

NITROGEN CONCENTRATION OF INTERSTELLAR DIAMOND

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The isotopic compositions of nitrogen and xenon provide the strongest possible arguments that nanometer-sized diamond (sobriquet C₈) in primitive meteorites originated as interstellar dust from a presolar source. As a corollary, nitrogen and noble gas measurements have been used to infer the presence of C₈ in meteorites of C1 to C3 type, and both ordinary and enstatite chondrites, an interpretation which has been confirmed by isolation of appropriate material either by workers at Chicago and/or ourselves. Huss and Lewis (1989) have been prepared to go even further, and use noble gas measurements made on HF/HCl residues as a means of estimating the abundance of C₈ (and other interstellar grains) in various meteorites. Whilst this is a convenient way of rapidly extending the database, for the purpose of learning about the events which took place in the early solar nebula, it relies on the premise that all the C₈ in the solar system had the same provenance or was intimately mixed. Both these concepts are fundamental and require substantiation before we proceed too far along such a route. Here, we explore the potential of nitrogen abundance for answering questions concerning the source of interstellar diamond: does it have a common source or, like SiC, the other well-characterised component of interstellar dust, did a number of stars contribute to its formation? One of the advantages we see for using nitrogen as an indicator of homogeneity of the nebula is that it exists in sufficiently high abundance for virtually every crystal of diamond to contain at least one nitrogen atom as a part of the structure, rather than trapped as an implanted species

Up to now our analytical efforts to investigate C₈ have focussed on obtaining precise measurements of the isotopic compositions of C and N (Arden *et al.*, 1989). To this end, a stepped combustion extraction method was employed, with small temperature increments to obtain plateau $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values, which would indicate resolution between residual traces of contaminants and the diamond *per se*. Such experiments are very tedious and difficult to replicate, especially since the C and N were measured on separate aliquots using two different extraction lines and mass spectrometers. Nitrogen abundances were calculated from the two sets of release data but were potentially subject to errors from sample heterogeneity, weighing and transfer problems. Thus it was impossible to decide whether the variations between 2000 and 7500 ppm reported by Ash, (oral contribution to the 20th Lunar and Planetary Science Conference) were real. A new technique for the simultaneous measurement of carbon and nitrogen abundance is now being explored.

The samples chosen for this study have been prepared according to the procedure described by Arden *et al.* (1989), omitting (except in the case of Cold Bokkeveld) the last stage at which diamond is separated from spinel, because ammonia - a potential nitrogen contaminant - is used for this purpose. The total carbon content of each residue was measured by a bulk, sealed tube combustion method using a system calibrated with diamond chips of the appropriate size (Russell *et al.* 1990). The major analytical programme was undertaken on apparatus described earlier (Boyd *et al.* 1988) but modified to include a capacitance manometer and take-off vessels to collect carbon dioxide. The following extraction procedure was devised based on our knowledge of full stepped combustion analyses but its final format is still evolving. The samples are first burned at 300°C and 400°C to remove any organic contamination and trapped atmospheric nitrogen, then combusted at 700°C for two hours to oxidise all the C₈ in a single step, without liberating the higher temperature carbon components in the residue. Towards the end of the major combustion step a trap is cooled to -175°C which freezes out around 90% of the released CO₂. The remaining 10% is recovered during clean-up of the nitrogen (Boyd *et al.* 1988) and the total CO₂ is quantitated and combined into a vessel which can be removed and transferred to a dynamic mass spectrometer for precise carbon isotopic determination if the sample size is sufficiently large. However, no data of this kind are reported here.

The yield of purified nitrogen is measured directly from the ion current of a static mass spectrometer as a by product of the $\delta^{15}\text{N}$ measurement. All the results obtained by the study are reported in the table below. From the $\delta^{15}\text{N}$ measurements, this method clearly does not successfully resolve N attributable to C₈ from that of other sources. For this reason, we have

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calculated the N concentration to a normalised $\delta^{15}\text{N}$ of -340% (Ash *et al.* 1989) assuming the contaminating nitrogen to be 0% . We anticipate that the need for this correction procedure can be eliminated by refining the measurement technique.

Sample	Wt.% C	$\delta^{15}\text{N}$ (‰)	N conc ⁿ . ppm	[N] in C δ ppm normalised to $\delta^{15}\text{N}-340\%$
Allende	13.22	-260.4	8977	6882
ARC IVDS1	23.26	-303.9	6867	6137
Bulk C=24%	26.20	-308.4	3918	3553
Murchison M2	17.83	-272.5	10738	8623
Bulk C=21%	17.95	-326.9	7866	7565
Cold Bokkeveld	63	-313.8	8926	8237
Bulk C=62%	31.2	-297.7	10003	8785
Inman	18.30	-286.3	3254	2740

To some extent, the limited database obtained is internally consistent but there are variations which cannot be ignored without more replicates. On two occasions, once for Allende and once for Cold Bokkeveld, the yield of carbon from the conjoint experiment is about half the value expected from the bulk analysis, which could infer a sample heterogeneity. In both instances, the nitrogen concentration calculated for C δ agrees within 10% with a measurement where carbon yields were concordant. One Allende sample gave a nitrogen in C δ concentration that was well below the other two. In this experiment the carbon yield was slightly above the bulk, which could reflect contamination from carbon with a low nitrogen content. The only Inman analysis gives the lowest nitrogen concentration; there is no bulk carbon value or replicate yet for this sample, which is in short supply, and so used conservatively.

The most consistent results obtained come from measurements of the two type CM2 meteorites; four measurements of the nitrogen concentration in C δ give a mean of 8302 ± 470 ppm, which is about 25% higher than the highest value yet obtained for the CV3 sample Allende. In our previous studies, which did not enjoy the benefit of conjoint C and N measurements, it was noted that C δ from CM2's appeared to be richer in nitrogen than Allende, and the latter appeared to be more variable, particularly when samples from different sources were compared.

Whether or not a variable nitrogen content for C δ is confirmed, one significant observation can be made from this study: the abundance of nitrogen easily exceeds the range encountered in terrestrial diamonds (which have a maximum of about about 3000 ppm N), whether they are natural or artificially produced. This undoubtedly points to a novel, and possibly non-equilibrium, method for nitrogen incorporation into interstellar diamond.

References

Arden *et al.* 1989 Lunar Planet. Sci. Conf. XX, 21-22; Boyd *et al.* 1988 J. Phys.E.Sci. Instrum. **21**, 876-885; Huss and Lewis 1989 Abstract 52nd Meteoritical Soc.p.91; Russell *et al.* 1990, this volume.