# THE STABLE ISOTOPIC COMPOSITION OF CARBON, NITROGEN AND HYDROGEN IN SOME YAMATO METEORITES

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*Abstract:* Three carbonaceous chondrites from the Yamato Mountains (Y-790112, Y-790032, Y-790003) were studied by stepwise combustion in order to distinguish different components of carbon, nitrogen and hydrogen on the basis of release temperature and isotopic composition.

## 1. Introduction

Isotopic analyses of carbon, nitrogen and hydrogen have been carried out on three carbonaceous chondrites from the Yamato Mountain region of Antarctica. The meteorites studied are Y-790003 (CM), Y-790032 (CM) and Y-790112 (CR) (NATIONAL INSTITUTE OF POLAR RESEARCH, 1982). Two ureilite samples (Y-74123 and Y-790981) were also analyzed, but for carbon only. The paper presents the results of this light element stable isotope study, the first to be reported for meteorite samples from Japanese Antarctica.

The purpose of the study was to compare data obtained from Antarctic meteorites with that from meteorite "falls". The method of stepwise combustion distinguishes different phases of carbon, nitrogen and hydrogen on the basis of release temperature and isotopic composition. This method has been used previously to reveal the presence of low abundance components with highly anomalous isotopic compositions in carbonaceous and ordinary chondrites: $\delta^{13}C$ up to $+1100\permil$ (SWART et al., 1983b), $\delta^{15}N$ down to $-270\permil$ (WRIGHT et al., 1983b) and $\delta D$ up to $+5700\permil$ (MCNAUGHTON et al., 1982). Stepwise combustion also allows characterization of graphite and diamond in ureilites, and two Yamato samples (Y-74123 and Y-790981) were examined for carbon, as part of a wider investigation of ureilites. In the case of carbon, the method distinguishes weathering products (formed during the lifetime of samples on the Antarctic ice) and other terrestrial contaminants from the indigenous components of interest (SWART et al., 1983a). Fresh Antarctic specimens, which have been less exposed to contamination from terrestrial organic species than non-Antarctic meteorites, are ideal specimens for further characterization of light element stable isotope compositions, hence samples from the Yamato collection were selected for this study.

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## 2. Experimental

The technique used to characterize carbon and nitrogen in the samples studied was one of combustion at progressively increasing temperatures in an atmosphere of oxygen (stepped combustion). Extraction of carbon and nitrogen is performed on separate aliquots of material; carbon is analyzed as carbon dioxide on a dynamic mass spectrometer (VG Micromass 602E) and nitrogen as $N_2$ in a static mass spectrometer. Full experimental details of the extraction procedures are published elsewhere (SWART et al., 1983a; WRIGHT et al., 1983a).

In the case of hydrogen, insufficient amounts of sample were available for separate determination, hence the water produced during extraction of carbon was collected in two steps (200–600°C, 700–1200°C) and analyzed as hydrogen gas, after conversion over uranium (McNAUGHTON et al., 1982). Blank contributions from this method are negligible.

Isotopic compositions are quoted as $\delta$-values (deviations in parts per thousand (‰) from a standard) in the usual way, relative to internationally accepted standards: PDB for carbon, AIR for nitrogen and SMOW for hydrogen gas.

The minimum amout of carbon which may be measured on the dynamic instrument is $2.5\,\mu g$ of carbon as $CO_2$, to a precision of $\pm 0.05$‰. For some of the temperature intervals, smaller quantities of $CO_2$ were produced, therefore a technique of isotope dilution was employed, in order to enable determination of the isotopic composition. This method consists of mixing a small amount of standard gas with the $CO_2$ produced on combustion of the sample. After necessary corrections, the isotopic composition of the sample may be determined to a precision of better than $\pm 1$‰ (LEWIS et al., 1983).

The minimum quantity of nitrogen gas required for analysis is about 2 ng, the precision of a $\delta^{15}N$ measurement being $\pm 2$‰. For hydrogen, a precision of $\pm 5$‰ is attainable on a minimum sample of $1\,\mu g$. It was not possible to determine precise yields of the water collected, although in each case amounts greater than $1\,\mu g$ were produced.

Fresh interior surfaces were selected for the analyses, in order to reduce the possibility of the addition of extraneous components formed by weathering in the severe Antarctic climate (GIBSON and YANAI, 1979). However, if such components are present, it is possible to distinguish them from indigenous phases on the basis of their very low thermal release temperatures (SWART et al., 1983a).

In order to obtain the maximum amount of information possible from the Antarctic carbonaceous chondrites, 15–20 mg of each was ground in a stainless steel pestle and mortar to a grain size of around 50 $\mu$m. This was then dissolved in orthophosphoric acid, in order to liberate carbon dioxide from carbonate minerals (McCREA, 1950). Acid digestion was carried out for 24 hr, a period of time which would allow dissolution of calcite, but not magnesian carbonates (WALTERS et al., 1972). The residue after acid dissolution was reclaimed, washed in distilled water until the supernatent liquid was neutral, then dried. This acid-resistant residue was used for the stepped combustion analyses of carbon, nitrogen and hydrogen in the carbonaceous chondrites.

## 3.  Results and Discussion

### 3.1.  Carbonaceous chondrites

Table 1 lists the results of stable isotopic analysis of the three Antarctic carbonaceous chondrites studied.   Included in the table, for comparative purposes, are data for Murchison and Renazzo, typical examples of the two carbonaceous chondrite groups to which the Yamato samples are thought to belong.   The data for each meteorite will be discussed individually.

*Table 1.   Carbon, nitrogen and hydrogen data from Murchison, Renazzo and the three Yamato chondrites studied.*

| Sample | Type | Total C* wt % | $\delta^{13}C_{PDB}$* ‰ | Carbonate ppm | $\delta^{13}C_{PDB}$ carbonate | Total N* wt % | $\delta^{15}N_{AIR}$* ‰ | $\delta D$* ‰ |
|---|---|---|---|---|---|---|---|---|
| Y-790112 | CR | 1.2 | −18.5 | 8 | +28.0 | 0.03 | +94.4 | +89.5 (200− 600°C) |
| Renazzo | CR | 1.7 | −9.9 | 500 | +65.1 | 0.05 | +175.3 | +528.4 (200−1200°C) |
| Y-790032 | CM | 4.4 | −12.1 | 600 | +30.4 | 0.08 | +21.1 | +24.8 (200− 600°C) |
| Y-790003 | CM | 2.0 | −27.6 | 300 | +67.8 | 1.20 | +214.1 | +21.4 (200− 600°C) +18.1 (600−1200°C) |
| Murchison | CM | 4.6 | −15.8 | 400 | +38.0 | 2.60** | +23.9** | +22.7 (600−1200°C) |

 *  C, N and H data are from orthophosphoric acid residues, with the exception of those for Renazzo, which are for an untreated sample.
 ** Data from pyrolysis, not combustion.

### 3.1.1.  Yamato-790112 (CR)

Orthophosphoric acid dissolution of 19.02 mg of Y-790112, a CR2 carbonaceous chondrite, yielded carbon dioxide from carbonate minerals in a concentration of only 8 ppm, with an isotopic composition of +28‰.  A similar treatment of Renazzo showed that carbonate minerals were present in far greater quantities, around 500 ppm, and that the carbonates were more enriched in $^{13}C$, at +65‰.   Al Rais (another CR chondrite, data not in table), also yielded isotopically heavier carbon (+54‰) than Y-790112 upon acid treatment, but in this case the carbonates were in similar concentrations to Y-790112, at 23 ppm.   There appears to be considerable variability in the isotopic composition and the concentration of carbonate minerals in CR2 meteorites. A possible explanation would be a contribution of isotopically light $CO_2$ from a source other than carbonates, as yet unidentified.   Indeed, Semarkona, Bishunpur and Chainpur, specimens of the most unequilibrated of type 3 ordinary chondrites, and all "falls", show some evidence for the presence of an isotopically light, non-carbonate phase which liberates carbon dioxide on $H_3PO_4$ treatment (GRADY et al., 1982a).   The acid-resistant residue was depleted in $^{13}C$, the heavier stable isotope of carbon, with a bulk isotopic composition of −18.5‰.   A simple mass balance calculation, assuming no carbon other than $CO_2$ was liberated by $H_3PO_4$, gives exactly the same $\delta^{13}C$ value.   The

$-18.5‰$ measured for Y-790112 is hence very different from the $-9.1‰$ calculated for Renazzo. Unfortunately, no orthophosphoric acid residue of Renazzo was available for combustion, and so at this time a measured isotopic composition of the non-carbonate fraction of Renazzo remains unknown.

Figure 1a shows the profile of carbon liberated during stepped combustion of 2.85 mg of Y-790112 orthophosphoric acid residue, between 200–1200°C. Below 500°C, some 91% of the carbon is released, with an average isotopic composition of *ca.* $-18$ ‰, probably representing organic macromolecular material. Between 500–600°C, a further 6.5% carbon is given off, with a much heavier isotopic composition, around
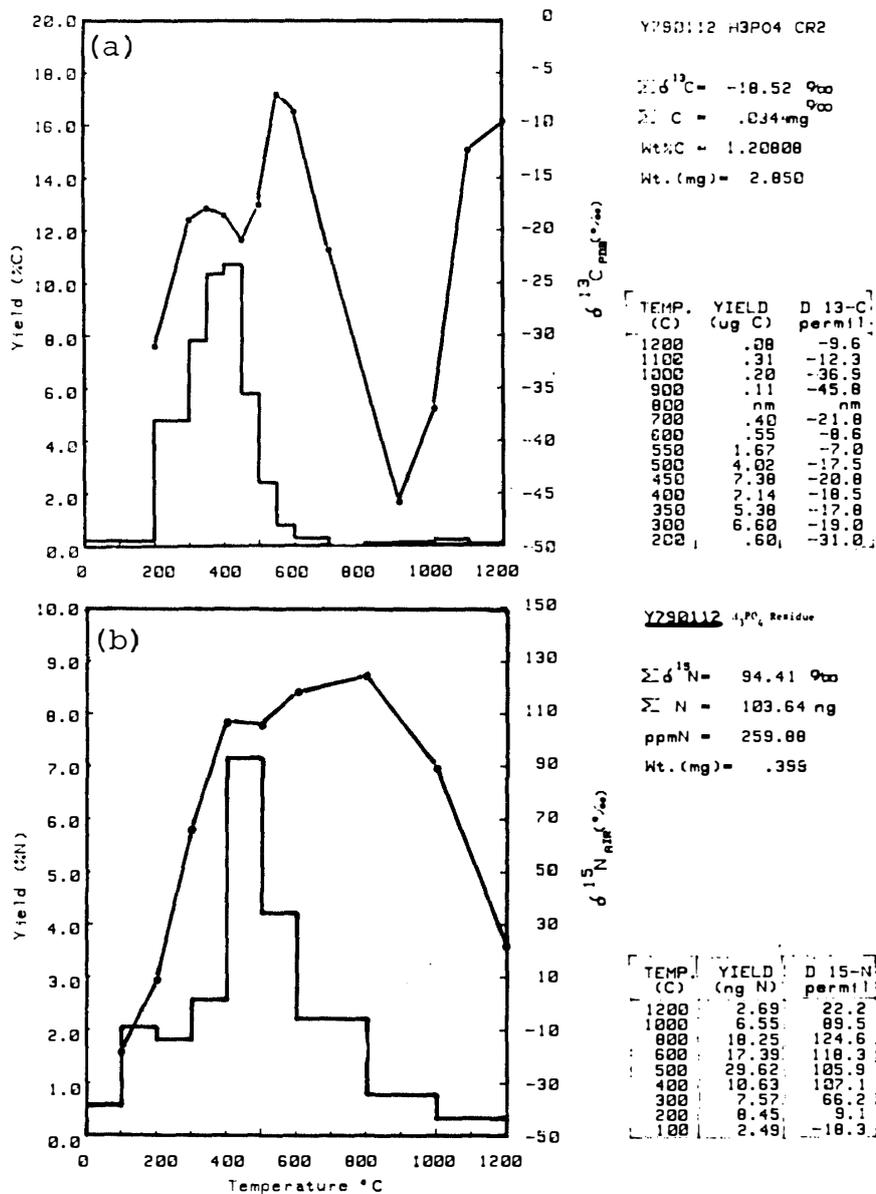


Fig. 1. Stepped combustion of Y-790112 (orthophosphoric acid residue).

(a)  C release (———) and $\delta^{13}C$ values (—·—).

(b)  N release (———) and $\delta^{15}N$ values (—·—).

$-8‰$. This temperature is appropriate for the thermal degradation of carbonate minerals, and it is conceivable that some isotopically heavy carbonate species have survived the $H_3PO_4$ treatment. Such carbonates cannot be calcite, but might possibly be magnesium-bearing species (magnesite, dolomite etc.). Variable proportions of different carbonate minerals in CR meteorites would help to explain the range of $\delta^{13}C$ values and yields of $CO_2$ in the $H_3PO_4$ expriment. Above 600°C, the remainder of the carbon is liberated in very small quantities by combustion from Y-790112; at first this seems to be isotopically light, but the $\delta^{13}C$ value rises rapidly at higher temperatures, in keeping with the presence of a small amount of the very heavy carbon first recognized in Murchison (SWART et al., 1983b).

Carbon in Renazzo-type carbonaceous chondrites does not possess a particularly diagnostic signature, whereas nitrogen is of sufficiently different isotopic composition from that found in CI, CM, CV and CO chondrites to be regarded as characteristic of CR's (KUNG and CLAYTON, 1978). A further 0.399 mg sample of the Y-790112 orthophosphoric acid-resistant residue was combusted in a stepwise manner to determine its nitrogen isotopic composition. The bulk of the nitrogen was liberated over a broader temperature range than carbon (73% between 400–800°C; see Fig. 1b). The maximum release of carbon and nitrogen in both cases is between 400 and 500°C. It is unwise at present to compare the results obtained for nitrogen and carbon too directly, as there are subtle differences between the two extraction procedures. For instance the oxygen pressure used for the carbon combustion is perhaps an order of magnitude higher than that employed for nitrogen ($>500$ mb, as opposed to around 50 mb). Clearly therefore we would expect slight differences in combustion temperatures for the same phases in the different experiments. The isotopic composition of the major nitrogen phase was heavy, averaging around $+110‰$. Since this $\delta^{15}N$ is fairly constant up
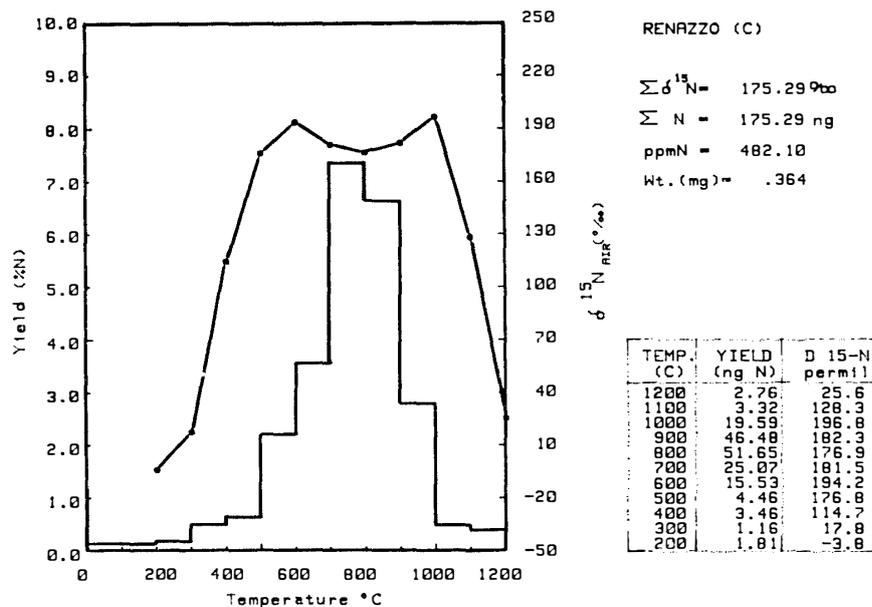


RENAZZO (C)

$\Sigma \delta^{15}N =$ 175.29 ‰
$\Sigma N =$ 175.29 ng
ppmN = 482.10
Wt.(mg) = .364

| TEMP. (C) | YIELD (ng N) | D 15-N permil |
|---|---|---|
| 1200 | 2.76 | 25.6 |
| 1100 | 3.32 | 128.3 |
| 1000 | 19.59 | 196.8 |
| 900 | 46.48 | 182.3 |
| 800 | 51.65 | 176.9 |
| 700 | 25.07 | 181.5 |
| 600 | 15.53 | 194.2 |
| 500 | 4.46 | 176.8 |
| 400 | 3.46 | 114.7 |
| 300 | 1.16 | 17.8 |
| 200 | 1.81 | -3.8 |

Fig. 2.  Stepped combustion of Renazzo (untreated sample).
N release (——) and $\delta^{15}N$ values (—·—).

to 800°C, it is probable that only one phase (the organic macromolecule) is being decomposed.

Nitrogen was also extracted by stepped combustion from a sample which had not been treated with orthophosphoric acid. The results were almost identical to the acid residue, 72% of the total nitrogen was again liberated between 400–800°C in a broader profile and $\delta^{15}N$ values were slightly heavier: $+130$ to $+140\%o$, rising to $+151\%o$ at 800°C. At present it is unknown what the $H_3PO_4$ treatment does to the indigenous nitrogen compounds, but it could be expected that water-soluble species such as amino acids would be extracted during the recovery procedure. It is interesting to note that the bulk $\delta^{15}N$ value for the acid residue is lighter ($+94\%o$) than that for the untreated sample ($+113\%o$). This is an effect which has also been observed in two other meteorites (Murchison, CM2 and Semarkona, LL3; WRIGHT *et al.*, 1983b), and seems to suggest that $H_3PO_4$ treatment might selectively liberate an isotopically heavy nitrogen component. Stepped nitrogen extraction data for untreated Y-790112 with comparable information from an analysis of Renazzo (Fig. 2) shows a general similarity—but the nitrogen released at 800°C from Renazzo is isotopically heavier, at $+177\%o$.

Water from bulk Renazzo has a variable isotopic composition, between $+944\%o$ (ROBERT and EPSTEIN, 1982) and $+528\%o$ (McNAUGHTON *et al.*, 1982); that from Y-790112 acid residue was $+89.5\%o$, the highest observed for any of the Yamato meteorites considered in this study; the $\delta D$ value of Y-790112 may be considered as being of a similar order to that of Renazzo.

Taken overall, light element stable isotope analyses of C, N and H in Y-790112 are consistent with this meteorite being an example of a Renazzo-type carbonaceous chondrite.

### 3.1.2.    Yamato-790032 (CM)

Orthophosphoric acid treatment of 13.48 mg of Y-790032 liberated 600 ppm of carbon dioxide, with an isotopic composition of $+30\%o$. The sample of Murchison analyzed at Cambridge (BM 1970, 6) contained carbonates with an isotopic composition of $+38\%o$ at a concentration of 400 ppm—although this value is somewhat lighter than those obtained by other investigators ($+45\%o$ KVENVOLDEN *et al.*, 1970; $+44\%o$ CHANG *et al.*, 1978), replicate analyses have confirmed the figure (GRADY, 1982). The isotopic variability has been ascribed to a mixture of two types of carbonate, one associated with matrix, and the other in high temperature inclusions (SWART *et al.*, 1982). Hence carbonates contained in Y-790032 appear to be similar in composition to those from Murchison, and fall within the range of carbonate mineral isotopic compositions of carbonaceous chondrites.

Figures 3a and 4 show the release profiles of carbon from 0.984 mg of Y-790032 orthophosphoric acid residue and 9.74 mg of Murchison $H_3PO_4$ residue respectively. Both meteorites gave comparable yields of carbon, with similar isotopic compositions (see Table 1). Most of the carbon is released from Y-790032 below 500°C—some 97%, again thought to be from combustion of organic polymer. In Y-790032 two phases of differing isotopic composition are resolved, the first oxidizing at lower temperatures (up to 400°C) with a composition slightly enriched in $^{13}C$ with respect to the second component at $-8\%o$, the other being combusted between 400–500°C, isotopically lighter at around $-18\%o$. Both phases are slightly heavier than the material in Y-790112.
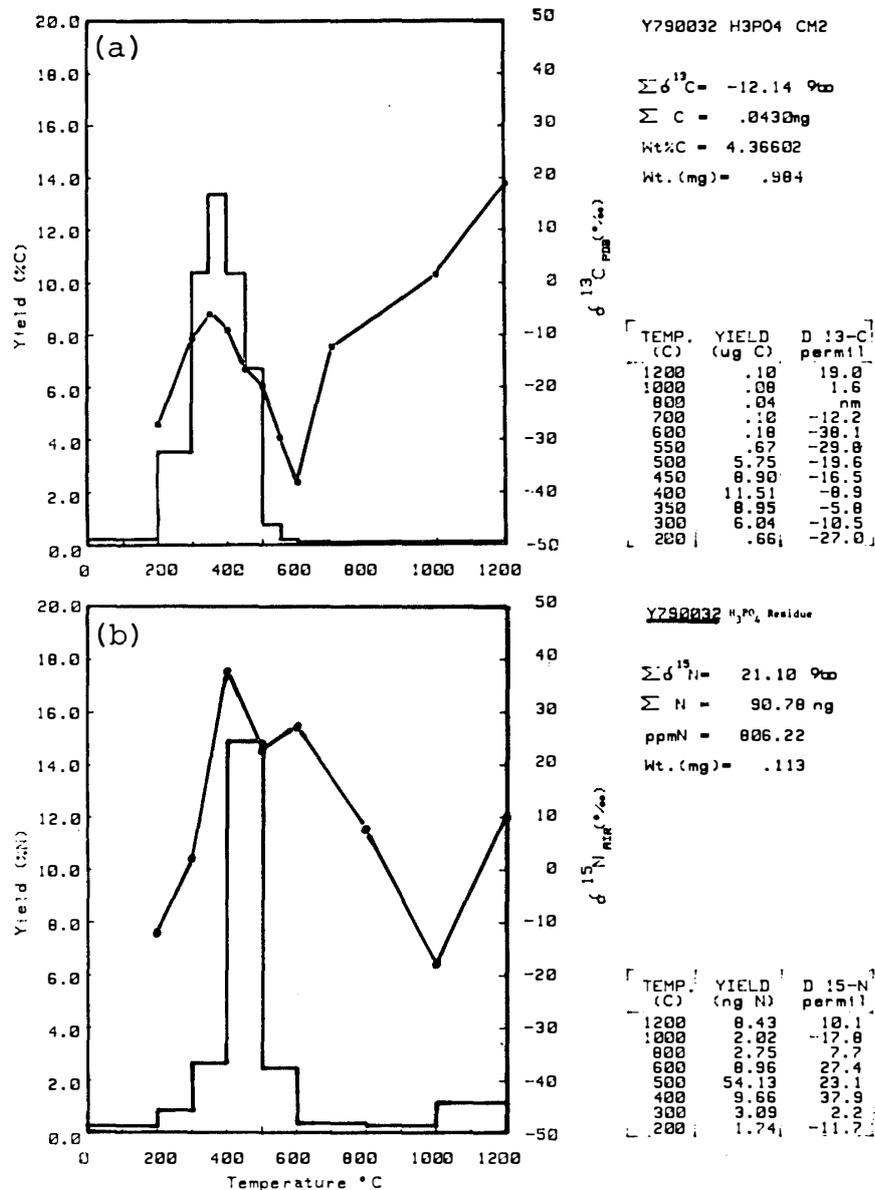
Fig. 3.   Stepped combustion of Y-790032 (orthophosphoric acid residue).

(a)   C release (——) and $\delta^{13}C$ values (—•—).

(b)   N release (——) and $\delta^{15}N$ values (—•—).

The Murchison acid residue, where two components are also apparently present gives $-13‰$ up to 400°C, $-18‰$ at 500°C. Stepped combustion of a bulk, untreated sample of Murchison afforded $\delta^{13}C$ value of $-9$ and $-14‰$ in the same temperature ranges.

Above 500°C, the remaining 3% of the carbon in Y-790032 is liberated in decreasing amounts. The isotopic composition of this carbon becomes progressively enriched in $^{13}C$, reaching $+19‰$ at 1200°C, where 0.2% of the total carbon budget is released. With the exception of the low $\delta^{13}C$ values in the 550 to 600°C temperature range, carbon in Y-790032 shows a similar trend to that exhibited in Murchison, although in the latter case, the isotopic composition climbs to $+140‰$ at 1200°C, where 0.04% of
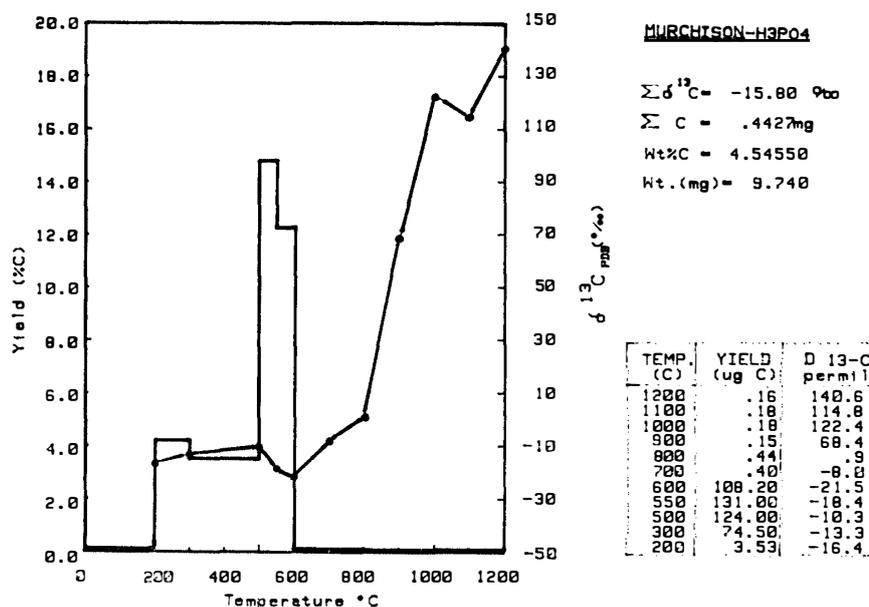
*Fig. 4. Stepped combustion of Murchison (orthophosphoric acid residue).*

C release (——) and $\delta^{13}C$ values (—·—).

the total carbon is combusted. Carbonate minerals decompose in the temperature range of 500–700°C, therefore the carbon released at 1200°C is not from such a source. Examination of a more demineralized acid residue of Murchison, 2C10f (prepared by oxidative acid attack of an HF/HCl residue) showed evidence of a very substantial enrichment in $^{13}C$ at higher temperatures, rising to $\delta^{13}C = +1100\%o$ (SWART et al., 1983 b). Isotopic values of such anomalous compositions cannot be explained by simple equilibrium fractionation mechanisms. The exotic carbon phase seems to be associated with $^{22}Ne$ and s-process Kr and Xe, hence it may represent interstellar grians ejected into the protosolar nebular from red giant stars. Y-790032, like Murchison and a number of other meteorites of the CM class (SWART et al., 1983b), has retained small amounts of the phase, probably within the fine-grained matrix.

The nitrogen data obtained from 0.113 mg of Y-790032 acid-resistant residue are shown in Fig. 3b. The total nitrogen isotopic composition of Y-790032, at +21‰, with a maximum $\delta^{15}N$ at +38‰, was found to be very similar to that of Murchison (+24 ‰), although the overall yield was a factor of three lower. The major release of nitrogen from the Antarctic sample was at a slightly higher temperature than that of the major carbon release, occurring across the range 300–600°C, and peaking at 500°C. This probably only reflects a difference in the extraction techniques, and the bulk nitrogen component most likely derives from the organic macromolecule.

The hydrogen isotopic composition, at +25‰, was close to the Murchison data (+23‰). Hence evidence from carbon, nitrogen and hydrogen all confirm that Y-790032 is typical CM chondrite.

### 3.1.3. Yamato-790003

The third Antarctic carbonaceous chondrite examined in this study, and previously classed with Y-790032 as a CM-type, provided some unexpected results. The yield of $CO_2$ produced during orthophosphoric acid digestion of carbonate minerals was
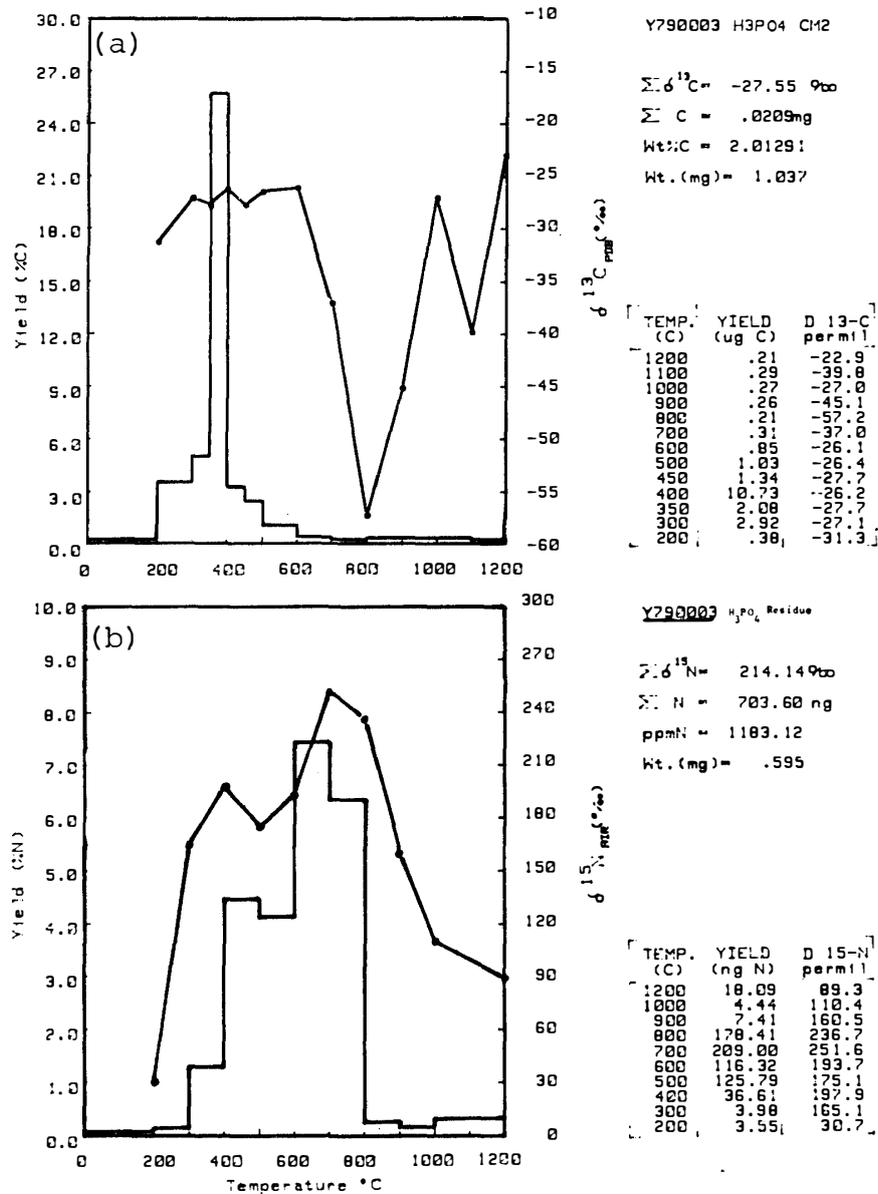
Fig. 5. *Stepped combustion of Y-790003 (orthophosphoric acid residue).*

(a)   C relaese (——) and $\delta^{13}C$ values (—·—).

(b)   N release (——) and $\delta^{15}N$ values (—·—).

300 ppm, comparable to that from Murchison, however the isotopic composition of these carbonates was much heavier: $+68\%_0$, and is in fact one of the most $^{13}$C-enriched carbonate measured in a CM chondrite. Nonetheless, the value is inside the range of compositions recorded for CI and CR chondrites, and is indeed close to the isotopic composition of carbonate from Renazzo ($+65\%_0$).

Figure 5a is the profile of carbon liberated during combustion of 1.037 mg of Y-790003 acid residue. Summing the isotopic compositions across the entire temperature range gives a value for the total isotopic composition of the acid residue of $-28\%_0$. This value is much lighter than that for other Yamato carbonaceous chondrites, and

indeed for any other CM chondrite, except Nogoya (BUNCH and CHANG, 1980).

Detailed examination of Fig. 5a shows that most of the carbon is liberated by 500° C (some 88%), with a constant isotopic composition of $-27‰$. In contrast to Murchison and Y-790032, there is no appreciable difference in $\delta$-value of the carbon released at low (up to 400°C) and intermediate temperatures (400–500°C). Above 700° C, 6% of the carbon is released in almost constant amounts up to 1200°C. However, the isotopic composition of this carbon is very variable—at 800°C, it drops to $-57‰$, then rises again, to $-23‰$ at 1200°C. The isotopic pattern is therefore more similar to that of Y-790112 (CR) than Y-790032 (CM).

The nitrogen isotopic data from Y-790003 are fascinating, as this sample, supposedly a CM2, was found to contain extremely heavy nitrogen (Fig. 5b). In fact the bulk sample yielded a total $\delta^{15}N$ value of $+187‰$ (with a phase released at 700°C as heavy as $+219‰$). Values of this nature are more consistent with CR-type meteorites, and so question the validity of Y-790003's classification as a CM2.

For Y-790112, it was noted that $H_3PO_4$ treatment tended to decrease $\delta^{15}N$ values, suggesting a phase enriched in $^{15}N$ was acid or water soluble. However, when undemineralized Y-790003 was analyzed, the nitrogen appeared to be more enriched in $^{15}N$. Thus acid or water soluble nitrogen must have a $\delta^{15}N$ between $+100‰$ and $+200‰$. The stepped combustion profile of the orthophosphoric acid residue (Fig. 5b) is conspicuous in its apparent bimodal release of nitrogen; this was presumably obscured in the bulk sample.

Water from the sample, however, was not enriched in deuterium, as were the CR chondrites, but with $\delta$-D values of $+18‰$ up to 600°C and $+21‰$ from 600–1200°C, fell into the range of isotopic compositions of CM samples. Notwithstanding this, the balance of the evidence from light element stable isotope studies of Y-790003 indicates that this meteorite is unlike other CM chondrites, but rather should be considered as a CR, or an anomalous carbonaceous chondrite.

### 3.2.  Ureilites

Petrographic examination of ureilites has revealed the presence of diamonds, closely intergrown with graphite (VDOVYKIN, 1970), both carbon species being concentrated in veins between the major silicate mineral assemblages (BERKELEY and JONES, 1982). The origin of the carbon in ureilites, and the formation of diamonds, has been discussed by several authors (e.g. WASSON et al., 1976; BERKELEY et al., 1976, 1980). Under debate has been the question of whether carbon was present throughout the lifetime of the ureilite parent body (bodies), or whether some of it was introduced as a late-stage injection product during a shock event. Current evidence from petrographic and textural features is ambiguous.

The purpose of carbon analysis of ureilite specimens was to determine the isotopic composition of the graphite and diamond in the samples, to see if any evidence was present which might indicate carbon from two or more provenances, and to verify that the diamonds were derived from the graphite. Y-74123 and Y-790981 have been examined by stepped combustion as part of a larger survey involving twelve ureilites which is to be published elsewhere (GRADY et al., 1982b). The carbon release profiles of Y-74123 and Y-790981, and ALHA77257 (for comparison) are shown in Figs.
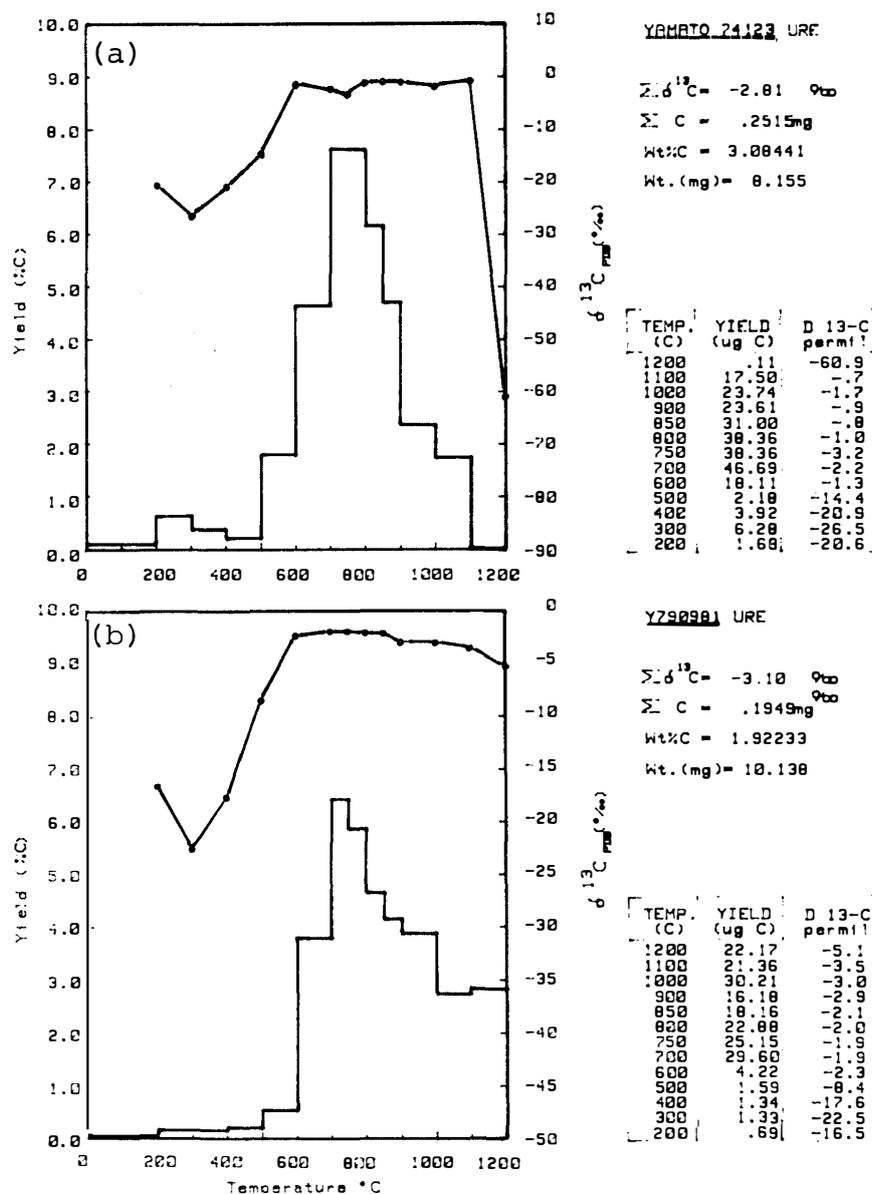
Fig. 6. C release pattern (——) and $\delta^{13}C$ values (—·—) during stepped combustion
of: (a) Y-74123, (b) Y-790981.

6a, 6b and 7 respectively. The three profiles are typical of all the specimens studied, in that a small amount of carbon is liberated at temperatures between 200–400°C, with the bulk being released above 500°C. The low temperature carbon phase, isotopically light in composition (−20 to −30‰) is presumably organic in nature, and could be from indigenous organic compounds. However, it is more likely to represent terrestrial contamination. Whatever the source, it is clearly resolvable from the major carbonaceous phase liberated at higher temperatures. ALHA77257 shows, in addition to a low temperature isotopically light phase, a component of carbon liberated below 200° C, with an isotopic value enriched in $^{13}C$, at around 0‰. The likely origin of such a component, with its very low release temperature is as a result of the presence of terres-
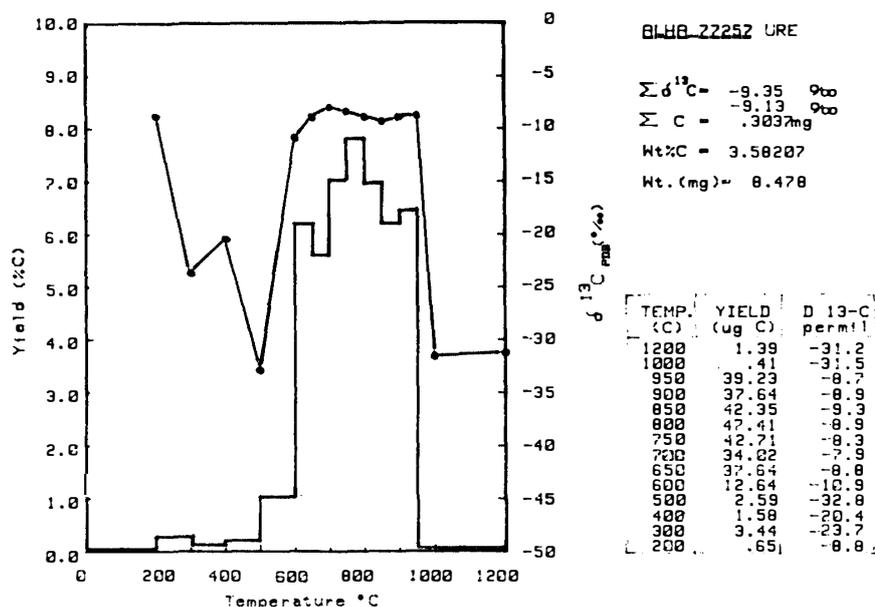
*Fig. 7. C release pattern (——) and $\delta^{13}C$ values (—·—) during stepped combustion of ALHA77257.*

trial inorganic weathering products, or trapped atmospheric gases, incorporated during the lifetime of the specimen of the Antarctic ice. Other Antarctic meteorites, particularly ordinary chondrites, collected by the United States recovery teams show this very low temperature phase (GRADY et al., 1982a), but it was not seen in any of the Yamato specimens studied here.

The major release of carbon occurs in the ureilites between 600–900°C, and is from the combustion of graphite and diamond. (N.B. carbon is liberated from Y-790981 up to 1200°C—this is due to incomplete combustion, caused by a depletion in the oxygen source.) From examination of standard materials, it is known that diamond combusts at a slightly higher temperature than graphite. It is noticeable that there is no apparent difference in isotopic composition between the two components, from which it may be inferred that the diamond was generated from the graphite, presumably by a shock mechanism. The $\delta^{13}C$ value of the graphite/diamond intergrowths in both Yamato samples is around $-2‰$; that of ALHA77257 is close to $-9‰$. Analysis of a suite of ureilites has shown that there is not a spectrum of isotopic compositions of the graphite/diamond, but rather that they fall into two distinct groups (GRADY et al., 1983). This division is not related to petrographic features, such as texture, but may be an indication that the ureilites were derived from two different parent bodies.

## Acknowledgments

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