# STEP-WISE CARBON ISOTOPIC ANALYSES OF THE ALLENDE METEORITE.

P.K. Swart, M.M. Grady and C.T. Pillinger. Planetary Sciences Unit, Dept. of Earth Sciences, University of Cambridge, Cambridge CB2 3EQ, U.K.

It is beyond dispute that carbon exists in a multitude of forms within meteorites and much interest has been paid to the acid resistant carbonaceous phases, a possible site of heavy noble gases and other anomalous components. Considerable effort has been spent in separating such material, notably from Allende, with the aim of distinguishing and characterising more precisely the location of anomalous gases. Few carbon isotopic analyses have been undertaken probably because of the inherent contamination risks during sample isolation procedures. The major study performed (1) must have been a great disappointment to its instigators because of the apparent homogeneity of the Allende separates in terms of carbon isotopes. In addition work has been conducted on the step-wise combustion of carbon and sulphur from Allende separates (2). Carbon with a light isotopic signature was released at low temperatures, becoming progressively heavier as temperature increased. This isotopic change was discussed as being a kinetic isotope effect and hence an experimental artifact rather than of analytical significance (1). Such an interpretation would be correct if all the carbon was present as either a single phase or a homogeneous mixture of closely related phases readily available to the oxygen.
Sample preparation: Our approach was to step-wise combust samples of <50 and >50μm Allende, without prior removal of the mineral matrix, in 100 or 200°C temperature steps from 200 to 1150°C. Preparation procedures were similar to those described elsewhere (3) and typical blanks for each step are <0.2μg. Combustions were carried out in 20 torr of $O_2$ for 30 minutes; $CO_2$ and $SO_2$ were separated using a variable temperature cryogenic trap; yields were measured with a capacitance manometer and the isotopic measurements made using a VG Micromass 602-E.
Results: Fig 1 shows the release pattern of C and S from the >50μm Allende sample and carbon isotopic compositions of both size fractions. The lower temperature of the carbon release maximum (Fig 2) in the finer fraction can be attributed to the more easily oxidisable nature of the sample. Similarly, a slight difference (0.21 wt % compared to 0.26 wt %) in total yield may also be explained by the variation in particle size. Isotopically the two samples showed similar trends, exhibiting a very negative (-23 to -25‰) low temperature release of carbon followed by several other forms exhibiting distinctly different isotopic signatures (ie. -16, -21, -7‰, see >50μm fraction Fig 1) at higher temperatures. Such a release pattern would not be expected if the change in isotopic composition were solely attributable to a kinetic effect. Other meteorites examined in a step-wise combustion mode, Bishunpur and ALHA 77299, also revealed complex patterns of carbon release and isotopic composition (Fig 3). We therefore suggest that the changes in carbon isotope values shown in Fig 1 are real and reflect the different forms of carbon oxidisable at varying temperatures. It may be that the presence of silicate phases protect carbon entities and allow their differentiation by step-wise heating. It is perhaps significant that combustion of Allende residues does not reveal the high temperature (1100°C) phase observed here. The $^{13}C$ enrichment in the high temperature carbon neatly counterbalances the lighter component (-21‰) released at intermediate temperatures (600-700°C) so that simultaneous combustion of both might be expected to give an isotope composition of ca -17‰, the value consistently achieved by total analysis of acid residues. A bulk isotopic value for Allende can be calculated by summing the weighted values of the individual temperature steps. The value obtained

compares well with published bulk data for Allende, -19.5‰ (3) and -16.4‰ (4). Conclusions: The step-wise oxidation technique provides an easy, contamination free method of identifying the carbon isotopic composition of the varying phases in meteorites. The wide spread of data points for Allende focuses attention on the meaning of bulk $\delta^{13}C$ values, which are no more than a sum of several individual carbon components varying greatly in isotopic composition. Through the use of even smaller combustion steps or longer extraction intervals, more data concerning carbon and its isotopic composition could be gleaned and correlations with rare gas release achieved. We would like to thank Ed Anders for stimulating our interest in the possibilities of stepwise combustion.

References: (1) Ott et al. (1981) GCA 45, 1751-1788. (2) Frick & Pepin (1980) LPS XI, 303-305. (3) Grady et al. (1981) Abs. 44th. Meteoritical Society Meeting p.28. (4) Chang et al. (1978) LPS IX, 157-159.