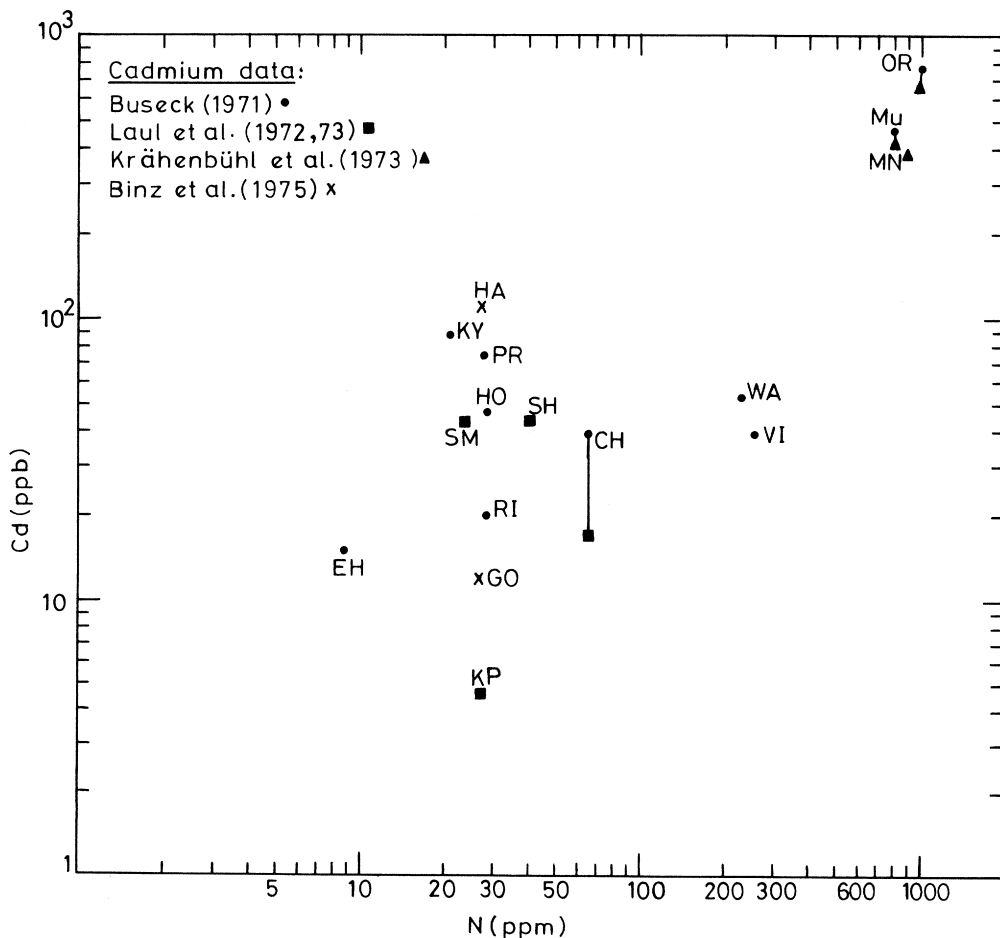
Fig. 8. Correlation of N vs. Cd. For abbreviations see Figs. 3, 4, 5. Nitrogen data from this laboratory.

of the valuable meteorite samples whose individual names are mentioned in Table 5. This research was partially supported through a grant from Indian Space Research Organization (ISRO) Bangalore.

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