

SEPARATION OF SPALLATION AND TERRESTRIAL  $^{14}\text{C}$  IN CHONDRITES

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Weathering products and contamination severely hamper our ability to accurately measure the  $^{14}\text{C}$  spallation component in meteorites, but can give insights into a sample's terrestrial history. A procedure has been developed to measure the  $^{14}\text{C}$  in these components using CO and CO<sub>2</sub> separations from temperature extractions from 200-500mg of material. The Bruderheim (L6) chondrite was chosen as a standard following the practice of previous workers [1,2], cross-checked against Peace River (L6), Abee (EH4) and Juvinas (EUC). Low temperature fractions (<900°C) give  $^{14}\text{C}$  signatures consistent with a modern terrestrial  $^{14}\text{C}$  source; melt fractions show elevated levels attesting to a spallogenic origin. Higher yields of CO in the melt fraction are less affected by the low levels of experimental contamination than the CO<sub>2</sub>. This fraction gave a mean CO:CO<sub>2</sub> ratio in Bruderheim of  $81.6 \pm 7.7$ ; the ratio of the spallation component is  $79.8 \pm 8.1$ . These values suggest equilibrium release of gases on the olivine-silica-pyroxene-iron buffer. This is corroborated by approximately equal release of the two components at 900°C. The chondrites gave an average saturation level of  $54.3 \pm 2.9$  dpm/kg; the achondrite gave  $49.6 \pm 2.0$  dpm/kg. No clear correlation with oxygen content is apparent, though shielding effects have yet to be evaluated.

Measurement of  $^{14}\text{C}$  from separated gas species from the step-wise pyrolysis of meteorites has been undertaken to further constrain and isolate the spallation component, and assess the role of recent weathering of meteorites, with particular reference to Antarctic samples. A series of samples of Bruderheim, ranging from 306 to 486mg, were step-heated under helium in a coil furnace, then taken to melt temperatures using induction heating of the molybdenum crucible. For each temperature step chosen, the evolving gases were removed by continuously cycling the helium using a peristaltic pump, trapping the condensibles in a variable temperature trap held at -180°C, and passing the non-condensibles over heated (to 650°C) cupric oxide, oxidizing any Co and CH<sub>4</sub> to CO<sub>2</sub>, which was then trapped in two liquid nitrogen-cooled traps. CO<sub>2</sub> was further purified by fractional distillation, diluted to give  $\approx 3$ cc and transferred to vials. Each aliquot is reduced over heated lithium to produce lithium carbide; water is added to generate acetylene, and this is dried and bled into a cracking chamber. A glow discharge is created between two aluminium pellets by means of a current controllable 60Hz, 1kV AC power supply, dissociating the gas and depositing 200-400 $\mu\text{g}$  of low density graphitic material on the surface of each pellet [3,4]. Each pellet provides a target for accelerator mass spectrometry, with  $^{14}\text{C}$  analysis carried out as described elsewhere [4]. The number of  $^{14}\text{C}$  atoms for each aliquot is determined and backgrounds are subtracted as outlined elsewhere [5].

To assess each fraction for contamination it is assumed that the recovered gas is entirely of recent origin, and the expected  $^{14}\text{C}$  content is calculated. This is subtracted from that observed to give a delta-14 value, relative to modern. Figure 1 shows the results from Bruderheim together with single runs on each of the other three falls. Almost all fractions up to 900°C plot within error (approximately three times the size of the symbols) of modern, indicating a terrestrial source. Above this temperature, breakdown of silicates and any indigenous graphite distorts the picture. The elevated levels from the melt fraction (here designated as 1600°C for Bruderheim and 1650°C for the other falls for clarity) are due to the spallation component, which is particularly evident in the CO fraction. Graphite in Abee and Juvinas act as natural dilutants, hence the low delta-14 for two of the other falls.

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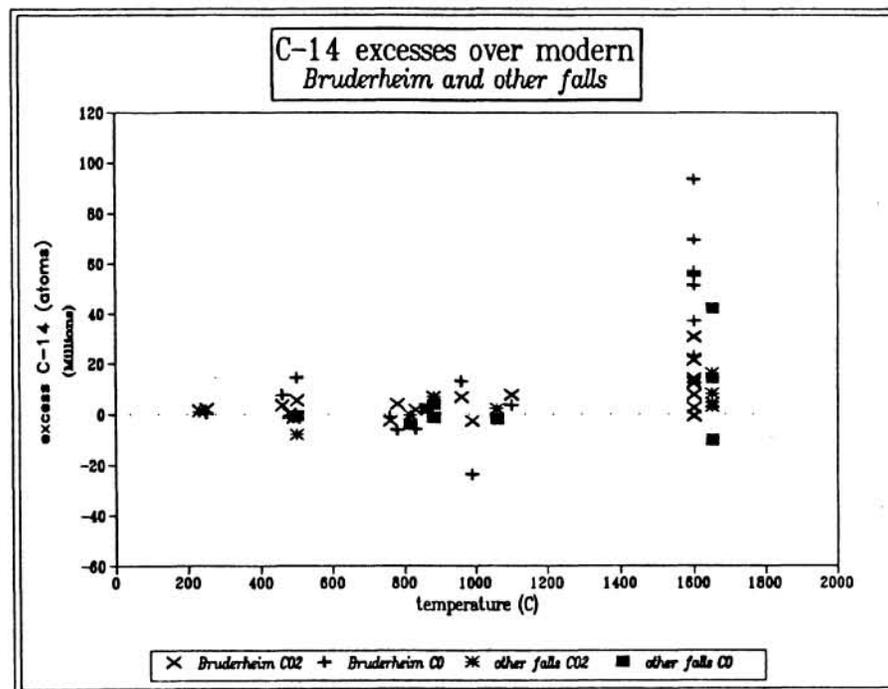


Figure 1.  $^{14}\text{C}$  excesses in Bruderheim and three other falls as a function of temperature and gas species. Zero represents a modern (terrestrial) carbon source. The excess  $^{14}\text{C}$  from spallation is clearly seen in the melt (1600°C) CO fraction.

The variable excess  $^{14}\text{C}$  in the melt is explained by incomplete diffusion for melts of less than six hours duration. Unlike the combustion method employed by Jull [2], whereby oxidation of the carbon facilitates rapid release of the spallation component, these pyrolysis extractions are controlled by diffusion, with expected diffusion rates of  $\approx 10^{-5.5}\text{cm}^2/\text{s}$  [6]. A diffusion rate of  $10^{-6}\text{cm}^2/\text{s}$  has been estimated from the results of this work on Bruderheim [7], requiring a minimum of five hour melts for complete extraction of the spallation component.

Kotra *et al.* [8] showed that finely ground carbonates decompose between 400°C and 800°C during continuous heating. Some finely crushed dolomite was tested using the same procedure adopted here for meteorites and found to completely decompose between 550°C and 920°C. This suggests an alternative to the hydrolysis method of separating weathering components used by Jull, *et al.* [9,10]. This has been used to assess the weathering component of nine Antarctic meteorites, and is reported elsewhere [5].

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