

THE DISTRIBUTION OF CARBON IN C1 TO C6 CARBONACEOUS CHONDRITES

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Abstract: The carbon content and $\delta^{13}\text{C}$ of carbon in ten carbonaceous chondrites, spanning petrologic grades 1 to 6, have been determined. There is a gradual change in the nature of the major carbonaceous components from C1 to C6 chondrites: C1 and C2 samples contain carbon as organics, whereas in higher petrologic types, carbon is predominantly amorphous or graphitic. This transition is consistent with carbon in C3 and C4 samples being formed either by dehydrogenation of organic materials during metamorphism on the parent body, or nebular heating followed by accretion at higher temperatures than prevailed during formation of C1-2 meteorites. In addition to a major carbonaceous component, ^{13}C -rich interstellar grains are found in C1 and C2 samples and, to a much lesser extent, CV3 meteorites. CO3 and C4-6 meteorites do not appear to contain ^{13}C -rich materials, a distribution controlled by primary accretion processes and not a result of secondary effects on parent-bodies. However, among the C1 to C3 meteorites aqueous activity might have acted to re-distribute ^{13}C -rich grains by either concentrating them into C1 meteorites, or alternatively by transporting them into the source region of CV3 samples.

1. Introduction

Carbonaceous chondrites are chemically primitive meteorites, with C1 samples in particular having a general similarity in composition, for all elements other than the most volatile, to the present day solar photosphere (*e.g.*, HOLWEGER, 1977; ANDERS and EBIHARA, 1982; ANDERS and GREVESSE, 1989). Notwithstanding their primitive chemistry, carbonaceous chondrites have undergone varying degrees of secondary, post-accretional, alteration on the parent bodies (*e.g.*, MCSWEEN, 1979). C1 and C2 types have suffered extensive low-temperature (<400 K) aqueous processing (*e.g.*, DU FRESNE and ANDERS, 1962; BOSTRÖM and FREDRIKSSON, 1966; FREDRIKSSON and KERRIDGE, 1988), whilst C4 to C6 types have been subjected to static thermal metamorphism. Higher petrologic grades, such as Mulga West, a C5/6 (BINNS *et al.*, 1977), may have experienced temperatures up to 1100 K. Type 3 carbonaceous chondrites might be expected to be the most pristine samples (MCSWEEN, 1979); however even these meteorites exhibit chemical fractionation trends (*e.g.*, KALLEMEYN and WASSON, 1981), and both Warrenton, CO3, (KERRIDGE, 1972) and Allende, CV3, (MIURA *et al.*, 1987) show evidence for aqueous alteration.

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Nevertheless, the least altered carbonaceous chondrites (*i.e.* C3 samples) give perhaps the best insight into the nature of solar system materials; these meteorites are made up from chondrules, irregular aggregates and calcium-aluminium rich inclusions (all of which are thought to have formed at high temperatures) set in a fine-grained dark matrix considered to be of much lower temperature origin (*e.g.*, WOOD, 1988). Carbonaceous chondrites of higher petrologic types than C3 samples show features consistent with the annealing of such an assemblage, whilst lower grades have characteristics (*e.g.* solar wind irradiation, brecciation) which suggest a regolith environment. In addition to petrological differences, accompanying variations in major and trace element and oxygen isotopic composition are interpreted as having been established at the time of meteorite formation. Whether these variations reflect (i) the existence of contemporaneous nebular reservoirs of distinctly different elemental/isotopic abundance, (ii) incomplete sampling, by the presently known carbonaceous chondrite groups, of a compositional continuum, or (iii) temporal changes in nebular chemistry, is an unresolved issue. Regardless, study of the different chemical groups and /or various petrologic grades of carbonaceous chondrites has the potential to provide information which might differentiate amongst some of these possibilities.

A subject currently attracting much attention concerns interstellar grains. These predominantly carbon-bearing components are generally recognized by an anomalous carbon isotopic signature, deriving from a primary nucleosynthetic feature, which has survived the passage into, and processing within, the solar system (*e.g.*, SWART *et al.*, 1983a; ZINNER *et al.*, 1987; TANG *et al.*, 1988). Grains with normal carbon isotopic signatures are somewhat more difficult to detect, but may be revealed by an unusual isotopic composition in an element other than carbon (*e.g.*, LEWIS *et al.*, 1983). Up to now, the quest for interstellar grains has been directed towards those meteorite samples which are (a) deemed to be most likely to contain such materials, and (b) available in the most plentiful supply. The work presented herein is of a much more general nature and was undertaken in an attempt to assess the distribution of interstellar grains within type 1 to 6 carbonaceous chondrites. An indication of the presence of interstellar grains is assessed, in this study, by the observation of elevated $^{13}\text{C}/^{12}\text{C}$ ratios.

Carbonate minerals enriched in ^{13}C were identified in primitive carbonaceous chondrites over 25 years ago (CLAYTON, 1963). Although CLAYTON alluded to the possibility that the carbonates may have a nuclear origin, fractionation of carbon isotopes between oxidized (*i.e.* carbonates) and reduced (*i.e.* organic materials) forms of carbon has been explained as a result of solar system processing through Fischer-Tropsch-type reactions (LANCET and ANDERS, 1970). A more detailed survey of carbonates in primitive meteorites has now shown that the range in ^{13}C -enrichment in C1 and C2 samples is equivalent to *ca.* 25 to 80‰ (GRADY *et al.*, 1988a) but this is still within the bounds of chemical fractionation. However, in addition to ^{13}C -rich carbonates, it is now well established that carbonaceous chondrites contain highly ^{13}C -enriched carbon (*e.g.*, CARR *et al.*, 1983; SWART *et al.*, 1983a; YANG and EPSTEIN, 1984; HALBOUT *et al.*, 1986; TANG *et al.*, 1988; ASH *et al.*, 1989). This material remained unnoticed during 25–30 years of study of carbonaceous chondrites, primarily on account of the co-existence of large amounts of isotopically normal carbon oc-

curing as macromolecular organic constituents somewhat analogous to terrestrial kerogen (the organic constituents of carbonaceous chondrites are reviewed by HAYATSU and ANDERS, 1981; MULLIE and REISSE, 1987). The ^{13}C -rich components were eventually identified due to their inherent stability and resistance to oxidation. SWART *et al.* (1982) showed that carbon released at high temperatures ($T > 800^\circ\text{C}$) during stepped combustion was isotopically heavier than the bulk sample for Allende; KERRIDGE, working independently, analyzed an HCl-HF resistant fraction of Murray (KERRIDGE, 1982), and found at the highest temperature of the experiment (850–1050°C) carbon with $\delta^{13}\text{C} \sim +110\text{‰}$, which was unusual, but not impossible, for a solar system fractionation process. KERRIDGE considered that this ^{13}C -enriched carbon was probably associated with the macromolecular organic material. Pursuing the ^{13}C -rich material further, SWART *et al.* (1983a) analyzed a number of residues prepared from Murchison by the use of mineral acids and oxidizing reagents; the extent of ^{13}C -enrichment was demonstrated to be far greater than could be reasonably explained by isotopic fractionation during solar system processing. Several isotopically heavy carbonaceous components, with $\delta^{13}\text{C}$ up to $+1100\text{‰}$, were considered to be individual minor constituents having a primary nucleosynthetic origin, and unrelated to the macromolecular organic material. It was postulated that the isotopically heaviest carbon was C_β (SWART *et al.*, 1983a), the carrier of s-Xe, a noble gas component showing the isotopic signature of the slow neutron-capture process, as calculated by CLAYTON and WARD (1978).

A number of studies aimed at elucidation of the origin and distribution of ^{13}C -rich carbon in C1 and C2 carbonaceous chondrites have followed in the wake of these initial discoveries. CARR *et al.* (1983) confirmed that there are at least three ^{13}C -rich components in C2 chondrites. One of these, referred to as C_α , is thought to be amorphous carbon and the host of Ne-E(L) (CARR *et al.*, 1983). Following on from a preliminary ion-probe investigation by ZINNER and EPSTEIN (1987), which showed that an additional ^{13}C -rich component had $\delta^{13}\text{C}$ of up to $+7000\text{‰}$, it was discovered by ZINNER *et al.* (1987) that there were two ^{13}C -rich silicon carbide components—one, C_β , is fine-grained with $\delta^{13}\text{C} \approx +1000$ to $+1500\text{‰}$ and the other, C_ϵ , is more coarse with $\delta^{13}\text{C}$ in excess of $+8000\text{‰}$, and may be associated with Ne-E(H). More recent work by TANG *et al.* (1989) demonstrated the complexity of SiC, the silicon isotopic record of large grains demanding at least four separate stellar sources.

The preliminary findings of isotopically anomalous carbon in Allende by SWART *et al.* (1982, 1983a) have now been consolidated by a more detailed study (ASH *et al.*, 1989). There are two ^{13}C -enriched carbonaceous components in Allende, one with $\delta^{13}\text{C}$ of $\sim +340\text{‰}$ and the other of greater than $+530\text{‰}$. However, distribution of ^{13}C -rich carbon in carbonaceous chondrites of other chemical groups, particularly higher petrologic types, has thus far not been appraised. The present study was undertaken in order to assess, in the most general of ways, whether any systematic trends could be observed in terms of this distribution.

2. Experimental

The carbon contents and isotopic compositions of the meteorites studied herein have been assessed using a combination of incremental heating (stepped combustion)

and analysis using static mass spectrometry. To this end, samples of carbonaceous chondrites (100 to 200 mg) were initially crushed in a stainless steel pestle and mortar to a grain size of $<50 \mu\text{m}$. Aliquots of approximately 20 mg were then treated with HCl (12 M at 18°C for 24 h). The residues were subsequently recovered after at least four washings with distilled water (until neutral), then a final rinse in methanol. The HCl-treatment served several purposes: the acid (1) destroys and ^{13}C -rich carbonates present which would interfere during stepped combustion analysis, (2) removes sulphides, which give SO_2 on combustion and make the task of CO_2 purification more difficult, (3) allows better access of oxygen for combustion purposes, and (4) produces weight-losses of *ca.* 60% so that smaller samples may be manipulated into the extraction system. The last mentioned is extremely important, since large amounts of volatile-rich, finely-powdered meteorite samples are difficult to evacuate, and could be partially lost from the combustion vessel to other parts of the glass-line during pump-down. Unfortunately, the effect of HCl on carbonaceous chondrites is poorly constrained: minerals which are known to be soluble include troilite/pyrrhotite, iron-nickel metal, magnetite, olivines, carbonates and sulphates. However, removal of these minerals alone does not account for the weight-loss observed. The effects of HCl on components such as tochilinite (MACKINNON and ZOLENSKY, 1984) and cronstedtite or FESON (TOMEOKA and BUSECK, 1985) are unknown, whilst reactions with clay minerals and other phyllosilicates are highly dependent upon the chemical composition of the minerals.

Between 2 and 4 mg of the HCl-resistant residues were weighed in small, previously-baked (1200°C in air) quartz buckets according to the procedure of CARR *et al.* (1986). After loading into an all-glass gas extraction line, the sample was evacuated in two stages. Firstly, it was pumped from atmosphere down to *ca.* 1 Torr using a capillary leak connected to the pumping system, to avoid the problems associated with fine powder mobility referred to above. The sample was then pumped to high vacuum ($\sim 10^{-8}$ Torr) using a conventional arrangement of high conductance valves, the whole procedure taking typically 30 min.

Carbon was extracted from the samples by stepped combustion, a process of incremental heating under oxygen described more completely by SWART *et al.* (1983b) and CARR *et al.* (1986). For the lower petrologic type samples, large amounts of carbon were released at low temperatures, hence several combustions at a single temperature were carried out in the temperature range $500\text{--}550^\circ\text{C}$ to ensure its complete removal. It was not always possible to determine the yield of this carbon, since the amounts of gas were sometimes too large to measure on the capacitance manometer employed in the system. In these cases, the CO_2 produced in the low temperature steps was pumped away without isotopic measurement. For Orgueil, Murchison, Vigarano, Coolidge and Mulga West, the bulk carbon content and $\delta^{13}\text{C}$ was ascertained by taking a second aliquot of the HCl-resistant residue and combusting it with spectroscopic-grade copper (II) oxide (GRADY *et al.*, 1982). For the higher petrologic grade samples, with lower bulk carbon abundance, it was possible to perform stepped combustions from room temperature up to 1300°C . Even so, multiple combustions at intermediate temperatures were also employed to ensure effective removal of any low temperature organic carbon of indigenous or contaminant origin.

The carbon dioxide produced by combustion was quantified to ± 0.2 ng on a capacitance manometer, following which its isotopic composition was determined by analysis in a noble gas-type static mass spectrometer (CARR *et al.*, 1986). Isotopic composition is reported in the δ -notation relative to the PDB standard (*i.e.* $\delta^{13}\text{C}_{\text{PDB}}$, denoted simply as $\delta^{13}\text{C}$). Errors on $\delta^{13}\text{C}$ are $\pm 1\%$ on sample sizes greater than 10 ng of CO_2 .

3. Results

3.1. Major carbonaceous components

A summary of the carbon concentration and isotopic data for the ten carbonaceous chondrites studied is given in Table 1. In accordance with the original observations of BOATO (1954) and WIJK (1956) meteorites of low petrologic type (*i.e.* the chemical groups CI, CM and CR) have the highest carbon contents. The value of 2.31 wt% carbon calculated for the whole-meteorite sample of Orgueil compares with a measured bulk abundance of 3.30 wt% (GRADY, 1982) and 3.67 wt% obtained from a stepped combustion of the whole-rock (WRIGHT *et al.*, 1985). Published carbon abundances in Orgueil vary from 2.71% to 4.1% (BOATO, 1954; OTTING and ZAHNINGER, 1967; WIJK, 1969; SMITH and KAPLAN, 1970; GIBSON *et al.*, 1971; ROBERT and EPSTEIN, 1982) hence there is a fair degree of sample heterogeneity to be taken into account when comparing carbon concentrations. Heterogeneity notwithstanding, the amount of carbon lost on HCl-dissolution was approximately 1 wt%. Carbonaceous components lost during the acid-treatment include carbonates, which constitute 0.016 wt% in Orgueil (GRADY *et al.*, 1988a), and solvent-soluble organic material (determined as

Table 1. Summary of carbon abundance and isotopic data from combustion of ten carbonaceous chondrites.

Sample	Type	% wt loss	Total carbon ($T < 1300^\circ\text{C}$)			$T = 700\text{--}1300^\circ\text{C}$		
			$[\text{C}]_{\text{HCl}}^\dagger$ (wt%)	$[\text{C}]_{\text{WR}}^\dagger$ (wt%)	$\delta^{13}\text{C}$ (‰)	$[\text{C}]_{\text{HCl}}^\dagger$ (ppm)	$[\text{C}]_{\text{WR}}^\dagger$ (ppm)	$\delta^{13}\text{C}$ (‰)
Orgueil	CI1	64.3	6.47*	2.31	-16.7	39.25	14.01	+82.7
Murchison	CM2	57.6	2.20	0.93	-15.3	12.62	5.35	+155.6
			2.20*	0.93	-10.4			
Renazzo	CR2	49.8	1.34	0.67	-22.4	73.75	37.02	-5.2
Felix	CO3	49.2	0.33	0.17	-17.6	24.19	12.29	-10.4
Ornans	CO3	52.0	—	—	—	81.58	39.16	-17.0
Allende	CV3	56.0	—	—	—	48.96	21.54	-9.0
Vigarano	CV3	51.9	1.24	0.60	-18.5	14.82	7.13	-5.3
			1.59*	0.76	-17.3			
Karoonda	CO4	56.3	0.20	0.09	-31.6	29.08	12.71	-31.5
Coolidge	CV4	46.7	0.15	0.08	-13.7	413.29	220.28	-6.6
			0.37*	0.20	-17.0			
Mulga West	CV5/6	58.1	0.33	0.14	-26.8	25.42	10.65	-22.8
			0.76*	0.32	-24.9			

† HCl—Amount of carbon measured in HCl-resistant residue; WR—Amount of carbon in the whole-rock meteorite calculated from HCl data.

* Sealed tube combustion.

0.11 wt%, SMITH and KAPLAN, 1970). Similarly, for Murchison (whole-rock carbon content=1.56 wt%; SWART *et al.*, 1983a) and Renazzo (whole-rock carbon content=1.7 wt%; GRADY *et al.*, 1983) the amounts of carbon lost during acid-treatment far exceeds that which can be accounted for by dissolution of carbonates and solvent-soluble organics. This effect was first described by SMITH and KAPLAN (1970), who noted that unidentified volatile organics were lost during treatment with HF and HCl, precluding mass balance calculations. Hydrochloric acid is often used to remove carbonates from terrestrial sedimentary rocks with no apparent loss of macromolecular organic matter, kerogen. Notwithstanding the difference in formation mechanisms between terrestrial and meteoritic organic materials (*i.e.* biogenic *vs.* abiogenic), the two entities are considered analogous in many ways. Indeed, the term "m-kerogen" has been coined to describe that material present in meteorites (KERRIDGE, 1983). Thus, it appears that meteoritic macromolecular material reacts to HCl treatment in a fundamentally different way from terrestrial kerogen, resulting in loss of almost 50% of the total carbon budget. This observation is inconsistent with the aromatic nature of m-kerogen, which should render the material more robust to degradation by HCl than corresponding terrestrial materials. It is possible that very fine-grained material is removed during sample processing—a layer of organics is often observed floating on the surface of the acid (J. ARDEN, pers. commun.), which might be lost if great care is not taken. However, it is unlikely that 50% of the residue could be lost in this way. Another possibility is that the carbon exists as a trapped gas, although *ca.* 1 wt% of carbon as a trapped gas in Orgueil is also unlikely.

Seven carbonaceous chondrites of varying chemical group and petrologic grade were combusted in steps from room temperature upwards. Data for these samples are shown in Fig. 1 and summarized in Table 1. In all seven samples, the carbon is almost totally combusted by 700–800°C, however its actual combustion temperature varies from sample to sample. One parameter used to describe the combustive behavior of carbon is its geometric mean combustion temperature (T_m). This is calculated by assuming all the carbon comes from a single component— T_m represents the temperature at which 50% of the carbon has been liberated. This is preferable to comparing the temperature at which the maximum release of carbon is obtained, since the temperature increments of the experiment are selected by the operator, and thus have no direct bearing on the sample under investigation. The assumption of a single carbon component is questionable, since carbonaceous chondrites are known to contain a variety of carbon-bearing materials. However, in each meteorite sample, the carbon inventory is dominated by one specific form; appraisal of T_m gives a measure of the resistance to oxidation of this major component. The value of T_m is undoubtedly dependent on experimental conditions, and in this context the use of repeated combustions at a single temperature may have acted to lower T_m . It is impossible to quantify the errors associated with T_m , and so in the following discussion it is the relative differences between T_m for the samples which are considered, rather than the absolute values.

T_m for the seven samples is given in Table 2. It is apparent that, in general, there is a progressive increase in T_m with increasing petrologic grade, from 380°C in Murchison (CM2) to 601°C in Coolidge (CV4), demonstrating the more stable nature of

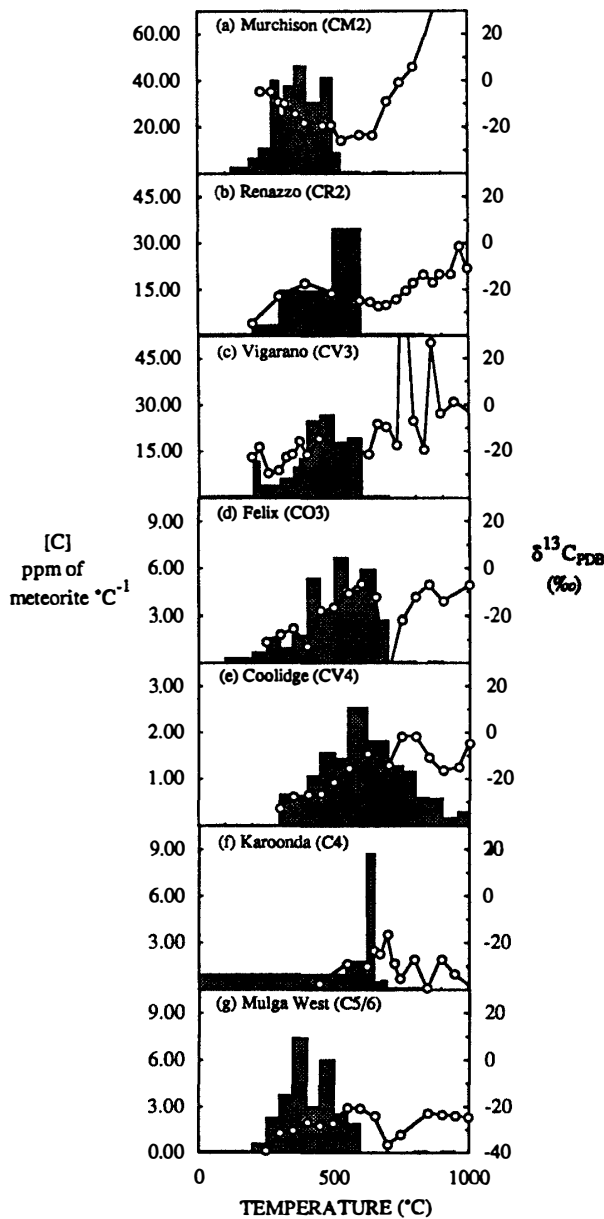


Fig. 1. Stepped combustion of seven carbonaceous chondrites below 1000°C, showing release of the major carbonaceous component(s). Yield of carbon, expressed as ppm C of meteorite °C⁻¹, shown as the shaded histogram. Note change in scale of the yield histogram between figures a-c and d-g. δ¹³C_{PDB} (‰) plotted as the open circles joined by the solid line. Errors in δ¹³C are less than the size of the symbol.

Table 2. Geometric mean combustion temperature (T_m) of carbon in C2 to C6 chondrites.

Sample	Type	Temp. °C
Murchison	CM2	380
Renazzo	CR2	504
Felix	CO3	522
Vigarano	CV3	457
Karoonda	CO4	476
Coolidge	CV4	601
Mulga West	CV5/6	397

the carbon towards oxidation in the higher compared to the lower petrologic grade meteorites. There are, however, exceptions to this trend. For instance, T_m for Renazzo is 504°C, which is considerably higher than that observed in the other C2 meteorite Murchison. The low value of T_m in Murchison may be rationalized as the presence of hydrogenated organic materials rather than amorphous carbon (which is less readily combustible, GILMOUR and PILLINGER, 1985). Noting that T_m for Renazzo is higher than that for Vigarano and Karoonda suggests that the carbon in Renazzo is more stable towards oxidation than that in some C3 meteorites. However, on the basis of δD data it is widely believed that the carbonaceous material in Renazzo is organic (YANG and EPSTEIN, 1983). The results here suggest that carbon as "organic" material in Renazzo forms only a relatively minor constituent and that the majority of the carbon exists as an amorphous or crystalline component.

In the case of Mulga West (CV5/6) T_m is 397°C (Table 2), which is lower than any C3/C4 meteorite or, indeed, Renazzo. Notwithstanding its low combustion temperature, carbon in Mulga West is similar in $\delta^{13}C$ to that in C3 and C4 chondrites. It might be anticipated that Mulga West, the highest grade chondrite analyzed in this study, should have the lowest carbon abundance, however, it contains more carbon than Felix (CO3) and Coolidge (CV4). The combustion temperature of carbon in both Felix and Coolidge argues against biological contamination; the nature of carbon in Mulga West, by virtue of its low combustion temperature, is more questionable. It is unlikely that Mulga West contains substantial indigenous organic material; rather, the low temperature carbon in this one meteorite is probably terrestrial organic contamination (Mulga West is a meteorite "find"). Thus, as was observed in the case of acid-resistant residues prepared from enstatite chondrites (GRADY *et al.*, 1988b) carbon in meteorites of higher petrologic type is more stable to oxidation than lower grade samples. In carbonaceous chondrites, this may be a reflection of graphitization during either pre-accretional nebular heating or post-accretional metamorphism.

Variations in $\delta^{13}C$ of the major carbon release suggests that in all of the samples studied there is more than one component present. The monotonic decrease in $\delta^{13}C$ in Murchison (from -4.8‰ at 190–230°C to -26.0‰ at 500–530°C) has been described previously and, indeed, has been observed in other CM2 meteorites (GILMOUR *et al.*, 1986). Results from Murchison are commensurate with there being two components: an isotopically heavy low temperature component ($\delta^{13}C > -4\text{‰}$) and a lighter component ($\delta^{13}C \approx -26\text{‰}$) combustible between 350°C and 550°C. The sample of Murchison analyzed herein had been solvent extracted prior to dissolution in HCl, so the isotopically heavy carbon is not simply solvent-soluble organic compounds, which from the work of CHANG *et al.* (1978), BECKER and EPSTEIN (1982) and EPSTEIN *et al.* (1987) are known to be enriched in ^{13}C . Thus, it is considered that the isotopically heavy, low temperature carbon component is related to the insoluble macromolecular organic material and may in fact be isotopically heavy aliphatic side chains or other functional groups. The isotopically light component in Murchison is a mixture of carbon from macromolecular material (thought to have a $\delta^{13}C$ value of $\sim -17\text{‰}$) and other elemental forms such as diamond (which in Murchison has $\delta^{13}C < -38\text{‰}$; SWART *et al.*, 1983a), or C_θ (carbon associated with spinel with $\delta^{13}C < -50\text{‰}$; TANG *et al.*, 1988).

The isotopic composition of carbon in Renazzo shows a similar trend to that in

Murchison although much less pronounced. From 300°C to 670°C, $\delta^{13}\text{C}$ drops from -17.2‰ to -27.4‰ , indicative of the presence of two components; the former is probably insoluble organic material, whilst the latter may again be related to diamond or C_θ , although thus far these components have not been identified in Renazzo.

Low temperature carbon in the five remaining chondrites (Felix, Vigarano, Coolidge, Karoonda and Mulga West) shows the opposite trend to Murchison and Renazzo: $\delta^{13}\text{C}$ generally rises with temperature. At least two components are combusting: at the lowest temperatures ($<300^\circ\text{C}$), there is a component with $\delta^{13}\text{C}$ ca. -30 to -40‰ (also seen Renazzo), most probably due to a handling blank or pump oils. Assuming only a two-component mixing in these samples, the most extreme $\delta^{13}\text{C}$ value gives the best estimate for $\delta^{13}\text{C}$ of the second component: -6.6‰ (Felix), -8.2‰ (Vigarano), -1.4‰ (Coolidge), -16.4‰ (Karoonda) and -20.3‰ (Mulga West). The $\delta^{13}\text{C}$ values for Karoonda and Mulga West would be consistent with amorphous carbon (similar to that detected in Allende and referred to as C_7 , SWART *et al.*, 1983a) but the values for Felix, Vigarano and Coolidge are considerably higher. Thus for these three samples, there might be an additional low temperature component which is relatively ^{13}C -rich (see below).

3.2. Minor carbonaceous components

The yields and $\delta^{13}\text{C}$ of carbon combusting above 600°C in nine carbonaceous chondrites are shown in Fig. 2 (Mulga West has been omitted, see below). It has been demonstrated, through analyses of highly processed chemical/physical separates (*e.g.*, CARR *et al.*, 1983; SWART *et al.*, 1983a), that there are at least three separate isotopically heavy carbonaceous components in C2 chondrites (C_α , C_β and C_ϵ) which burn at temperatures greater than 600°C. The data given here, derived from HCl-resistant residues which have not been fully demineralized or oxidized, can only be used as an indicator of the presence of isotopically anomalous components. In the C1 and C2 chondrites, $\delta^{13}\text{C}$ gradually rises with increasing temperature, *i.e.* Orgueil, Murchison and Renazzo demonstrably contain ^{13}C -rich components (with $\delta^{13}\text{C}$ up to $+500\text{‰}$). The difference in maximum $\delta^{13}\text{C}$ of the three samples may reflect either (a) the same components occurring in varying quantities (Orgueil containing the most), or (b) different components with distinct isotopic compositions. Detailed examination of the isotope profiles of Orgueil, Murchison and Renazzo shows there to be subtle effects, such as changes in slope, *etc.* which testify to the presence of multiple components; these remain unresolved at the scale of the extractions performed herein.

On a general level, the most notable feature of Fig. 2 is that, excepting the three meteorites just described, none of the other chondrites manifests increasing $\delta^{13}\text{C}$ with temperature, demonstrating a hiatus between C1–2 meteorites and samples of higher petrologic type (regardless of chemical group). This is not to say that carbonaceous chondrites of higher petrologic grade do not contain isotopically anomalous components. A recent study of Allende (CV3) by ASH *et al.* (1989) has shown there to be two ^{13}C -rich components, one of which has many similarities to C_β (SiC), and which is probably responsible for the characteristic isotopic profiles of C1 and C2 chondrites. However, C_β is only revealed in Allende after extensive chemical processing (over and above simple HCl-treatment) and prolonged gas-phase oxidation at low

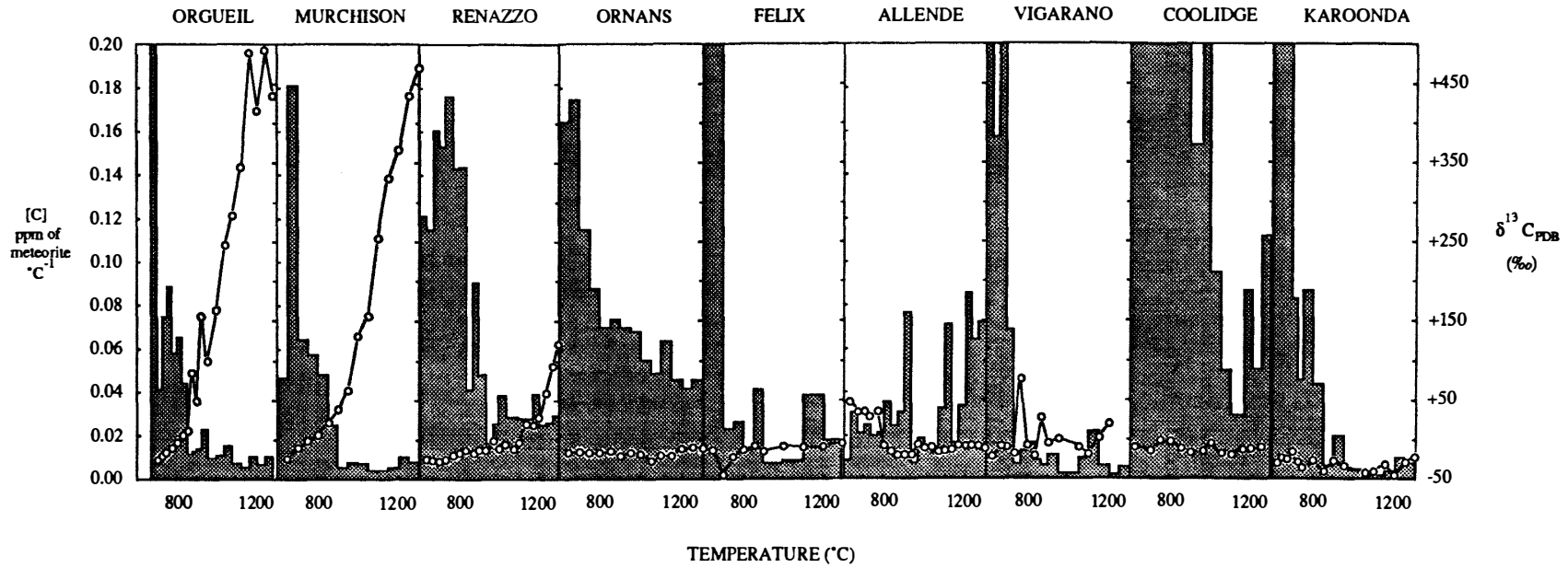


Fig. 2. Stepped combustion of nine carbonaceous chondrites above 600°C, highlighting the isotopic composition of minor carbonaceous components. Yield of carbon, expressed as ppm C of meteorite °C⁻¹, shown as the shaded histogram. δ¹³C_{PDB} (‰) plotted as the open circles joined by the solid line. Errors in δ¹³C are less than the size of the symbol.

temperatures. This in itself demonstrates that even if the ^{13}C -rich component in Allende is C_β , then it probably resides in a different location from that in C1 and C2 meteorites. From the evidence shown here, it is apparent that if C_β , or equivalent materials, are to be sought in high petrologic type carbonaceous chondrites, then extensive preparative treatments of the type administered to Allende (ASH *et al.*, 1989) are necessary.

The two CV3 meteorites, Allende and Vigarano, both contain a ^{13}C -rich component unrelated to the material which gives the gradual rise in $\delta^{13}\text{C}$ with temperature observed for C1 and C2 samples (Fig. 2). There is a release of ^{13}C -rich material between 600–800°C in Allende and 700–900°C in Vigarano. That of Allende is commensurate with a single component, but in Vigarano the isotope profile appears to be complex, suggesting the presence of more than one component (at the very least, one isotopically heavy and one light). ASH *et al.* (1989) demonstrated that an HF-, HCl- and HNO_3 -resistant residue from Allende, when combusted several times at 500°C to remove C_γ (the bulk of the isotopically normal carbon, SWART *et al.*, 1983a), reveals ^{13}C -rich material with $\delta^{13}\text{C}$ *ca.* +340‰ which burns between 600 and 800°C. This noble gas-poor component has been called C_\aleph (aleph) and is thought to have affinities with C_α (ASH *et al.*, 1989), or amorphous carbon, the putative carrier of Ne-E(L) in C1 and C2 chondrites. Since the most extreme $\delta^{13}\text{C}$ values measured herein are +47.3‰ (Allende) and +77.3‰ (Vigarano), these values must be compromised by the combustion of C_γ and C_δ (two ^{13}C -depleted components), which are not removed by the HCl-treatment. None of the other samples of petrologic type 3 or greater show evidence for ^{13}C -enriched material, although in Ornans, Coolidge and Karoonda relatively large amounts of isotopically normal carbon are liberated below 800°C, thereby possibly masking any signal. Felix, on the other hand, has isotopically light carbon ($\delta^{13}\text{C} < -45\text{‰}$) burning between 600 and 800°C (seen more clearly in Fig. 1), thus, detection of an isotopically heavy component in Felix might also be compromised. ASH *et al.* (1988) uncovered, in an extensively processed residue from Allende, a small amount of isotopically light carbon ($\delta^{13}\text{C} < -260\text{‰}$), burning between 750 and 800°C. It was given the non-committal designation C_λ . It is possible that Felix also contains C_λ , detectable in an HCl-resistant residue.

Data for the remaining chondrite, Mulga West, were compromised by an analytical problem, hence are not given in Fig. 2. However, over 80 ng of carbon combusted at temperatures above 800°C, an amount well in excess of blank levels. The high temperature of combustion argues against both contamination from organic materials and an indigenous elemental form of carbon, unless this latter is “protected” in some way (entrapped in residual silicate minerals, for instance).

4. Discussion

There is a gradual transition in the nature of the major carbonaceous components from C1 to C4 chondrites, reflected by an increasing resistance towards oxidation, manifested by progressively higher values of T_m . GILMOUR and PILLINGER (1985) have demonstrated that the combustion temperature of terrestrial organic materials increases with decreasing hydrogen content. Thus, on the basis of the data presented

herein, it is apparent that the carbon in the higher petrologic type meteorites is less hydrogenated. C1 and C2 meteorites contain organic materials (*sensu stricto*), whereas C3 and C4 chondrites possess carbon with a more graphitic nature. With the change in form of the carbon is a concomitant decrease in carbon abundance. The significance of these observations is hampered somewhat by the meteorites of different petrologic type belonging to various chemical groups. Furthermore, as noted in the previous section, carbon is distributed heterogeneously throughout individual meteorites, and without detailed studies of each sample it is impossible to assess the extent of this effect. Thus, while it is evident that the C1 and C2 meteorites contain the most carbon, the carbon contents of the C3 to C6 chondrites cannot be said to display a coherent trend.

Notwithstanding the provisos outlined above, the information given in Fig. 1 (excluding the data for Mulga West, due to contamination) has some importance. It is necessary to try and explain why higher petrologic type samples have carbon of a more graphitic nature (there is no direct evidence from this study that the carbon has a graphite structure; the term graphite is taken to include amorphous carbon). In this context it is appropriate to consider the abundance patterns of volatile elements in chondritic meteorites, which are generally depleted as petrologic grade increase. Still under debate is the question of whether volatile materials (*e.g.* organics in C1 and C2 meteorites) were originally incorporated in all meteorites and subsequently lost during the metamorphic processes which affected higher petrologic grade samples, or whether events taking place in the nebula controlled the material that accreted into meteorite parent-bodies. Accounts of these opposing views may be obtained from DODD (1969) and LARIMER and ANDERS (1967). If metamorphic effects control the nature of carbon in carbonaceous chondrites, then high petrologic grade chondrites must originally have contained large amounts (up to several wt%) of organic materials. Devolatilization during parent-body heating would lead to loss of carbon in the form of gases such as CH₄, CO and CO₂, leaving a residue of predominantly aromatic (*i.e.* graphitic) organics.

Alternatively, graphitic carbon might have been incorporated directly into the parent-bodies of C3 to C6 chondrites. In a gas of solar composition (C/O ≈ 0.6) carbon does not condense as graphite; rather CO, the dominant form at high temperatures, is transformed into organic materials by grain catalysis reactions (*e.g.*, HAYATSU and ANDERS, 1981). Thus, within the framework of this simple model, in order to incorporate graphitic carbon directly into a meteorite parent-body, organic material must be graphitized by a nebula heating process, a contorted pathway which seems a little *ad hoc*. Although graphite can condense directly in environments with C/O ≈ 0.9 (LARIMER, 1975), such as the environment in which enstatite chondrites formed, there is no correspondence between $\delta^{13}\text{C}$ of graphite in enstatite chondrites ($\delta^{13}\text{C} = -9$ to $+4\%$, GRADY *et al.*, 1986) and the graphitic component in C3 and C4 chondrites ($\delta^{13}\text{C} \leq -17\%$). In light of the similarity in $\delta^{13}\text{C}$ between the graphitic component in C3 and C4 samples and organic materials in C1 and C2 meteorites, it seems reasonable to suppose some sort of genetic relationship. However, from the results presented herein, it is not possible to distinguish between a graphitization process occurring in the nebula or during planetary metamorphism.

Figure 2 shows that at the highest temperatures of the stepped heating experiments

$\delta^{13}\text{C}$ decreases as $\text{C1} > \text{C2}(\text{CM} > \text{CR}) > \text{C3} > \text{C4}$. If an enrichment in ^{13}C is taken to signify the presence of interstellar grains, then clearly C1 and C2 meteorites contain them. The evidence presented herein from CV3 meteorites is less convincing although it is considered that there are sufficient data to conclude that interstellar grains are present in these meteorites. Indeed, ASH *et al.* (1989) have demonstrated conclusively that ^{13}C -rich materials are present in Allende and can be readily concentrated provided substantial preparative chemistry is employed beforehand. On the other hand, there is little indication of ^{13}C -rich grains in either CO3 or C4–6 samples. The question arises as to whether all carbonaceous chondrites originally contained interstellar grains (subsequently destroyed by planetary processes) or whether they were only present in low petrologic types. The most ^{13}C -enriched phases in C1 and C2 chondrites are SiC (ZINNER *et al.*, 1987); the fate of SiC during metamorphism is unknown, but its refractory nature suggests that it should survive (at very least in C3 samples, which have been subjected only to mild re-heating). It is therefore reasonable to assume that isotopically anomalous carbonaceous components were never accreted into the source material of CO3 or C4–6 meteorites. The ^{13}C -rich components in CV3 meteorites are probably different from those in C1 and C2 meteorites: in addition to apparent differences in concentration, which may make detection difficult in C3 samples, the isotopic enrichments are detected at different temperatures.

Of the meteorites which have not been subjected to high degrees of metamorphism, the sample which has suffered the highest degree of aqueous alteration (Orgueil), also exhibits the largest ^{13}C -enrichment. Conversely, Allende, which shows only scant evidence for aqueous effects, contains the smallest ^{13}C -enrichment (even in more processed HF/HCl-resistant residues, $\delta^{13}\text{C}$ never exceeds $+550\%$, ASH *et al.*, 1989). Hydrothermal activity on the parent-body might therefore have influenced distribution of isotopically anomalous materials (their fine-grained nature allowing easy transport). Thus, as ^{13}C -rich dust was accreted onto primitive parent-bodies at a late-stage in their formation, aqueous fluid movements may have acted to concentrate such materials in C1 meteorites, or alternatively carry minor quantities into CV3 samples. The presence of carbonates with (relatively minor) ^{13}C -enrichments in C1 and C2 samples might also be related to aqueous processing of ^{13}C -rich interstellar grains. However, evidence to support this idea has been sought, but not found (GRADY *et al.*, 1988a).

5. Summary

The transition from C1 to C4 carbonaceous chondrites is accompanied by a decrease in carbon content and a change in the nature of the major carbonaceous component. Carbon in C1 and C2 chondrites occurs predominantly as organic materials. However, the CR2 meteorite Renazzo appears to contain amorphous elemental carbon rather than hydrogenated organic compounds. This is at odds with D/H studies which have uncovered a deuterium enrichment in Renazzo which is thought to be associated with organic materials (YANG and EPSTEIN, 1983). Further study of Renazzo is necessary in order to characterize fully the major carbonaceous phases in this chondrite, which might lead to the placing of more firm constraints on their origins.

C3 and C4 meteorites contain carbon in the form of an amorphous or graphitic component. This is isotopically similar to the organic materials in C1 and C2 samples, suggesting a genetic relationship. During condensation in the solar nebula, carbon is transformed from gas-phase species into organic materials, hence the amorphous/graphitic component in C3 and C4 samples may arise from dehydrogenation of pre-existing organic compounds. It is not possible to assess conclusively whether this process took place prior to accretion in the solar nebula, or following meteorite formation during planetary heating. All C3 and C4 samples analyzed herein have been metamorphosed to some degree. Whilst this process can undoubtedly convert organic materials into amorphous/graphitic carbon, it is not clear whether the extent of metamorphism would be sufficiently severe to ensure complete transformation (the CV3 meteorites for instance have suffered only mild metamorphic effects). Laboratory simulation studies should help constrain the effects of this process on organic materials.

C1 and C2 meteorites contain interstellar grains, ^{13}C -enriched materials such as SiC. Orgueil (C1) exhibits the largest ^{13}C -enrichment, while Renazzo (CR2) shows the lowest. It is not clear whether this reflects a difference in the isotopic compositions of grains from carbonaceous chondrites of various chemical groups, or petrologic types, or a difference in relative abundances (the largest ^{13}C -enrichment symptomatic of the highest concentration). CV3 meteorites show only scant evidence for the presence of ^{13}C -rich grains in HCl-resistant residues, although it is known that they are present in these meteorites (ASH *et al.*, 1989). On the basis of the limited evidence collected herein, CV3 chondrites appear to possess a different complement of ^{13}C -rich grains from CI/CM/CR samples. CO3 and C4–6 meteorites, on the other hand, apparently do not contain ^{13}C -enriched carbonaceous components.

ASH *et al.* (1989) proposed that Allende contained, at most, 10–20 ppb of SiC, compared to ~ 2000 ppb in CM2 chondrites (TANG *et al.*, 1988). SiC should survive the mild metamorphism experienced by C3 meteorites, hence its low abundance in CV3 samples and absence in CO3 meteorites suggests that only minute amounts of SiC were ever present in these samples. C4–6 meteorites presumably never contained any SiC. Abundant ^{13}C -enriched grains occur only in meteorites which have suffered aqueous alteration, which may either be coincidence (*i.e.* large concentrations in regolith samples), or the distribution may be related to aqueous processing (*i.e.* hydrothermal activity may have acted to concentrate, or re-distribute the grains).

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