

# Fullerenes in Allende meteorite

**SIR** — The detection of fullerenes ( $C_{60}$  and  $C_{70}$ ) in deposits from meteor impacts<sup>1-3</sup> has led to renewed interest in the possibility that fullerenes are present in meteorites. Although fullerenes have not previously been detected in the Murchison and Allende meteorites<sup>4</sup>, the Allende meteorite is known to contain several well-ordered graphite particles which are remarkably similar in size and appearance to the fullerene-related structures carbon onions and nanotubes<sup>5</sup>. We report here that fullerenes are in fact present in trace amounts in the Allende meteorite.

The fullerenes were detected in

impact deposits<sup>1</sup> and the residues collected from the Long Duration Exposure Facility<sup>2</sup> formed as a result of impact synthesis, but comparable to the levels reported in sedimentary deposits associated with the Cretaceous/Tertiary impact event<sup>3</sup>, possibly produced by extensive wild fires. Because there is no evidence of any shock event in the Allende meteorite's history, the fullerenes found in this meteorite were probably not the result of impact synthesis. Failure to detect fullerenes in previous studies of meteorites<sup>4</sup> could be because of the well-known inhomogeneous distribution

duced under conditions involving many competing processes; the presence of hydrogen need not preclude fullerene production in the interstellar medium.

In some interstellar environments, hydrogen is present at  $H/He < 10^{-9}$ , levels much lower than the mean cosmic abundance<sup>9</sup>. Fullerenes can form in the outflows from WC stars<sup>10</sup>, for example, which consist mainly of He and C and little or no hydrogen. In fact, the presence of hydrogen may lead to the conversion of fullerenes,  $C_{60}$ , to fullerenes,  $C_{60}H_n$ , resulting in a wide range of molecules from fully aromatic to fully aliphatic, which could explain the variability observed in interstellar/stellar spectra.

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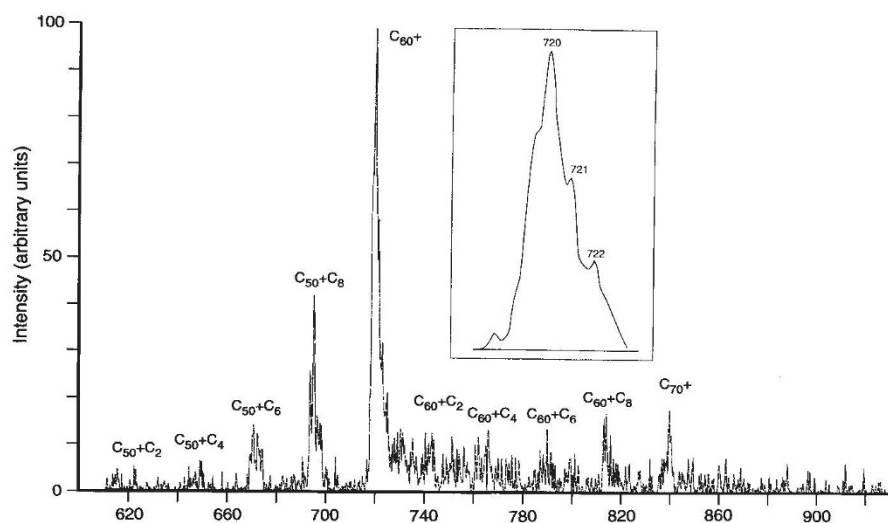
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## Low-frequency oscillation

**SIR** — The singular spectrum approach (SSA) has recently been used to identify a low-frequency oscillation in the global surface air temperature record<sup>1</sup>. SSA<sup>2,3</sup> considers  $M$  lagged copies of a series  $X(t)$  sampled at equal intervals  $\tau$ ,  $X_i = X(t_0 + i\tau)$ ,  $i = 1, N$ , and estimates the eigenvalues  $\lambda_k$  and the corresponding eigenvectors  $\rho_k$  of their time-lag covariance matrix  $C$ , where  $1 < k < M$ . The eigenvectors are of interest if, among the  $k$  eigenvalues, there exist several whose magnitudes are significant. As such, SSA can be used to separate signal from noise.

The figure (a) shows the estimated eigenvalues,  $\lambda_k$ , of the time-lag covariance matrix  $C$  (with  $M = 40$ ) for the detrended IPCC global temperature record<sup>4</sup>. Detrending the data is recommended in such cases (when  $M$  represents a fairly large fraction of  $N$ ); otherwise the first eigenvalues representing the trend can mask or alter long-period cycles that might be present. Here we detrend the data throughout by removing the least-squares line. In agreement with the earlier work<sup>1</sup>, we observe the first two eigenvalues appreciably higher than the so-called 'noise floor'. These two eigenvalues correspond to eigenvectors (insert in the figure) that exhibit an oscillation of approximately 60–70 years. Eigenvalues far from the 'noise floor' are often



Laser desorption (reflectron) mass spectrum from sample 15/21. A major peak occurred at  $m/z = 720$  ( $C_{60}^+$ ) and a less prominent peak at  $m/z = 840$  ( $C_{70}^+$ ). Fragmentation peaks from  $C_{52}^+$  to  $C_{58}^+$  and  $C_{62}^+$  to  $C_{68}^+$  were observed at power levels much higher than those originally used to detect  $C_{60}^+$  and  $C_{70}^+$  in the Allende extracts. The insert of sample 15/21 shows  $C_{60}^+$  resolved into 3 peaks at 720, 721 and 722 AMU ( $m/\Delta m = 300$ ), corresponding to  $^{12}C_{60}^+$ ,  $^{12}C_{59}^{13}C^+$  and  $^{12}C_{58}^{13}C_2^+$ , respectively. Sample preparation and analysis of the Allende extracts using a KRATOS (reflectron) time-of-flight instrument is described in detail elsewhere<sup>1</sup>. Sample 15/21 was obtained from the Smithsonian Museum (USNM 3529, split 15 position 21).

separate Allende samples by laser desorption (linear and reflectron) time-of-flight mass spectrometry. The observed  $C_{60}^+/C_{70}^+$  peak ratio is 12 (see figure). The  $C_{60}$  content for Allende was estimated to be 0.1 p.p.m., considerably less than the fullerene concentrations reported for both the 1.85-billion-year-old Sudbury

of organics within meteorites<sup>6</sup> or perhaps because fullerenes are present at concentrations too low to be detected previously.

In addition to fullerenes, we detected many polycyclic aromatic hydrocarbons (PAHs) in the Allende meteorite, consistent with previous reports<sup>6</sup>. In particular, we detected benzofluoranthene and corannulene ( $C_{20}H_{10}$ ), five-membered ring structures which have been proposed as precursors to the formation of fullerenes in the gas phase<sup>7</sup>. The observation of fullerenes together with PAHs suggests that PAHs may have been involved in fullerene synthesis, perhaps within circumstellar envelopes or other sites in the interstellar medium. This is somewhat surprising, as such environments generally contain a high abundance of atomic and/or molecular hydrogen<sup>8</sup>. However, the production of fullerenes in sooting flames<sup>7</sup> implies that fullerenes can be pro-

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