LIGHT NITROGEN, ITS COMPOSITION, ASSOCIATION AND LOCATION IN THE ALLENDE AND MURCHISON METEORITES. I.P. Wright, S.J. Norris and C.T. Pillinger, Department of Earth Sciences, University of Cambridge, Cambridge CB2 3EQ, U.K. and R.S. Lewis and E. Anders, Enrico Fermi Institute, University of Chicago, Illinois 60637, U.S.A.

The first evidence for isotopically very light nitrogen in C-chondrites was reported by Kung and Clayton (1). They obtained $\delta^{15}N$=-66°/oo for an Allende HF/HCℓ residue, and inferred from isotopic balance that a still lighter component, of possibly -500°/oo, might exist in this sample. Stepped combustion of similar residues (2,3) indeed showed light N, but only to -115°/oo. The release of this light N correlated with that of anomalous noble gases (especially s-process Kr, Xe and Ne-E; less closely with CCFXe). Bulk Allende gave less extreme values, the lowest $\delta^{15}N$ found in pyrolysis of a powdered sample being -90°/oo at 900°C (4).

Using stepped pyrolysis or combustion techniques similar to those of the Clayton group and Frick and Pepin, respectively, we have undertaken nitrogen isotopic analyses of bulk Allende and Murchison as well as acid-insoluble fractions from both. All nitrogen released by either extraction method was analysed quantitatively and isotopically in a static mass spectrometer.

Allende Stepped pyrolysis of two bulk Allende size fractions (<50μ and >50μ) seems to demonstrate that a light nitrogen component resides preferentially in the finer, matrix fraction. Overall, the <50μ sample affords a $\delta^{15}N$ of -24°/oo from 26ppm nitrogen, compared to +2°/oo and 31ppm for the >50μ material. The 800-900°C temperature step in each case gave the minimum $\delta^{15}N$ encountered, -86°/oo and -49°/oo, respectively. Qualitatively, these results are in good agreement with the data of (4). A colloidal, acid insoluble residue (Allende BB) had a very light bulk composition ( -90 °/oo vs -66°/oo of (1)), but its most extreme value upon stepped pyrolysis was only -128°/oo at 1000°C (Fig. 1).

Murchison Pyrolysis of size fractions again suggests that the light component is associated with matrix. A <50μ split yielded 424ppm of nitrogen with a $\delta^{15}N$ value of +33°/oo whereas replicate samples of unsieved bulk Murchison gave yields of 369ppm ($\delta^{15}N$=+41°/oo) and 281ppm ($\delta^{15}N$=+44°/oo) from combustion The bulk nitrogen abundances obtained here are low compared to those published by others but so too is the bulk carbon from this particular Murchison specimen (5).

More extreme $\delta^{15}N$ values were obtained for the fine (<1μ) noble gas rich residue 2C10f. This sample, prepared by treatment with acids and alkaline oxidising agents (6), on pyrolysis released 5190ppm N over a broad temperature range (600-1200°C), but most prolifically from 900 to 1100°C. The nitrogen was extraordinarily light, all fractions falling between -210°/oo and -270°/oo. Stepped combustion of 2C10f (Fig.2) showed singularly light N in the main fractions at 600 and 700°C, but distinctly heavier components at other temperatures. In stark contrast, the medium, 1-3μ, fraction 2C10m gave an order of magnitude less N, with positive rather than negative $\delta^{15}N$ values.

Discussion As a result of the present study, the $\delta^{15}N$ of meteoritic light nitrogen has been reduced from -115°/oo to -273°/oo, well towards the -500°/oo value postulated for "light planetary nitrogen" (7). We have no way of knowing if this isotopic composition is at its limit as the measured samples could still contain heavier types of nitrogen. However, a $\delta^{15}N$ of -273°/oo is already sufficiently low to require a nucleosynthetic process, probably thermonuclear reactions in stars.

The two Murchison separates contain several other isotopically anomalous components, in varying amounts: CCFXe, s-Xe, heavy C (≡$\delta^{13}C$+1100°/oo) (5), and

Ne-E(L). To see which of these anomalies correlate with light N, we calculate nominal amounts of light N in the two samples (Table) by assuming that their nitrogen is a binary mixture of the most extreme components seen in the Murchison samples: $-273^{o}$/oo and $+58^{o}$/oo. The light N value for the medium fraction is only an upper limit, as there is no clear evidence for light N in this sample.

The fine fraction is greatly (>166x) enriched in light N, and all of the other anomalous components, only CCFXe shows a similar enrichment (570x). We have shown previously that CCFXe is located in colloidal carbon of fairly normal composition, $\delta^{13}C=-38^{o}$/oo (5) which is consistent with a solar system origin. But the apparent association with nitrogen of $\delta^{15}N=-273^{o}$/oo argues strongly for a nucleosynthetic origin of CCFXe, if N and CCFXe indeed belong to the same phase. We cannot yet prove such an association on the basis of release temperature, as the data for the C and N combustion do not match up exactly. This may be due to differences in temperature programme (our carbon experiment involved repeated combustion at 500°C), or it may mean that N is released later than C, owing to slower breaking of C–N bonds during oxidative degradation. It appears that N is chemically bound to the carbonaceous phase as its release temperature can be lowered 300°C by combustion. Also, the 2C10f/2C10m ratios for C and N are virtually constant at *ca* 18 (Table)

On the other hand, the heavy, +1100°/oo, carbon, in Murchison separates seems to correlate with *s*-Xe, as shown by the similar enrichment factors for 2C10f (3.5x and 2.9x). There ought to be an isotopically distinctive nitrogen component associated with heavy C but the prime candidate, the small amount of nitrogen released by combustion from 2C10f at 900-1100°C is hardly anomalous at $-14^{o}$/oo. Some dilution by very light nitrogen could, of course, be masking a higher $^{15}N$ content.

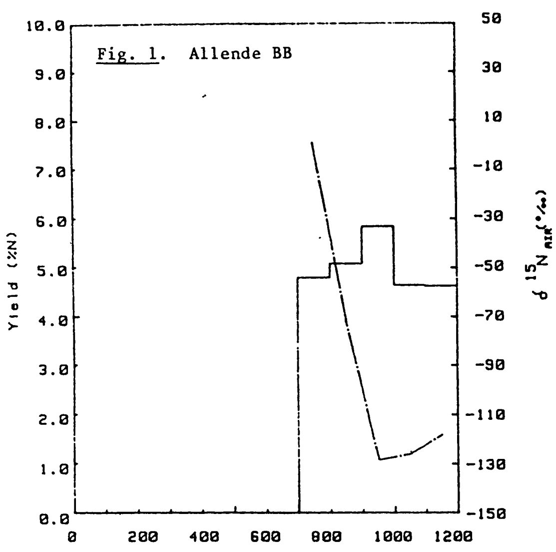

Fig. 1. Allende BB

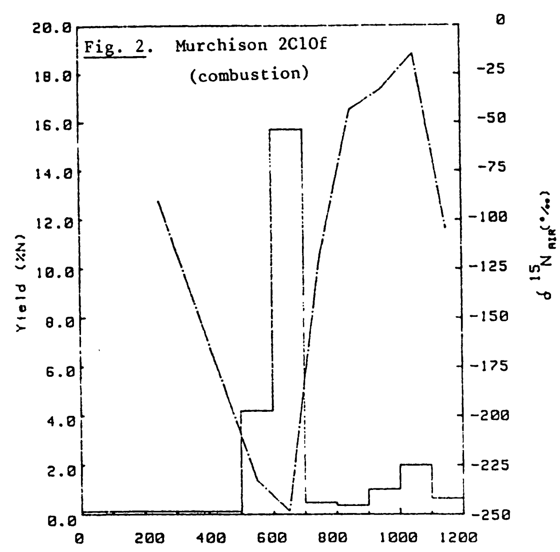

Fig. 2. Murchison 2C10f (combustion)

| | Wt % | C % | C heavy ppm | N ppm | N light ppm | $Xe_s^{130}$ $10^{-10}$cc/g | $Xe_f^{136}$ $10^{-10}$cc/g |
|---|---|---|---|---|---|---|---|
| 2C10m | 0.048 | 3.46 | 1300 | 293 | <26 | 0.66 | 0.73 |
| 2C10f | 0.078 | 65.0 | 4500 | 5190 | 4400 | 1.90 | 419 |
| f/m | | 18.8 | 3.5 | 17.7 | >166 | 2.9 | 574 |

References. (1) Kung & Clayton, EPSL 38, 421-435 (1978) (2) Frick & Pepin, EPSL 56, 64-81 (1981) (3) Frick & Pepin, Meteoritics in press (4) Thiemans & Clayton, EPSL 47, 34-42 (1980) (5) Swart et al., Science in press (6) Alaerts et al., GCA 44, 189-209 (1980) (7) Geiss & Bochsler, GCA 46, 529-548 (1982)