

## Carbon and nitrogen in carbonaceous chondrites: Elemental abundances and stable isotopic compositions

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**Abstract**—We have undertaken a comprehensive study of carbon and nitrogen elemental abundances and isotopic compositions of bulk carbonaceous chondrites. A strategy of multiple analyses has enabled the investigation of hitherto unconstrained small-scale heterogeneities. No systematic differences are observed between meteorite falls and finds, suggesting that terrestrial processing has a minimal effect on bulk carbon and nitrogen chemistry. The changes in elemental abundance and isotopic composition over the petrologic range may reflect variations in primary accreted materials, but strong evidence exists of the alteration of components during secondary thermal and aqueous processing. These changes are reflected within the CM2 and CO3 groups and follow the published alteration scales for those groups. The nitrogen isotope system appears to be controlled by an organic host, which loses a <sup>15</sup>N-rich component with progressive alteration. This study recommends caution, however, over the use of bulk carbon and nitrogen information for classification purposes; variance in relative abundance of different components in carbonaceous chondrites is significant and reflects intrameteorite heterogeneities.

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### INTRODUCTION

The carbonaceous chondrite meteorites are considered to be the most primitive extraterrestrial materials available for analysis. They represent fragments of organic-rich asteroids that have escaped much of the geological processing experienced by larger bodies such as the Earth, and provide geochemists with the opportunity to assess the evolution of early solar system materials. Carbonaceous chondrites contain carbon (C) and nitrogen (N) yet display significant variations in these elements between individual meteorites (Kerridge 1985). These variations are considered to reflect, in part, post-accretionary asteroidal alteration. Differing extents of such secondary alteration have produced carbonaceous chondrites with specific chemical and mineralogical features that allow them to be separated into distinct chemical or petrologic types (Boato 1954; Van Schmus and Wood 1967; Wiik 1956).

A study by Kerridge (1985) into C and N in whole-rock carbonaceous chondrites was unable to identify any systematic trends between elemental abundance and extent of alteration, although his study presented results obtained by a

variety of analytical techniques. More recent work (Grady et al. 2002; Sephton et al. 2003, 2004b) has suggested that whole-rock isotopic compositions may be representative of preterrestrial alteration processes. Despite the potential to use these elements as proxies for preterrestrial processing, there has not been a comprehensive survey of bulk C and N abundances and isotopic compositions in carbonaceous chondrites that has covered all petrologic types using a single analytical method.

C- and N-bearing components in meteorites are present in both organic and inorganic form. The most abundant C- and N-bearing phase in carbonaceous chondrites is organic material, the majority of which is present as a condensed aromatic macromolecule. This organic network is believed to form, in part, by the accumulation of interstellar precursors and their subsequent modification on the asteroidal parent body (Cronin and Chang 1993). Studies have shown the macromolecule to be enriched in the heavy stable isotopes of H (D,  $\delta D = +300$  to  $+1600\text{‰}$ ) (Becker and Epstein 1982; Cronin and Chang 1993; Kerridge 1983, 1985; Robert and Epstein 1982) and N (<sup>15</sup>N,  $\delta^{15}N =$  up to  $170\text{‰}$ ) (Alexander et al. 1998; Robert and Epstein 1982). Such enrichments are

believed to be generated in interstellar environments (Becker and Epstein 1982; Sandford 2002). Some individual components are also enriched in C ( $^{13}\text{C}$ ,  $\delta^{13}\text{C} = -5$  to  $-25\%$ ) (Alexander et al. 1998; Sephton et al. 1998). However, experiments simulating asteroidal processing (Sephton et al. 2003) have shown that the macromolecule undergoes significant isotopic changes during thermal and aqueous alteration, the effects of which may be apparent in whole-rock samples (Sephton et al. 2004b). Such processing reduces  $^{13}\text{C}$  and  $^{15}\text{N}$  enrichments in the organic fraction, effects which should be systematically evident in meteorites of different petrologic types.

Aqueous processing is also responsible for the precipitation of carbonate species, which are the second most dominant C-bearing component in carbonaceous chondrites (0.2% whole rock) (Smith and Kaplan 1970). Although carbonates are present in all samples, they do not account for much of the C inventory within carbonaceous chondrites (Kerridge 1985). One exception is the Tagish Lake carbonaceous chondrite, which contains a significantly higher proportion of carbonates than any other carbonaceous chondrite (Zolensky et al. 2002). However, meteoritic carbonates can have a significant influence on bulk isotopic compositions, since they are enriched in  $^{13}\text{C}$  ( $\delta^{13}\text{C} > +20\%$ ) (Grady et al. 1991) relative to organic material. The first  $\delta^{13}\text{C}$  measurement of carbonates in the Orgueil carbonaceous chondrite revealed a  $\delta^{13}\text{C}$  value of  $+60\%$ , around  $+40$  to  $+70\%$  greater than that of terrestrial carbonates (Clayton 1963). Subsequent analyses of carbonates from CI and CM chondrites revealed similar values in the range of  $+40$  to  $+50\%$  (CMs) and  $+50$  to  $+60\%$  (CIs) (Grady et al. 1988).

Exotic hosts of C and N include diamond, graphite, and silicon carbide, which represent input from several source regions such as the interstellar medium, circumstellar disks, nebulae, and asymptotic giant branch stars (Anders and Zinner 1993). Although these can have highly anomalous isotopic values, their abundance in carbonaceous chondrites is minor and they are not thought to contribute greatly to the isotopic compositions of whole-rock samples (Kerridge 1985).

It is evident, therefore, that C- and N-bearing phases have specific sources and physiochemical properties, and their characteristic chemical responses to solar system processing constitute a valuable record of past events.

Over fifty years after Boato's (1954) first analyses, this study intends to re-examine the use of whole-rock C and N abundances and isotopic compositions as a tool for classifying carbonaceous chondrites and identifying anomalous samples. Through multiple analysis using a consistent method, heterogeneity between and within meteorites has been assessed and the effects of preterrestrial alteration processes over a range of carbonaceous chondrite groups constrained. The resultant data set is far larger than that previously available and reveals important variations attributable to secondary processing in the early solar system.

## EXPERIMENTAL METHODS

### Samples

Twenty-six samples of CM, CO, CV, CI, CR, and CK carbonaceous chondrites plus one ungrouped sample, in powder or chip form, were chosen for analysis of their C and N abundance and isotopic composition. Five of these meteorites, Kivesvaara, Colony, Leoville, Maralinga, and Coolidge, are finds; all others are recorded falls. Crushing was carried out using an agate pestle and mortar and sample size varied between 0.47 and 21.90 mg. Subsamples and splits of some meteorites were analyzed to examine intrameteorite variations. Samples were loaded into commercially available ultra-clean tin buckets (Elemental Microanalysis Ltd.) for analysis.

### C and N Abundance and Isotopic Composition

C and N abundances and isotopes were measured using an elemental analyzer (Europa ANCA-SL) coupled to a continuous-flow mass spectrometer (Europa GEO 20-20). Using a continuous flow enabled the entire sample gas to be analyzed, allowing the analysis of much smaller sample sizes than used by most authors previously. Samples were purged with He at  $40 \text{ ml min}^{-1}$  and then introduced to a ceramic furnace at  $1000 \text{ }^\circ\text{C}$ , where ignition of the tin bucket in a pulse of oxygen raised the temperature locally to  $1800 \text{ }^\circ\text{C}$ . Complete sample combustion was assured by passage through chromium oxide at  $1000 \text{ }^\circ\text{C}$ . Sulphur and halides were removed by passing the gases through copper oxide and silver wool. An He carrier gas (99.999% purity) at  $60 \text{ ml min}^{-1}$  flow rate was employed. A second furnace at  $600 \text{ }^\circ\text{C}$  containing copper enabled sample reduction to produce molecular  $\text{N}_2$ . Any  $\text{H}_2\text{O}$  present was removed with anhydrous magnesium perchlorate. The gas stream then passed into the GC oven set at  $70 \text{ }^\circ\text{C}$  (50 cm,  $\frac{1}{2}$  inch outer diameter packed Carbosieve G 60-80 column) for separation and then to the mass spectrometer where the stable isotope ratios of carbon ( $^{13}\text{C}/^{12}\text{C}$ ) and nitrogen ( $^{15}\text{N}/^{14}\text{N}$ ) were determined. The abundance of stable isotopes are expressed using the  $\delta$  notation. These indicate the difference in per mil ( $\%$ ) between the ratio in the sample and the same ratio in the standard.

$$\delta\% = [(R_{\text{sample}} - R_{\text{standard}}) / R_{\text{standard}}] \times 1000 \quad (1)$$

where  $R = ^{13}\text{C}/^{12}\text{C}$  for carbon and  $^{15}\text{N}/^{14}\text{N}$  for nitrogen. The standard for carbon (PDB) has  $^{13}\text{C}/^{12}\text{C} = 0.0112372$  and that for nitrogen (air) has  $^{15}\text{N}/^{14}\text{N} = 0.003765$ . The reference material used was a mixture of cane sugar (calibrated against the IAEA standard NBS 21) and IAEA standard N-3. Comparison of the reference material at the  $100 \mu\text{g}$  level gave data reproducibility of  $\pm 0.09\%$ . Batches of samples were bracketed between system blanks and replicates of the

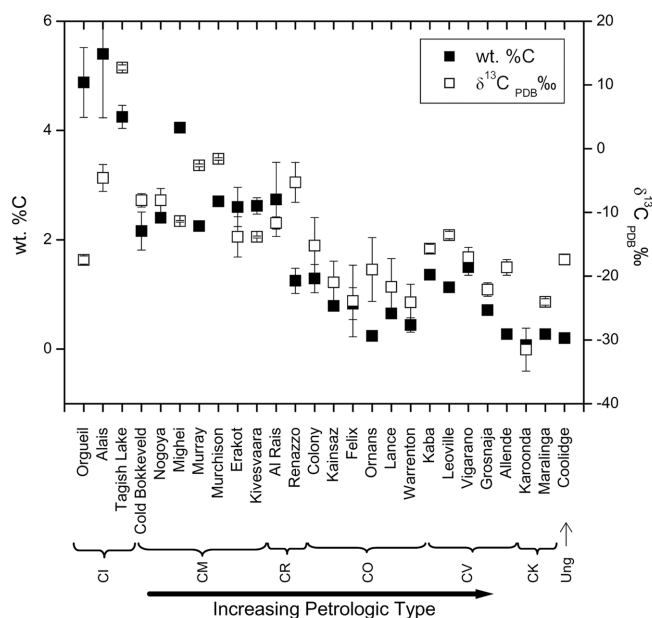


Fig. 1. The mean carbon abundance (wt% C) and carbon isotopic composition ( $\delta^{13}\text{C}$ ) of all carbonaceous chondrites, grouped according to increasing petrologic type. CM chondrites are grouped according to alteration scale of Browning et al. (1996). CO and CV chondrites are grouped according to metamorphic grades (Scott and Jones 1990; Bonal et al. 2004). Error bars denote standard deviations as defined in Table 1.

reference materials to allow comparison of samples to references and to monitor instrument drift.

### Statistical Analysis

In order to identify the presence of anomalous samples and/or results, standard deviations were calculated for individual meteorites and meteorite groups. A measured value  $>2\sigma$  (95% confidence) from the mean was taken to imply that the subsample or meteorite was anomalous. Student's *t*-tests (unpaired, two-tailed) were carried out in order to test the significance of the data between each chondrite group with a null hypothesis stating that the abundances and isotopic compositions are the same for each petrologic group ( $\mu_1 - \mu_2 = 0$ ,  $\alpha = 0.05$ ). The critical level of statistical significance was chosen to be  $p < 0.05$  (95% confidence).

## RESULTS AND DISCUSSION

Figures 1 and 2 display the mean C and N elemental abundances and isotopic compositions for all carbonaceous chondrites. The CI chondrites display the highest abundances of C and N; the CK chondrites display the lowest. Similarly, there is a trend of more negative  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values with increasing petrologic type from type 1 to 4 (Figs. 1 and 2). The CR chondrites are outliers with relatively heavy  $\delta^{15}\text{N}$

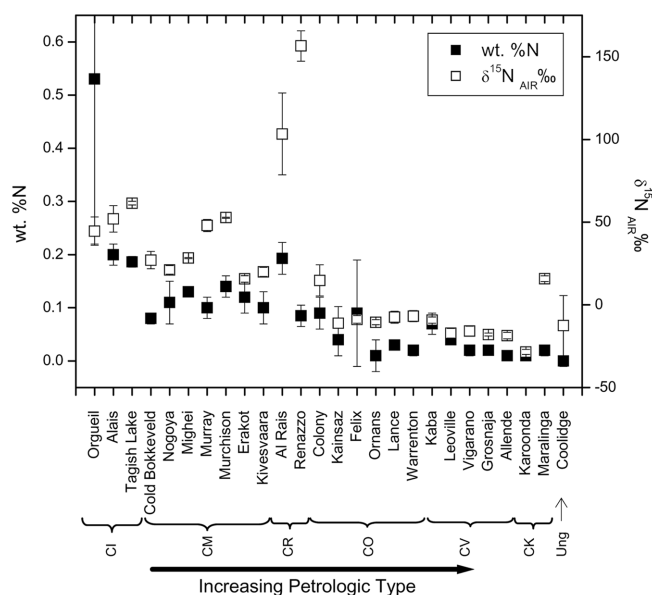


Fig. 2. The mean nitrogen abundance (wt% N) and nitrogen isotopic composition ( $\delta^{15}\text{N}$ ) of all carbonaceous chondrites, grouped according to increasing petrologic type. CM chondrites are grouped according to alteration scale of Browning et al. (1996). CO and CV chondrites are grouped according to metamorphic grades (Scott and Jones 1990; Bonal et al. 2004). Error bars denote standard deviations as defined in Table 1.

values as has previously been observed (Alexander et al. 1998; Kolodny et al. 1980; Kung and Clayton 1978). These broad trends have been reported in previous studies (Grady et al. 1991; Kerridge 1985; Robert and Epstein 1982) and have been interpreted as reflecting changes in elemental abundance and isotopic compositions brought about by preterrestrial processing.

Figure 3 compares the results of this study with published data (available in the Appendix). For C abundance (Fig. 3a) our data falls within, but at the top of, the range of published values. Our relatively high values may reflect the improved sensitivity of the analytical method employed here compared with that of studies over the previous 50 years. The ranges of published values for most carbonaceous chondrites are tightly constrained and include our data. Exceptions to this are Nogoya (CM2) and the CI chondrites which display variance of 2 to  $>3$  wt% C within each meteorite. This internal variance may reflect the analysis of heterogeneous fragments that contain varying amounts of C-bearing components, in particular carbonates that may influence bulk C abundances.  $\delta^{13}\text{C}$  values (Fig. 3b) measured for each carbonaceous chondrite in this study are comparable with and plot within the range of published values. Exceptions to this are Tagish Lake (CI2), Erakot (CM2), and Karoonda (CK4), for which there are too few other data points for accurate comparison. However, it is believed that both analyses of Tagish Lake (this study; Grady et al. 2002) were of the carbonate-rich lithology, which strongly

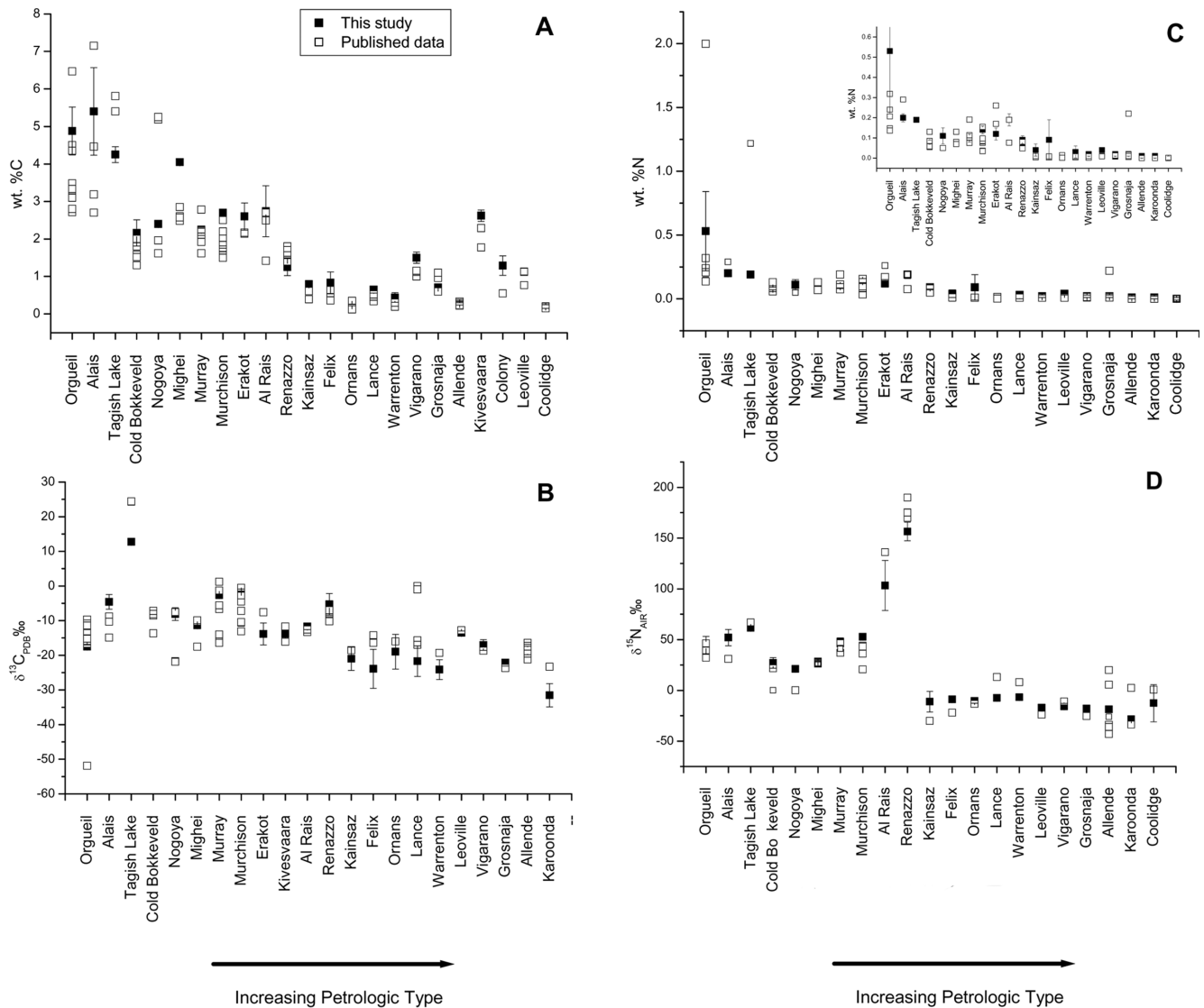


Fig. 3. A comparison of bulk carbon and nitrogen abundances and isotopic compositions from this study with published data (Appendix 1). The insert in Fig. 3c shows  $\delta^{15}\text{N}$  values, with data points greater than 1‰ excluded for data clarity. Error bars denote standard deviations as defined in Table 1.

influences  $\delta^{13}\text{C}$  towards positive values. Even taking this into consideration, the published data does not reflect the decrease in  $\delta^{13}\text{C}$  with increasing petrologic type seen in Fig. 1. Indeed, Fig. 3b suggests the CI, CM, and CR chondrites display similar  $\delta^{13}\text{C}$  values (approximately  $-9\%$ ), yet the CO, CV, and CK chondrites display lighter yet consistent  $\delta^{13}\text{C}$  values (approximately  $-20\%$ ). Although this is not the same trend as seen in Fig. 1, it does indicate that samples of low petrologic type have relatively higher  $\delta^{13}\text{C}$  values than samples of higher petrologic type.

Figure 3c compares the N abundances obtained in this study with published values. As with C, our data plots within the ranges defined by previous studies. However, Grady et al. (2002) reported values for Orgueil (CI1) and Tagish Lake

(CI2) which plot outside our ranges and those of other previous studies. Figure 3c (insert) omits this data in order to examine the relationships between our data and that of previous studies at low N abundances. The CI, CM, and CR chondrites show variance in N abundance compared with CO, CV, and CK chondrites (Fig. 3c insert). However, this may generally reflect a greater availability of previous data for type 1 and 2 chondrites than for types 3 and 4. This difference in variance precludes an accurate identification of any trends within the data, e.g., the trend of decreasing N abundance with increasing petrologic type (Fig. 2) cannot be verified. N isotopes from this study plot within the ranges defined by previous studies (Fig. 3d). The CR chondrites Al Rais and Renazzo display similar anomalous  $\delta^{15}\text{N}$  values relative to

Table 1. Mean elemental abundances and isotopic compositions for all chondrites analyzed in this study.

Sample	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)
Falls					
Orgueil	CI1	4.88 ± 0.64	-17.5 ± 0.64	0.53 ± 0.31	+44.6 ± 8.55
Alais	CI1	5.40 ± 1.17	-4.60 ± 2.11	0.20 ± 0.02	+52.0 ± 7.98
Tagish Lake	CI2	4.25 ± 0.21	+12.73 ± 0.41	0.19 ± 0.01	+61.5 ± 1.11
Cold Bokkeveld	CM2	2.16 ± 0.35	-8.10 ± 1.04	0.08 ± 0.01	+27.0 ± 5.20
Nogoya	CM2	2.40 ± 0.04	-8.10 ± 1.85	0.11 ± 0.04	+21.1 ± 2.57
Mighei	CM2	4.05 ± 0.02	-11.4 ± 0.13	0.13 ± <0.01	+28.3 ± 0.38
Murray	CM2	2.25 ± 0.09	-2.62 ± 0.22	0.10 ± 0.02	+48.0 ± 3.71
Murchison	CM2	2.70 ± 0.06	-1.61 ± 0.19	0.14 ± 0.02	+52.7 ± 0.31
Erakot	CM2	2.60 ± 0.36	-13.9 ± 3.16	0.12 ± 0.03	+15.7 ± 2.02
Al Rais	CR2	2.74 ± 0.68	-11.7 ± 0.96	0.19 ± 0.03	+103 ± 24.7
Renazzo	CR2	1.25 ± 0.23	-5.29 ± 3.13	0.09 ± 0.02	+157 ± 9.18
Kainsaz	CO3	0.79 ± 0.03	-21.0 ± 3.36	0.04 ± 0.03	-11.1 ± 10.1
Felix	CO3	0.83 ± 0.29	-23.9 ± 5.63	0.09 ± 0.10	-8.80 ± 2.29
Ornans	CO3	0.24 ± 0.02	-19.0 ± 5.02	0.01 ± <0.01	-10.5 ± 3.63
Lancé	CO3	0.65 ± 0.09	-21.7 ± 4.44	0.03 ± 0.03	-7.40 ± 1.46
Warrenton	CO3	0.44 ± 0.13	-24.1 ± 2.87	0.02 ± 0.01	-6.80 ± 3.34
Kaba	CV3	1.36 ± 0.08	-15.7 ± 0.69	0.07 ± 0.02	-9.20 ± 1.92
Vigarano	CV3	1.50 ± 0.15	-17.0 ± 1.55	0.02 ± 0.01	-15.9 ± 3.00
Grosnaja	CV3	0.71 ± 0.01	-22.1 ± 1.05	0.02 ± <0.01	-18.0 ± 0.90
Allende	CV3	0.27 ± 0.03	-18.6 ± 1.23	0.01 ± <0.01	-18.6 ± 1.96
Karoonda	CK4	0.07 ± 0.02	-31.5 ± 3.37	0.01 ± 0.01	-28.4 ± 1.55
Finds					
Kivesvaara	CM2	2.62 ± 0.15	-13.8 ± 0.15	0.10 ± 0.03	+19.9 ± 2.74
Colony	CO3	1.29 ± 0.26	-15.2 ± 4.41	0.10 ± 0.03	14.8 ± 9.47
Leoville	CV3	1.14 ± 0.09	-13.6 ± 0.56	0.04 ± 0.01	-17.1 ± 2.91
Maralinga	CK4	0.28 ± 0.01	-24.0 ± 0.51	0.02 ± 0.01	17.0 ± 1.76
Coolidge	C4	0.01 ± 0.05	-17.4 ± 0.78	0.01 ± 0.01	<0.01 ± 18.3

other carbonaceous chondrites, as observed in Fig. 2. The published data reflects the trend of decreasing  $\delta^{15}\text{N}$  with increasing petrologic type. However, the CO, CV, and CK chondrites display less variance than CI, CM, and CK chondrites, either due to the scarcity of previously published data or the inherent accuracy problems associated with isotopic analyses at such low abundances.

In general, the results of this study fall within the limits of previously published work for both C and N elemental abundance and isotopic composition. Our initial observation of decreasing  $\delta^{13}\text{C}$  values with increasing petrologic type (Fig. 1) cannot be supported by published data (Fig. 3b). For C and N abundance and  $\delta^{15}\text{N}$ , the trends observed solely from our study are reflected within the published data (Figs. 3a, 3b, and 3d). However, adequate comparisons of chondrites of low petrologic type are hindered by the scarcity of previous data. It is appropriate therefore to use our data and that of previous studies to undertake further investigations of bulk chondrite chemistry.

### Intrameteorite Heterogeneity

In order to establish how informative these trends are, it is necessary to consider the carbonaceous chondrites as

multicomponent systems, since C and N may reside in several entities. Previous studies have highlighted the inhomogeneous distribution of C- and N-bearing components in the carbonaceous chondrites that can lead to small-scale heterogeneities (Grady et al. 1991; Kerridge 1985; Robert and Epstein 1982). The approach of using multiple analyses in this study provides a means to investigate the magnitude of the heterogeneities within each chondrite and reduce their influence. Such heterogeneities can lead to incongruous mean values, data spread, and the potential masking of geochemical trends.

Standard deviations (Table 1) of each sample mean have been calculated in order to constrain the extent of sample heterogeneity and allow a quantitative comparison between the carbonaceous chondrites. Individual analyses of each carbonaceous chondrite are given in Table 2. The CI chondrites show the largest standard deviations and by implication, the greatest degree of heterogeneity in C abundance compared with chondrites of higher petrologic type.  $\delta^{13}\text{C}$  values are heterogeneous for the CO3 chondrites Colony, Felix, and Lancé in particular. However, no subsamples of any carbonaceous chondrite over the entire petrologic range showed standard deviations outside the 95% confidence level.

Table 2. Bulk abundances and isotopic compositions obtained for each carbonaceous chondrite.

Sample	Sample size (mg)	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)
Falls						
Orgueil	5.23	CI1	4.72	-16.7	0.56	38.0
	4.49		4.40	-17.2	0.82	41.4
	4.44		4.57	-18.2		
	7.66		5.82	-17.8	0.21	54.2
Alais	8.95	CI1	4.70	-1.73	0.19	45.7
	12.1		4.70		0.18	41.8
	14.4		4.58			
	15.4		4.45			
	16.4		4.34			
	6.61		6.98	-6.77	0.24	61.1
	6.26		6.77	-5.20	0.21	56.5
	5.38		6.67	-4.70	0.18	54.7
Tagish Lake	5.64	CI2	4.44	12.4	0.20	60.7
	6.43		4.02	13.0	0.18	62.3
Cold Bokkeveld	4.29	CM2	2.39	-9.27	0.08	25.4
	2.42		1.98	-7.98	0.08	24.4
	2.94		1.96	-7.46	0.07	27.2
	2.43		2.04	-6.86	0.09	23.8
	2.55		1.84	-7.58	0.06	37.4
	1.39		2.76	-9.43	0.08	24.2
	2.28		2.40	-6.74	0.09	19.7
	2.13		2.39	-7.66	0.09	20.3
Nogoya	2.40	CM2	2.43	-7.47	0.09	21.1
	2.28		2.45	-7.13	0.09	22.1
	3.03		2.39	-6.72	0.06	25.4
	1.35		2.43	-9.12	0.15	17.8
	1.21		2.33	-11.7	0.17	
	10.0		4.06	-11.6	0.13	28.8
Mighei	10.3	CM2	4.02	-11.3	0.13	28.0
	9.94		4.05	-11.4	0.13	28.1
	10.5		4.07	-11.3	0.13	28.2
	10.6		2.31	-2.68	0.11	48.8
Murray	10.5	CM2	2.29	-2.64	0.11	47.9
	10.1		2.33	-3.01	0.11	48.0
	10.5		2.30	-2.60	0.11	47.7
	2.32		2.19	-2.42	0.09	41.9
	2.07		2.09	-2.38	0.07	53.5
	10.1		2.82	-1.42	0.15	53.0
Murchison	9.81	CM2	2.70	-1.83	0.15	52.5
	11.2		2.82	-1.80	0.15	52.4
	11.1		2.77	-1.44	0.15	52.6
	2.80		2.39	-1.54	0.11	53.1
	2.90		2.74	-16.6	0.14	15.0
Erakot	2.86	CM2	2.11	-16.5	0.10	17.5
	1.72		2.96	-11.8	0.10	17.2
	2.00				0.15	13.2
	1.45		2.59	-10.5		
Al Rais	6.62	CR2	3.22	-12.3	0.21	121
	2.20		2.26	-11.0	0.17	85.8
Renazzo	12.2	CR2	1.09	-7.50	0.07	150
	13.4		1.41	-3.08	0.10	163
Kainsaz	2.02	CO3			0.07	-5.94
	3.03		0.77	-21.9	0.04	-11.0
	2.40		0.83	-23.8	0.06	-2.33
	11.4		0.77	-17.3	0.01	-25.3

Table 2. *Continued.* Bulk abundances and isotopic compositions obtained for each carbonaceous chondrite.

Sample	Sample size (mg)	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)
Felix	2.08	CO3	0.77	-27.1	0.06	-6.14
	1.95		0.85	-26.8	0.06	-10.6
	0.58		1.32	-29.8	0.24	-7.66
	19.6		0.62	-16.7		
	17.1		0.60	-19.1	0.01	-10.8
Lancé	2.33	CO3	0.73	-26.4	0.06	-6.03
	3.47		0.54	-23.7	0.03	-7.33
	1.89		0.69	-26.3	0.07	-8.94
	7.94		0.59	-19.7	0.01	
	16.2		0.57	-18.2	0.01	
	24.3		0.75	-15.8	<0.01	
Ornans	12.0	CO3	0.24	-22.2	0.01	-11.8
	9.80				0.01	-11.9
	20.8		0.27	-11.5	<0.01	-7.34
	16.4		0.22	-20.8	<0.01	-15.1
	15.9		0.23	-21.4	<0.01	-6.29
Warrenton	4.25	CO3	0.53	-27.6	0.04	-7.02
	3.63		0.26	-26.1	0.02	
	3.39		0.29	-25.9	0.03	-5.09
	11.9		0.49	-23.2	0.01	-11.7
	20.5		0.48	-21.7	0.01	-7.48
Kaba	24.9	CV3	0.59	-20.2	<0.01	-2.70
	13.4		1.41	-15.2	0.09	-7.87
Vigarano	19.1	CV3	1.30	-16.2	0.05	-10.6
	10.9		1.42	-17.9	0.01	
	13.9		1.41	-17.6	0.02	
	24.2		1.77	-14.3	0.03	-19.3
	15.		1.44	-18.1	0.01	-13.8
Grosnaja	17.0	CV3	1.49	-17.3	0.01	-14.5
	17.7		0.72	-21.4	0.02	-17.3
Allende	18.4	CV3	0.70	-22.9	0.02	-18.6
	14.9		0.26	-18.8	<0.01	
	11.7		0.26	-20.0	0.01	
	18.4		0.26	-18.6	0.01	-16.6
Karoonda	21.1	CK4	0.24	-18.9	0.01	-18.6
	18.4		0.33	-16.7		-20.6
	5.84		0.08	-33.0		
	9.26		0.08	-34.0	0.01	
	15.9		0.09	-25.6	0.01	-29.5
	18.5		0.06	-32.8		
Finds Kivesvaara	30.7	CM2	0.04	-32.3	<0.01	-27.4
	3.15		2.74	-13.4	0.11	22.3
	3.16		2.70	-13.6	0.10	20.6
	3.14		2.75	-14.5	0.10	23.0
	3.41		2.65	-13.4	0.15	17.0
	2.19		2.38	-13.6	0.09	16.3
	2.09		2.50	-14.5	0.07	20.3
Colony	1.39	CO3	1.72	-21.6	0.15	
	2.66		1.24	-19.4	0.09	4.05
	2.20		1.05	-18.2	0.10	5.21
	8.89		1.09	-12.1	0.09	27.2
	11.2		1.05	-11.1	0.09	23.7
	19.4		1.42	-11.0	0.10	12.3
11.5	1.48	-13.1	0.05	16.3		

Table 2. *Continued.* Bulk abundances and isotopic compositions obtained for each carbonaceous chondrite.

Sample	Sample size (mg)	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)
Leoville	14.4	CV3	1.02	-13.6	0.03	
	6.28		1.21	-14.2	0.03	
	24.8		1.22	-12.6	0.03	-19.7
	21.0		1.04	-13.3	0.03	-14.1
	14.4		1.19	-13.7	0.04	-15.2
	17.8		1.13	-13.8	0.05	-19.6
Maralinga	10.7	CK4	0.28	-25.1	0.02	15.8
	10.7		0.27	-23.0	0.01	18.3
Coolidge	23.6	C4	0.17	-17.9	<0.01	-3.93
	22.3		0.17	-18.1	<0.01	-0.73
	22.2		0.21	-17.2	<0.01	-39.9
	23.7		0.27	-16.4	<0.01	-5.79

Table 3. Calculated  $p$  values from unpaired, two-tailed student's  $t$ -tests comparing mean results from each petrologic group.  $p < 0.05$  indicates a significant statistical difference at the 95% confidence level.

	C (wt%)			$\delta^{13}\text{C}$ (‰)			N (wt%)			$\delta^{15}\text{N}$ (‰)		
	CM	CO	CV	CM	CO	CV	CM	CO	CV	CM	CO	CV
CI	0.020	0.001	0.002	0.641	0.184	0.258	0.219	0.147	0.139	0.03	<0.001	0.001
CM		<0.001	0.001		0.001	0.025		0.006	0.003		0.001	0.002
CO			0.078			0.152			0.63			0.060

The use of a consistent method in this study allows the cause of the heterogeneities, either indigenous to the sample or a function of sampling, storage, operator, or instrument error, to be investigated. The same aliquot of Alais, for example, was sampled on two separate occasions, for eight individual analyses, and gave C yields of between 4.34 and 4.70 wt% C (from five multiple analyses) and between 6.69 and 6.98 wt% C (from three multiple analyses). The possible effects of operator or machine error can be discounted, as other samples and standards analyzed on these occasions show no such differences. To some extent, a similar range in C abundance is also found within published data for Alais, from 2.70 wt% (Grady et al. 1991) to 7.15 wt% C (Wright et al. 1986). In addition to these variations in C and N abundance and isotopic composition within Alais subsamples, Alais has been reported to have distinct and varied mineralogy compared with other CI chondrites (MacDougall and Kerridge 1976) that may contribute to the heterogeneous geochemistry observed. Figure 3 shows that, in general, previous data plots outside the errors of our analytical technique and outside the standard deviations of our replicate analyses. The variance in data observed in Fig. 3 for CI, CM, and CR chondrites is unlikely to be due wholly to analytical differences and more likely represents heterogeneity of the analyzed fragments. These data are consistent with suggestions that the measured differences are sourced from the meteorites themselves.

The cause of heterogeneities in C and N abundances and isotopic composition can be further explored through a comparison between meteorite falls and finds, since the

chemistry of meteorite finds may be readily influenced by terrestrial weathering processes (Sephton et al. 2004a). However, statistical comparisons are severely hindered by the relatively small size of the finds data set. Nonetheless, when comparing individual falls with finds from the same petrologic group, there are no obvious differences in standard deviations between the falls and finds data sets. This suggests that terrestrial weathering processes do not significantly influence bulk C and N abundances and isotopic compositions and that they are indigenous to each meteorite.

Due to the variance in published data brought about by indigenous heterogeneities, it is appropriate to use our own reproducible data set for further interpretations.

### Bulk C and N Abundances, Isotopic Compositions, and Classification

It is useful to investigate whether the commonly accepted group classification scheme for carbonaceous chondrites defined by Van Schmus and Wood (1967) is reflected in the results obtained in this study. C and N abundances and isotopic compositions may define the chondrite groups to the extent that these elements could be used for the classification of new chondrites. Student's  $t$ -tests (Table 3) were carried out to assess the statistical significance of any differences between the mean bulk data obtained for each petrologic group. On the basis of the data obtained, the CK chondrites could not be distinguished from the CV group, perhaps supporting proposals that the CK



Table 4. Calculated mean values for each parameter measured for every meteorite. Group means are shown in rows in bold. Columns labeled “No. of  $\sigma$ ” show the number of standard deviations ( $\sigma$ ) away from the group mean displayed by the individual meteorites. Greater than  $2\sigma$  are considered anomalous to their petrologic group.

Sample	Type	C (wt%)	No. of $\sigma$	$\delta^{13}\text{C}$ (‰)	No. of $\sigma$	N (wt%)	No. of $\sigma$	$\delta^{15}\text{N}$ (‰)	No. of $\sigma$
Orgueil	CI1	4.88	0.09	-17.5	0.95	0.53	1.15	+44.6	0.96
Alais	CI1	5.40	0.95	-4.60	0.10	0.20	0.55	+52.0	0.08
Tagish Lake	CI2	4.25	1.04	+12.73	1.05	0.19	0.60	+61.511	1.04
CI group mean, $n = 3$		4.83		-3.12		0.31		52.68	
Cold Bokkeveld	CM2	2.16	0.72	-8.10	0.21	0.08	1.12	+27.1	0.41
Nogoya	CM2	2.40	0.33	-8.10	0.21	0.11	0.11	+21.1	0.83
Mighei	CM2	4.05	2.34	-11.4	0.96	0.13	0.62	+28.3	0.32
Murray	CM2	2.25	0.57	-2.62	1.04	0.10	0.33	+48.0	1.08
Murchison	CM2	2.70	0.15	-1.61	1.27	0.14	1.02	+52.7	1.42
Erakot	CM2	2.60	0.01	-13.8	1.52	0.12	0.48	+15.7	1.22
Kivesvaara	CM2	2.62	0.76	-13.9	0.84	0.10	0.79	+19.9	0.94
CM group mean, $n = 6$		2.68		-8.50		0.11		+30.4	
Al Rais	CR2	2.74		-11.7		0.19		+103	
Renazzo	CR2	1.25		-5.29		0.09		+157	
CR group mean, $n = 2$		2.00		-8.47		0.14		+130	
Kainsaz	CO3	0.79	0.80	-21.0	0.30	0.04	0.19	-11.2	0.44
Felix	CO3	0.83	0.96	-23.9	0.41	0.09	1.13	-8.80	0.04
Lancé	CO3	0.65	0.24	-21.7	0.13	0.03	0.07	-7.43	0.32
Ormans	CO3	0.24	1.33	-19.0	0.79	0.01	0.60	-10.5	0.31
Warrenton	CO3	0.44	0.56	-24.1	0.46	0.02	0.31	-6.81	0.44
Colony	CO3	1.29	0.25	-15.2	1.73	0.09	0.15	+14.8	0.94
CO group mean, $n = 6$		0.71		-20.8		0.05		-4.98	
Kaba	CV3	1.36	0.31	-15.7	0.82	0.07	1.37	-9.22	1.07
Leoville	CV3	1.13	0.79	-13.6	1.52	0.04	0.14	-17.1	1.16
Vigarano	CV3	1.50	0.72	-17.0	0.32	0.02	0.58	-15.9	0.22
Grosnaja	CV3	0.71	1.51	-22.1	1.53	0.02	0.61	-18.0	0.62
Allende	CV3	0.27	2.75	-18.6	0.26	0.01	0.96	-18.6	0.74
CV group mean, $n = 5$		1.00		-17.4		0.03		-15.8	
Maralinga	CK4	0.27		-24.0		0.02		15.8	
Karoonda	CK4	0.07		-31.5		0.01		-28.4	
CK group mean, $n = 2$		0.17		-27.8		0.01		-6.32	
Coolidge	C4	0.20		-17.4		<0.01		-12.6	

chondrites should not be classified as distinct from CV chondrites (Greenwood et al. 2004). However, statistical analysis could not be carried out for the CK or CR groups due to the small number of samples in those groups. The data in Table 3 indicates that the CI group was distinct in its C abundance and  $\delta^{15}\text{N}$  composition. The CM group was statistically different from all other groups in terms of mean wt% C, wt% N,  $\delta^{13}\text{C}$ , and  $\delta^{15}\text{N}$ . The CO and CV chondrites could not be separated into statistically significant groups using either mean elemental abundance or mean isotopic compositions of individual meteorites.

Only two samples were identified as being statistical outliers to their petrologic groups as defined by the standard deviations of each group (Table 4); both Allende (CV) and Mighei (CM) were anomalous to their groups for C abundance. Allende's C abundance was significantly lower

than that of other CV chondrites, consistent with the results of previous studies (Fig. 3) (McSween 1977). All other chondrites fall within  $2\sigma$  of the mean of the established chondrite groups, verifying their inclusion based on statistical analyses.

With these anomalies removed, the t-tests were repeated, revealing a significant difference between CI and CV groups in their  $\delta^{15}\text{N}$  values, which had previously been obscured. However, even with the Allende and Mighei data removed, it was still not possible to discern between the CO and CV groups, suggesting that C and N abundances and isotopic compositions should not be used to distinguish between these two groups. A tentative use of C and N abundances and isotopic compositions for classification of the remaining groups is recommended on the basis of student's t-test analyses.

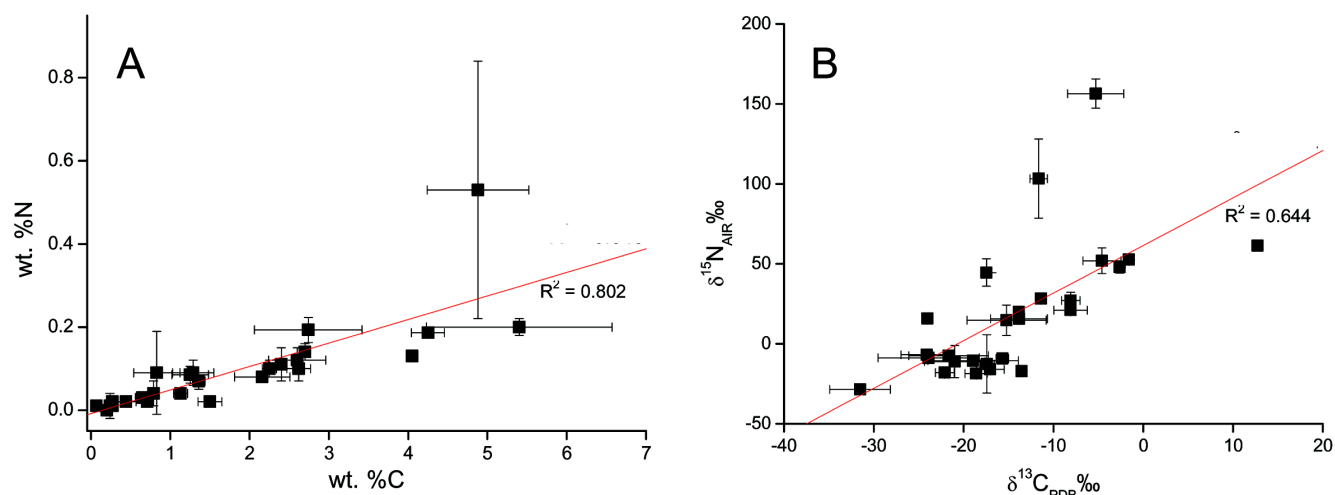


Fig. 4. a) Mean carbon versus mean nitrogen abundances (wt%) for all carbonaceous chondrites analyzed. A trend is displayed between the two measured parameters with an  $R^2$  value of 0.802. b) Carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) isotopic compositions for all carbonaceous chondrites analyzed. A trend is displayed between the two parameters with an  $R^2$  value of 0.644.

### Bulk Chemistry and Preterrestrial Alteration

By establishing that the bulk C and N abundances and isotopic compositions can be used for some classification purposes and that the means of the replicate analyses obtained in this study are representative of each chondrite, it is possible to examine this large data set in terms of chemical processes that may have influenced chondrite bulk composition.

#### Broad Trends

Chondrites of low petrologic types (1 and 2) display elevated C and N abundances and  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values; chondrites of higher petrologic types (3 and 4) display lower abundances and isotopic compositions (Figs. 1 and 2). This suggests a relationship between bulk C and N abundances and isotopic compositions and petrologic type. The CR chondrites (type 2) are an exception to this as they show anomalously heavy  $\delta^{15}\text{N}$  relative to all other chondrite groups. Elevated C and N elemental abundances in CI (type 1) chondrites may suggest a high relative abundance of C- and N-bearing material during primary accretion or the introduction of these elements during secondary processing.

The classification of chondrites into petrologic types is based on the mineralogical and chemical evidence that carbonaceous chondrites have experienced varying degrees of preterrestrial, asteroidal processing. The decrease in C and N abundance and  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values with increasing petrologic type suggest that parent body processing may play a significant role in influencing bulk chondrite chemistry. Published data for  $\delta^{13}\text{C}$  does not reflect this trend; however, the same conclusion would be valid for the observations of Fig. 3b where CI, CM, and CR chondrites contain aqueously generated,  $^{13}\text{C}$ -enriched carbonates which elevate their  $\delta^{13}\text{C}$

values. However, chondrites of types 1 and 4 represent end members of the classification scheme, and it is believed that organic matter in type 2 chondrites represent the most primitive and least altered among the carbonaceous chondrites (Alexander et al. 1998; Sephton et al. 2003). Therefore, there must be processes operating on the asteroidal parent body to invoke the trends observed over the entire petrologic type.

A positive trend between C and N abundance ( $R^2 = 0.643$ ) and  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  ( $R^2 = 0.414$ ) (Fig. 4) suggests that these elements reside in a common host. However, there is much scatter on the isotope trend, consistent with the multicomponent nature of these carbonaceous chondrites and the variance seen in Figs. 1, 2, and 3.

Thermal alteration (e.g., types 3 and 4) can result in oxidation and redistribution of indigenous organic material (Martinez and Escobar 1995) and a decrease in C and N abundances as observed (Alexander et al. 1998; Kung and Clayton 1978). Aqueous processing (e.g., types 1 and 2) results in high overall abundances of C and N (Figs. 1 and 2), possibly indicating the synthesis, accumulation, or preservation of organic material during aqueous processing (Pearson et al. 2002) and the growth of carbonates.

The CR chondrites show anomalous  $\delta^{15}\text{N}$  values, indicating a greater abundance of  $^{15}\text{N}$ -rich material than in other carbonaceous chondrites. Such enrichments are characteristic of an interstellar source (Becker and Epstein 1982) and suggest CR chondrites may contain more  $^{15}\text{N}$ -rich interstellar material than other carbonaceous chondrites. This assertion supports proposals that the CR chondrites contain the most primitive organic material, relatively unaffected by parent body processing (Alexander et al. 1998; Pearson et al. 2006). For the remaining carbonaceous chondrite groups, increasing alteration (thermal or aqueous) may

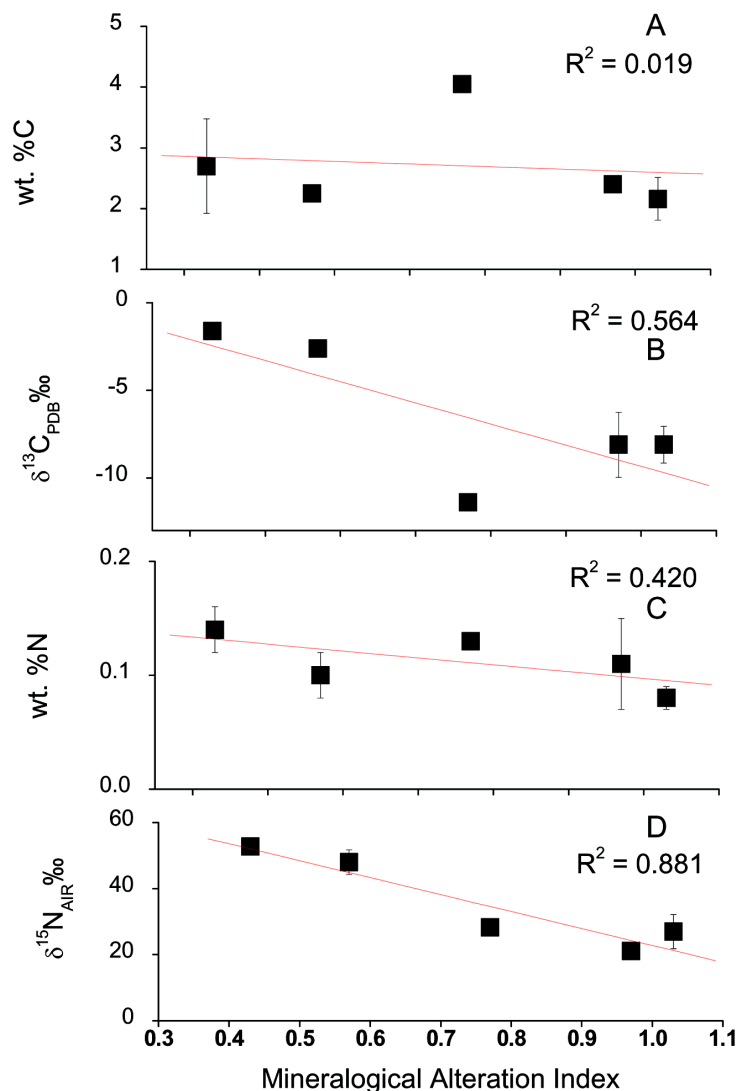


Fig. 5. Mean CM2 chondrite data obtained in this study plotted against mineralogical alteration index (MAI) as defined by Browning et al. (1996). The MAI was developed to illustrate an order of increasing alteration: Murchison (MAI = 0.43) < Murray (0.57) < Mighei (0.77) < Nogoya (0.97) < Cold Bokkeveld (1.03). Error bars denote standard deviations as defined in Table 1.

remove a  $^{13}\text{C}/^{15}\text{N}$ -rich component from the samples (Sephton et al. 2003, 2004b) resulting in decreasing  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in the more altered chondrites. These  $^{13}\text{C}/^{15}\text{N}$ -rich components may contribute to the volatile or soluble organic fraction (Sephton et al. 1998, 2003, 2004b) in type 1 and 2 chondrites but may be lost during the thermal metamorphism of type 3 and 4 chondrites, consistent with Figs. 1, 2, and 3. Such a scenario may explain how a potentially primitive starting material such as Renazzo (CR2), which has undergone only mild aqueous activity, is low in soluble organic material and has an enriched N isotopic signature (Figs. 2 and 3) (Alexander et al. 1998).

In summary, carbonaceous chondrites of low petrologic type display elevated C and N abundances and elevated  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ . The converse is true of carbonaceous chondrites of

high petrologic type. CR chondrites are an exception to this as they display anomalous and enhanced  $\delta^{15}\text{N}$  relative to other carbonaceous chondrites, suggesting they preserve more interstellar material, which has been less affected by asteroidal processing.

#### Bulk Trends within Groups

It is appropriate to use this data set to investigate samples within each chondrite group in terms of published alteration sequences. Browning et al. (1996) proposed an alteration sequence for the CM2 chondrites based on hydrogen abundance and mineralogy (Murchison < Murray < Mighei < Nogoya < Cold Bokkeveld). This alteration sequence (mineralogical alteration index [MAI]) is broadly reflected in the results of this study (Fig. 5); the more altered CM

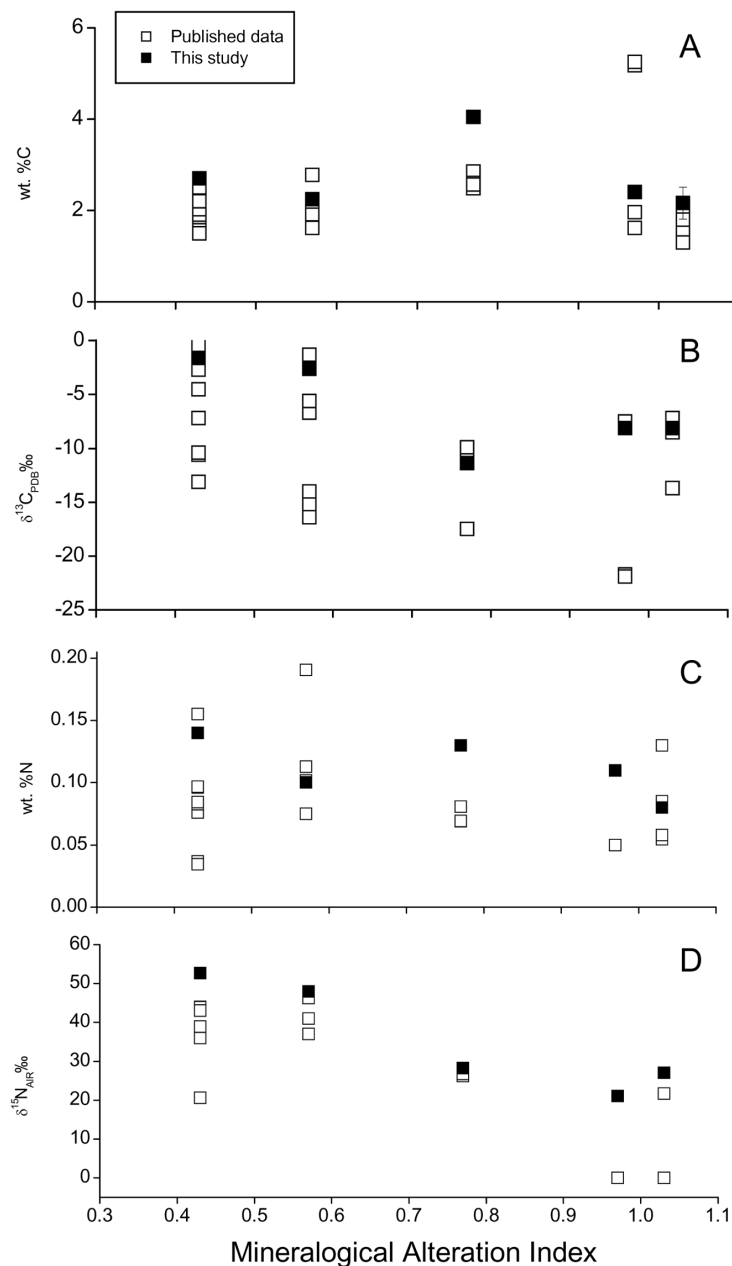


Fig. 6. A comparison of CM2 bulk carbon and nitrogen abundances and isotopic compositions from this study with published data (Appendix). Error bars denote standard deviations as defined in Table 1.

chondrites display decreasing N abundances and a shift to lighter  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values (Fig. 5). No such trend is observed for C abundance. When considering published values (Fig. 6), no trend is observed for C and N abundance or  $\delta^{13}\text{C}$  but these may be masked by large data variance. However, a trend may be inferred between published  $\delta^{15}\text{N}$  and MAI (Fig. 6d). The trends observed in our data and in the published data suggest that with increasing alteration (in this case aqueous processing),  $^{13}\text{C}$ - and  $^{15}\text{N}$ -rich components are removed from the chondrites, resulting in an accompanying decrease in N abundance. Sephton et al. (2004a, 2004b) have

suggested that  $^{13}\text{C}$ - and  $^{15}\text{N}$ -rich organic species may become liberated from the organic macromolecular material during asteroidal hydrous processing. These components were proposed to contribute to the soluble organic fraction. However, the decrease in bulk N abundance may infer that some components are removed rather than redistributed from the chondrite on processing.

The metamorphic sequence (with subgroups) for the CO3 chondrites (Colony [3.0] < Kainsaz [3.1] < Felix [3.2] < Ormans [3.3] < Lancé [3.4] < Warrenton [3.6]) defined by Scott and Jones (1990) is reflected in the results of this study

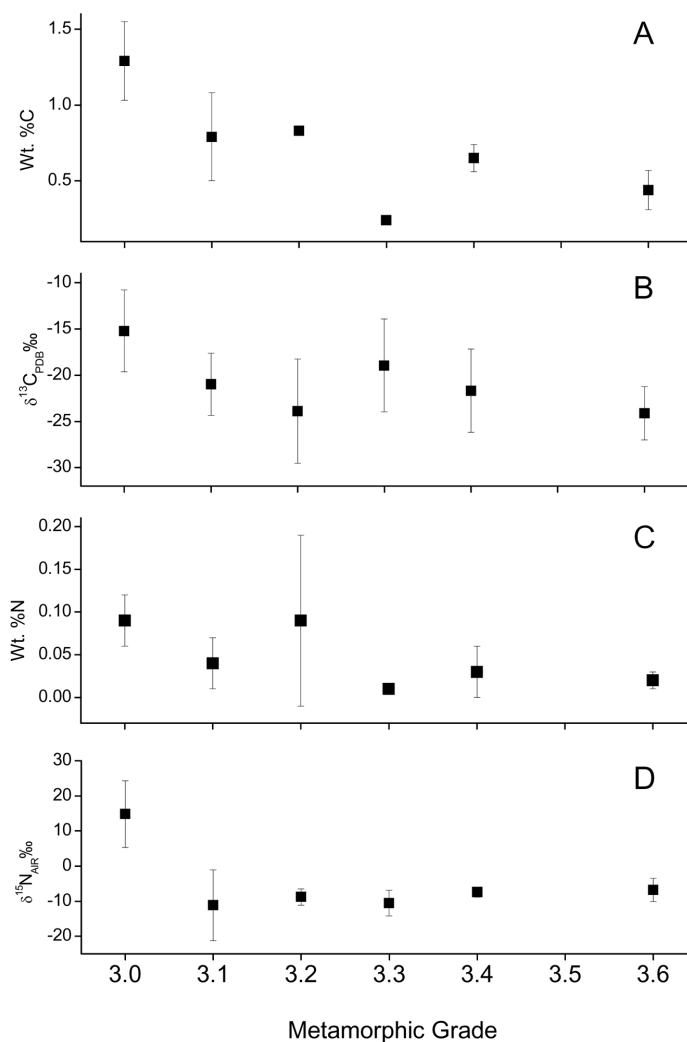


Fig. 7. Mean CO<sub>3</sub> chondrite data obtained in this study plotted against metamorphic grade as defined by Scott and Jones (1990): Colony (3.0), Kainsaz (3.1), Felix (3.2), Ormans (3.3), Lance (3.4), and Warrenton (3.6). Error bars denote standard deviations as defined in Table 1.

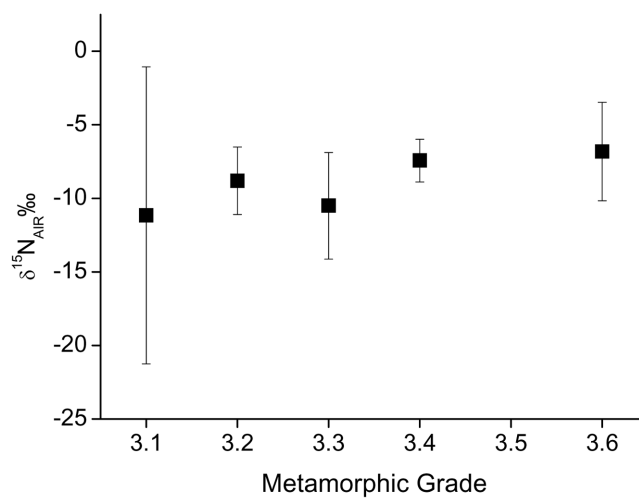


Fig. 8. CO<sub>3</sub> chondrite  $\delta^{15}\text{N}$  values from this study plotted against metamorphic grade (Scott and Jones 1996). Colony has been excluded from this plot to emphasize the trend between a move to heavier  $\delta^{15}\text{N}$  and increasing metamorphism seen in Fig. 7 for remaining CO<sub>3</sub> chondrites. Error bars denote standard deviations as defined in Table 1.

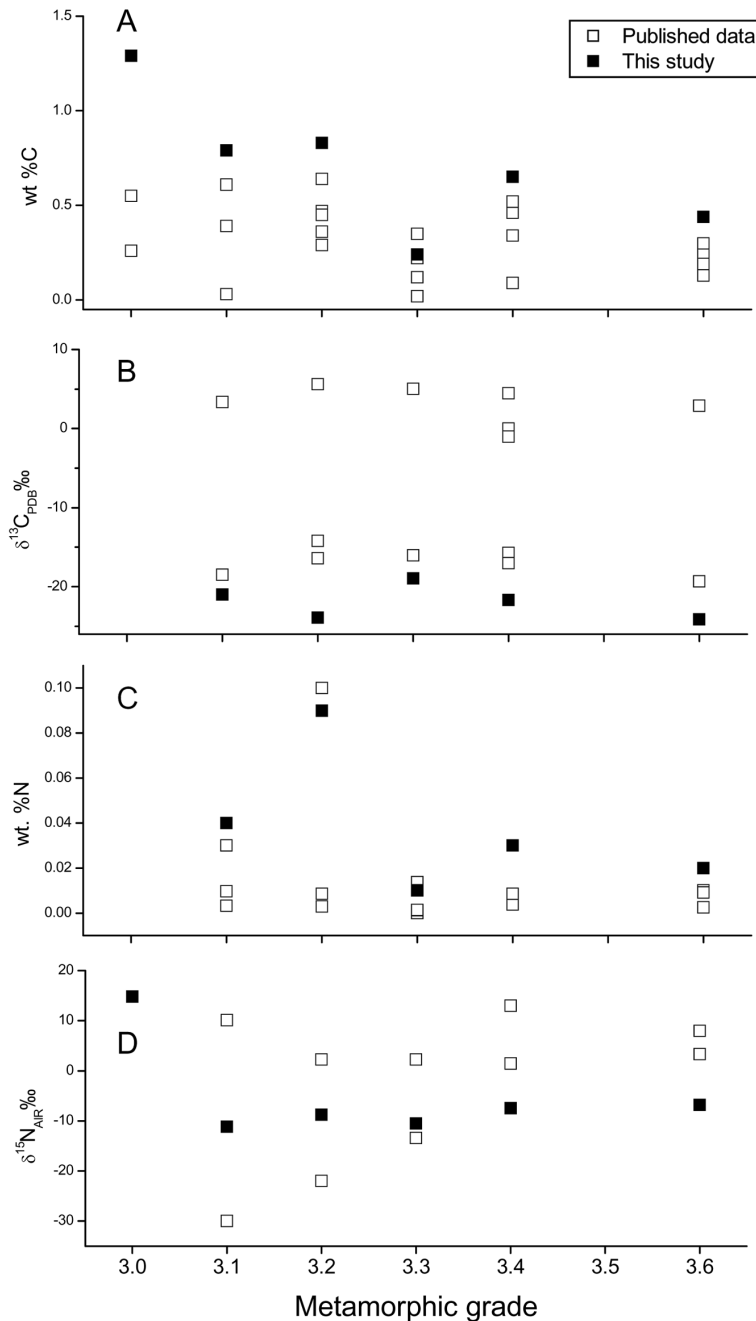


Fig. 9. A comparison of CO<sub>3</sub> bulk carbon and nitrogen abundances and isotopic compositions from this study with published data (Appendix) plotted against metamorphic grade (Scott and Jones 1996). Error bars denote standard deviations as defined in Table 1.

(Fig. 7); C abundances decrease and  $\delta^{13}\text{C}$  values become heavier with increasing metamorphic subgroup (Figs. 7a and 7b). This same trend has been identified in a recent study of CO<sub>3</sub> chondrite alteration (Greenwood and Franchi 2004). There is a much weaker trend for N abundance (Fig. 7d) with increasing subtype, and although not statistically different, Colony appears to display unusually high relative N abundances. The remaining CO<sub>3</sub> chondrites show a converse trend to the bulk data set (Fig. 8), also decrease within

increasing subgroup; however Fig. 7 displays the CO<sub>3</sub> chondrites, displaying a shift to heavier  $\delta^{15}\text{N}$  values with increasing metamorphic subgroup. This is contrary to the bulk trends over the entire data set (Fig. 2) and to the  $\delta^{13}\text{C}$  trends (Fig. 7b). Published data for the CO<sub>3</sub> chondrites (Fig. 9) do not display any trends for this group due to the large variance in data points.

Although it has been suggested that  $^{15}\text{N}$ -rich organic entities are released during the metamorphism of chondritic

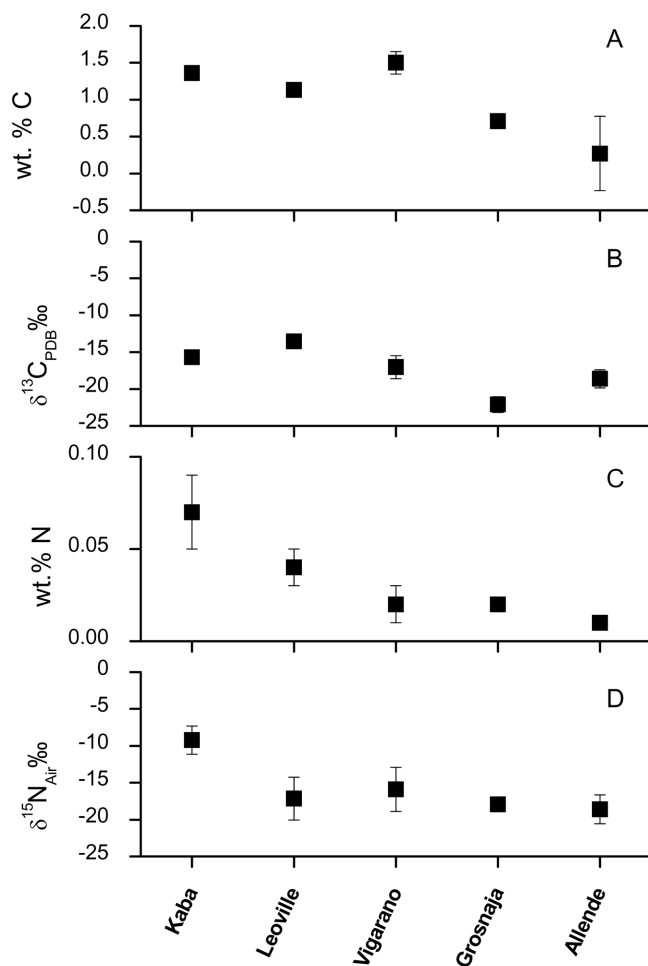


Fig. 10. Mean CV3 chondrite data obtained in this study grouped according to the thermal alteration sequence proposed by Bonal et al. (2004). Error bars denote standard deviations as defined in Table 1.

macromolecular material (Sephton et al. 2003, 2004b), our data suggests that either  $^{15}\text{N}$  is preferentially retained over  $^{14}\text{N}$  in CO3 organic macromolecular material or  $^{14}\text{N}$ -rich entities are liberated to leave a  $^{15}\text{N}$ -rich macromolecular structure. It has been suggested that the CO3 chondrites have experienced a complex parent body alteration history (Greenwood and Franchi 2004; Keller and Buseck 1990) encompassing both thermal and aqueous processing. Two-stage processing may result in the exhaustion of labile  $^{15}\text{N}$ -rich species available for release following the first alteration period and subsequent breakdown of the macromolecular network during a second alteration event liberating kinetically favored  $^{14}\text{N}$ -rich species.

The division of CV3 chondrites into oxidized (Grosnaja, Kaba, Bali, and Allende) and reduced (Vigarano and Leoville) subgroups is not reflected in the data obtained in this study consistent with previous studies of bulk CV chemistry (Kallemeyn and Wasson 1981). However, an

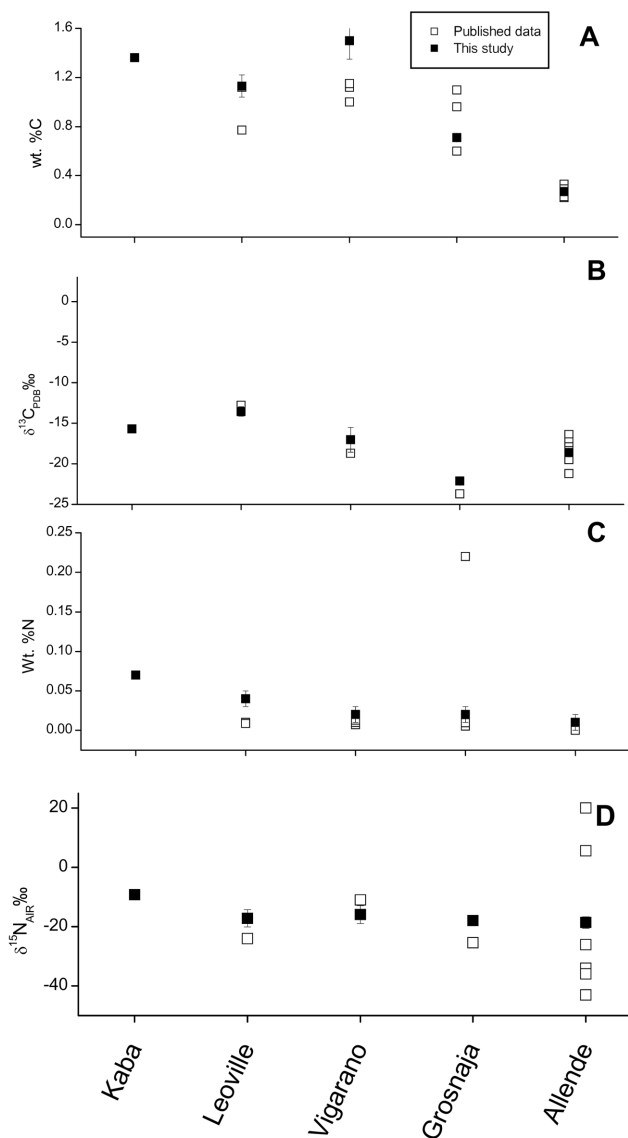


Fig. 11. A comparison of CV3 chondrite bulk carbon and nitrogen abundances and isotopic compositions from this study with published data (Appendix) grouped according to proposed thermal alteration sequence proposed by Bonal et al. (2004). Error bars denote standard deviations as defined in Table 1.

alteration scheme for CV3 chondrites was recently proposed on the basis of degree of thermal alteration of indigenous organic material (Leoville = Vigarano < Grosnaja = Mokoia < Allende) (Bonal et al. 2004). The C and N chemistries obtained in this study for CV3 chondrites broadly reflect this alteration scheme (Fig. 10), supporting the proposal that bulk C and N chemistry in CV3 chondrites reflects changes in organic material with progressive alteration. These trends are also consistent with the broad trends observed over the entire petrologic range (Figs. 1 and 2). However, the variance of published data precludes observations of these trends.

In summary, the results of this study reflect published alteration sequences for carbonaceous chondrites (Bonal et al. 2004; Browning et al. 1996; Scott and Jones 1990), implying that C and N abundances and isotopic compositions are useful indicators of preterrestrial alteration. This validates the use of these bulk measurements for studies of asteroidal processing.

### CONCLUSIONS

This study has used a sampling strategy whereby multiple subsamples of meteorites have been analyzed to discern the extent of inter- and intrameteorite heterogeneities. Both meteorite falls and finds have been compared and no significant differences between these two data sets were observed. This suggests that terrestrial weathering/contamination is not a significant influence on bulk C and N abundances and isotopic compositions. By analyzing chondrites over the entire range of carbonaceous chondrite petrologic types (1 to 4), the effects of preterrestrial parent body alteration have been investigated. As seen in previous studies, C and N abundances decrease with increasing petrologic type, reflecting either variations in the primary material accreted or alteration of the constituents during secondary processing.

Over the entire petrologic range (types 1 to 4),  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values decrease with increasing petrologic type. On a smaller scale, this trend is reflected within the CV3 and CO3 chondrite groups. In the CM2 chondrites,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values increase with increasing aqueous processing (Murchison to Cold Bokkeveld; Browning et al. 1996). During alteration (aqueous or thermal),  $^{13}\text{C}$ - and  $^{15}\text{N}$ -rich units are liberated from the organic macromolecule (Sephton et al. 1998, 2003, 2004b). In petrologic types 3 and 4 these units are lost during thermal alteration (Sephton et al. 2003, 2004b), hence decreasing the bulk  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values. In petrologic types 1 and 2, these organic units are conserved as soluble organic entities (Sephton et al. 1998, 2003, 2004b) and contribute to a heavy isotopic signature. The generation of isotopically heavy carbonate species in these aqueously altered samples also contributes to heavy carbon stable isotopic compositions. The CR chondrites display a level of N enrichment not observed in any other carbonaceous chondrites. This may reflect the preservation of interstellar material and support the proposals that they are the most primitive carbonaceous chondrites and possibly representative of chondritic parent material.

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### APPENDIX

Table A1. Published C and N abundances and isotopic compositions for the carbonaceous chondrites analyzed in this study.

Sample	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)	References <sup>a</sup>		
Alais	CI1	2.70	-8.8			1		
		7.15	-14.9			2		
		3.19				3		
		3.19		0.290		4		
		4.46	-10.3		31	5		
Orgueil	CI1			0.148	46.2	6		
		3.30	-13.1			1		
		3.10				7		
			-15.6			8		
			-51.9			8		
			-9.7			8		
			-10.9			9		
		2.72	-15.6	0.136	39	10		
			-11.6			11		
			-11.4			12		
		2.8				13		
		3.33				3		
		3.45		0.318		14		
		3.48		0.318		15		
		3.33		0.240		4		
6.47	-16.7			2				
4.35	-15.2	1.99	32	16				
Tagish Lake		5.4	24.3			16		
		5.81	24.4	1.218	67	16		
		1.30				7		
Cold Bokkeveld	CM2	1.58	-8.5			1		
				0.055	21.7	17		
				0.058	21.7	17		
		1.52				18		
		1.30				4		
			-7.2			11		
		2.00	-13.7	0.085	0.035	5		
		1.80		0.130		15		
		Erakot	CM2	2.14		0.260		15
					-7.6			11
2.17				0.170		15		
Kivesvaara	CM2	1.77	-11.7			1		
		2.29	-16.1			2		
Mighei	CM2	2.85	-10.3	0.0807	26.2	6		
		2.48				7		
			-10.3			11		
		2.60	-9.9	0.069	27	12		
				0.069	26.8	17		
		2.57		0.130		15		
2.57	-17.5			2				

Table A1. *Continued.* Published C and N abundances and isotopic compositions for the carbonaceous chondrites analyzed in this study.

Sample	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)	References <sup>a</sup>
Murchison	CM2	1.76	-4.5	0.076	39	11
		2.15	-0.5	0.085	43	19
		2.50	-7.2	0.083	44	20
		1.56	-10.6	0.037	36	21
				0.035	43	21
		1.85				22
		1.50	-13.1			1
		2.04	-1.9			1
		2.20	-10.4			2
		1.91		0.096		23
				0.083	43.9	17
				0.085	43	7
		2.03		0.155		15
		2.2	-2.7	0.0968	20.6	24
Murray	CM2	2.20	-6.7			1
		2.78				4
		2.02	-15.2			2
				0.102	46.3	6
		1.92	-1.3	0.075	37	11
		2.22	1.2	0.113	41	11
			-16.4			8
			-14			8
			-16.4			8
			-5.6			11
		2.19		0.191		15
Nogoya	CM2	1.62				25
		5.18	-21.7			26
		5.25	-21.9			26
		1.62				25
Al Rais	CR2	1.96	-7.5	0.050	0.013	5
		1.42	-13.3			1
		2.49				3
		2.49		0.190		4
Renazzo	CR2	2.70	-12.6	0.077	136	27
				0.052	172	17
				0.051	170	17
		1.50	-10.2	0.080	190	11
		1.70	-9.9	0.050	175	28
		1.80	-9.1			1
		1.40	-7	0.048	175	27
		1.70	-9.9	0.050	175.3	28
		1.56	-10.2	0.080	190	11
		1.44				29
		1.40	-7	0.048	175	27
Colony Felix	CO3	0.55				30
		0.47	-16.4			12
		0.45				7
		0.36				31
		0.64	-14.2	0.003	-22	5
Kainsaz	CO3	0.64		0.008		15
		0.39	-18.5	0.003	-30	5
		0.61		0.010		15
Lance	CO3	0.34	-15.7			12
		0.46	-17	0.004	13	5
			0			8

Table A1. *Continued.* Published C and N abundances and isotopic compositions for the carbonaceous chondrites analyzed in this study.

Sample	Type	C (wt%)	$\delta^{13}\text{C}$ (‰)	N (wt%)	$\delta^{15}\text{N}$ (‰)	References <sup>a</sup>
			-1			8
		0.46				4
Ormans	CO3	0.52		0.009		15
				0.001	-13.4	17
		0.35				3
		0.22		0.014		15
		0.12	-16	0.002		5
		0.35				4
Warrenton	CO3	0.25	-19.3	0.003	8	5
		0.19				7
		0.30		0.009		15
Allende	CV3	0.27	-16.4	0.004	-43	19
		0.30	-17.3	0.002	5.6	6
				0.002	-26	32
				0.002	-34	32
				0.002	-36	32
		0.33	-21.2	0.002	20	5
				0.002	-43	17
		0.22				33
		0.27				33
		0.25	-17.9			33
		0.23	-18.4			33
		0.29				34
		0.25		<0.001		15
		0.23				35
			-19.5			36
Grosnaja	CV3			0.005	-25.4	17
		0.96				3
		0.60		0.017		15
		0.96		0.220		4
		1.10	-23.7	0.010		5
Leoville	CV3	0.77	-12.8	0.010	-24	5
		1.12				4
				0.009	-24	17
		0.77				4
Vigarano	CV3	1.00	-18.7	0.007	-11	5
		1.12		0.010		4
		1.15		0.013		15
Karoonda	CK4			0.001	2.3	17
			-23.3			8
				<0.001	-33.8	37
Coolidge	C4-UNGR			0.0015	0.7	17
		0.19				38
		0.15				15

<sup>a</sup>Published bulk elemental data taken from: 1) Grady et al. 1991; 2) Wright et al. 1986; 3) Mason and Wiik 1962; 4) Wiik 1969; 5) Kerridge 1985; 6) Injerd and Kaplan 1974; 7) Wiik 1956; 8) Krouse and Modzeleski 1970; 9) Yang and Epstein 1983; 10) Robert and Epstein 1982; 11) Pillinger 1984; 12) Boato 1954; 13) Fredriksson and Kerridge 1988; 14) Anders and Grevesse 1989; 15) Gibson et al. 1971; 16) Grady et al. 2002; 17) Kung and Clayton 1978; 18) Wöhler 1858; 19) Chang et al. 1978; 20) Kvenvolden K. et al. 1970; 21) Swart et al. 1983; 22) Jarosewich 1971; 23) Fuchs et al. 1973; 24) Sephton et al. 2003; 25) Friedheim 1888; 26) Bunch and Chang 1980; 27) Bischoff et al. 1993; 28) Grady et al. 1983; 29) Mason 1963; 30) Rubin and Grossman 1985; 31) Merrill 1901; 32) Thiemens and Clayton 1981; 33) Jarosewich et al. 1987; 34) Clarke et al. 1970; 35) Murae and Masuda 1990; 36) Grady et al. 1981; 37) Sugiura and Zashu 1995; 38) Jarosewich 1966.