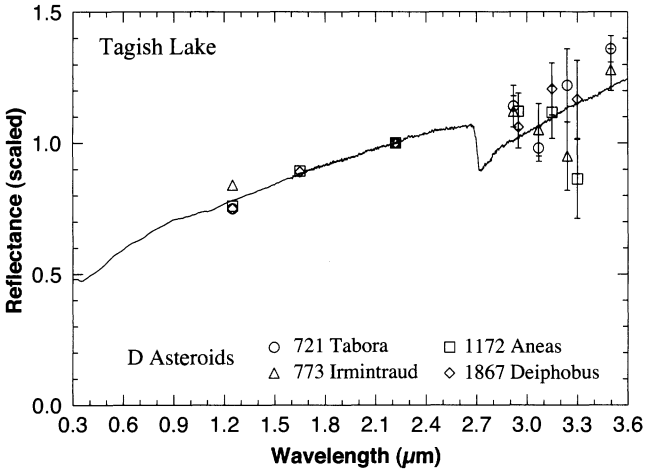
Fig. 3. The 3-μm reflectance spectra (*14*) of four D-type asteroids compared with visible and FT-IR spectrum of the Tagish Lake meteorite sample.



motion resonance with Jupiter. Their IRAS albedos (*8*) of 0.032 and 0.033 are both close to the reflectance of the Tagish Lake sample at 0.55 μm under a viewing geometry of (6, −6) (Fig. 2). More detailed analysis is needed to determine whether ejecta from this asteroid, or other D-type asteroids, can traverse the distance to the chaotic zones [Supplementary fig. 2 (*9*)] when driven by impact ejection energy and the Yarkovsky effect [e.g., (*19, 20*)]. In addition, more extensive telescopic observations are needed to determine whether the apparent spectral difference between 773 Irmintraud and the Tagish Lake meteorite is significant or whether there is a better candidate for the Tagish Lake parent body among other D-type asteroids. The launch efficiency from the Tagish Lake parent body may also be problematic to model because of the low density (1.7 g/cm^3) (*20*) and mechanical weakness of the Tagish Lake meteorite. The kinetic energy of impactors may be absorbed more efficiently on the Tagish Lake parent body than on stronger bodies, resulting in a smaller kinetic energy of the ejecta.

Linking asteroid types to well-studied meteorite classes is a difficult endeavor because extraterrestrial materials that fall to Earth are limited by physical conditions and form an inherently biased sample in time and space. Recovery of the Tagish Lake meteorite suggests that perhaps many more mechanically weak classes of meteorites are destroyed during atmospheric entry before they can be recovered.

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The Organic Content of the Tagish Lake Meteorite

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The Tagish Lake meteorite fell last year on a frozen lake in Canada and may provide the most pristine material of its kind. Analyses have now shown this carbonaceous chondrite to contain a suite of soluble organic compounds (~100 parts per million) that includes mono- and dicarboxylic acids, dicarboximides, pyridine carboxylic acids, a sulfonic acid, and both aliphatic and aromatic hydrocarbons. The insoluble carbon exhibits exclusive aromatic character, deuterium enrichment, and fullerenes containing "planetary" helium and argon. The findings provide insight into an outcome of early solar chemical evolution that differs from any seen so far in meteorites.

The biogenic elements have a long cosmic history that spans their stellar nucleosynthesis through complex stages of interstellar, nebular, and planetary processes that preceded life. Some of this prebiotic chemical evolution is recorded in carbonaceous chondrites, a primitive type of meteorite containing organic carbon. Although a majority of this carbon is bound in a kerogenlike, insoluble material, CM and, to a lesser extent, CI chondrites also contain a large and complex suite of soluble organics (*1*). The presence of amino acids and other compounds having terrestrial counterparts has led to speculation that comets and meteorites could have seeded early Earth with bioprecursor molecules (*2*).

The Tagish Lake meteorite fell in January 2000 in Canada and is a carbonaceous chondrite of uncertain classification (*3*). Its fall, brief environmental exposure, and collection at subfreezing conditions were exceptional, raising expectations for a pristine sample. We report here the findings from analyses of the soluble and insoluble organic content of about half (4.5 g) of a pristine (*3*) stone surrounded by fusion crust. We conducted water and solvent extractions of interior powdered samples and nuclear magnetic resonance (NMR) analyses of the insoluble organic material obtained after dissolution of mineral phases. The analytical procedures followed closely those established for the study of similar carbonaceous meteorites (*4–7*).

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The Tagish Lake meteorite contains about 100 parts per million (ppm) of soluble organic compounds (Table 1). The most varied between them are the aliphatic dicarboxylic acids of the water extract (*5*), a homologous suite of saturated and unsaturated compounds of up to 10 carbon atoms (*8*). Linear, saturated acids predominate, decreasing in amount with increasing chain length (Fig. 1) and occurring also as dicarboximides in the case of the more abundant species. A sample of the Murchison meteorite, a well-characterized CM chondrite (*1*), was analyzed under identical conditions and showed a dicarboxylic acid suite of remarkably similar abundance and distribution, the only difference being a lower ratio of linear to branched acids. The similarity extends to the isotopic composition, as shown by the gas chromatography–combustion–isotope ratio mass spectrometry (GC-C-IRMS) of individual diacids (*9*) (Table 2). Although the Tagish Lake compounds were consistently lighter, the $\delta^{13}C$ values were at least 60% of the Murchison counterparts in each case.

Aromatic dicarboxylic acids were also observed in benzene/methanol extracts as a series of imide derivatives of phthalic acid, homophthalic acid, and some methyl and dimethyl homologs not seen in Murchison. A total of nine were detected on the basis of their mass spectra, and $\delta^{13}C$ values of five of these imides were greater than terrestrial values (*10*), varying from +1.5 per mil (‰) for homophthalimide to +17.1‰ for a dimethyl homolog of phthalimide (*11*).

Carboxylated pyridines were found in the water extract with the amino acids, with which these molecules share amphoteric properties and biochemical relevance. Nicotinic acid, its two isomers, and 12 methyl and dimethyl homologs, with substituents both in the aromatic ring and the carboxylated side chains, were identified by gas chromatography–mass spectrometry (GC-MS) of their isopropyl derivatives. These compounds have not been described in Murchison but were searched for in this study and observed as a similar suite in that CM also.

Most of the other water-soluble compounds found in Tagish Lake are less abundant than in Murchison by one (carboxylic acids) to three orders (amino acids). They are also far less numerous, and the amounts of each series are dominated by the lower homologs. This distribution is in stark contrast with the extensive isomerism of CM chondrites and is exemplified by Tagish Lake monocarboxylic acids. These compounds account for a substantial fraction of soluble organics, but only the linear acids up to C_9 were identified and then only in trace amounts beyond C_2; i.e., individual formic and acetic acids are only slightly less abundant than in Murchison (*12*). The case of sulfonic acids is extreme as they are represented in Tagish Lake by only methane sulfonic acid (MSA). A larger $\delta^{13}C$ and a ^{33}S anomaly (opposite sign) for Murchison MSA relative to higher homologs suggested a distinct origin for meteoritic MSA (*13*). Tagish Lake results are consistent with that suggestion.

Tagish Lake aliphatic hydrocarbons include both saturated and cyclic and/or unsaturated species. The normal alkanes predominate with peak abundance at C_{23}. In view of the pristine nature of this meteorite, the finding is interesting in regard to the question of whether the Murchison n-alkanes are indigenous (*14*) or terrestrial contaminants (*6*, *15*). The $\delta^{13}C$ values of several n-alkanes determined by GC-C-IRMS range from −18.8‰ (C_{16}) to −29.0‰ (C_{20}), whereas those of branched and unsaturated and/or cyclo alkanes are similar (−18.4‰ to −27.1‰). These values are in the range of terrestrial organic compounds (*10*); however, other Tagish Lake materials that are considered indigenous, such as the macromolecular carbon, also have low $\delta^{13}C$ values. The low $\delta^{13}C$ value (−25‰) obtained for phenanthrene may indicate a general depletion in ^{13}C in this meteorite's hydrocarbons relative to other organics.

HF/HCl digestion of Tagish Lake powders

Fig. 1. *m/z* 44 (**bottom**) and 45/44 (**top**) traces of a GC-C-IRMS analysis of Tagish Lake dicarboxylic acids (*8*, *9*). 1, succinic acid; 2, methyl succinic acid; 3, glutaric acid; and 4, 2-methyl glutaric acid.

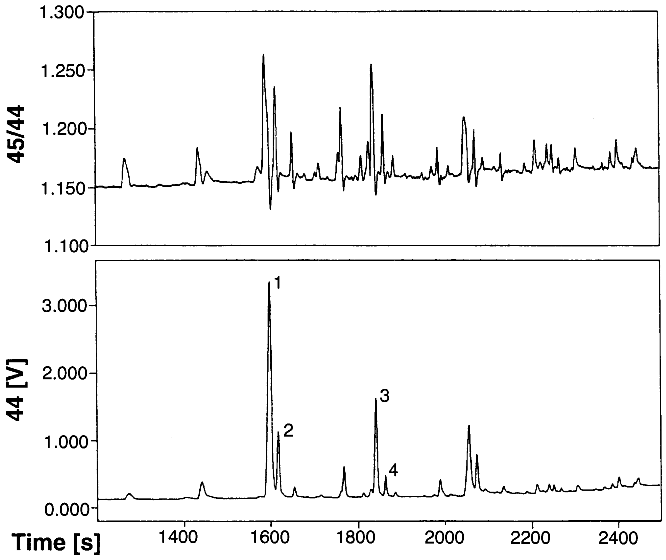


Table 1. Soluble organic compounds in the Tagish Lake and Murchison meteorites.

Class	Tagish Lake		Murchison (*29*)	
	Concentration* (ppm)	Compounds identified†	Concentration (ppm)	Compounds identified
Aliphatic hydrocarbons	5	12	>35	140
Aromatic hydrocarbons	≥1	13	15–28	87
Dicarboxylic acids	17.5	18	>30	17
Carboxylic acids	40.0	7	>300	20
Pyridine carboxylic acids	7.5	7	>7	7‡
Dicarboximides	5.5	4	>50	3‡
Sulfonic acids	≥20	1	67	4 (*13*)
Amino acids	<0.1	4	60	74
Amines	<0.1	3	8	10
Amides	<0.1	1	n.d.§	4 (*30*)
Hydroxy acids	b.d.‖	0	15	7

*Concentrations are based on chromatographic peak intensities and include compounds identified by reference standards and mass spectra. Variability was not estimated at this time, as measurements were obtained by analyses of one meteorite stone. †Compounds identified with reference standards. ‡This study. §Not determined. ‖Below detection (GC-MS).

Table 2. $\delta^{13}C$ values (‰) [by GC-C-IRMS, corrected for esterification (*31*)] of individual meteoritic dicarboxylic acids.

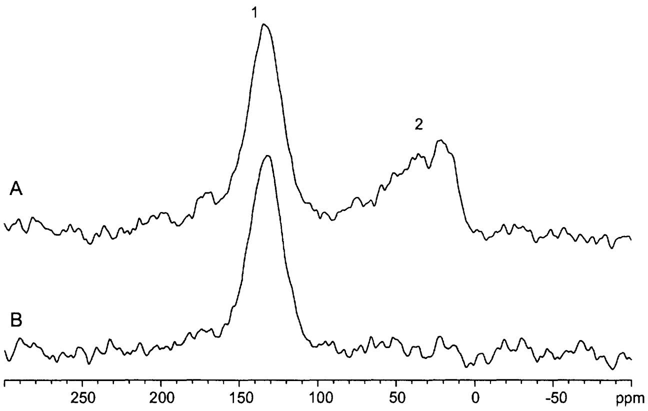
Acid	Murchison	Tagish Lake
Succinic	+28.1 (±1.1)	+22.5 (±0.6)
Methyl succinic	+26.5 (±0.7)	+15.4 (±3.0)
Glutaric	+26.8 (±0.3)	+22.9 (±1.5)
2 Methyl glutaric	+27.9 (±1.0)	+18.6 (±0.7)

yielded ~2.4% of total weight as a carbonaceous residue with the general formula $C_{100}H_{46}N_{10}O_{15}$ accounting for over 99% of total organic carbon. This macromolecular material was analyzed by solid-state ^{13}C NMR (*16*) along with a sample of the Orgueil meteorite, a CI chondrite that compares with Tagish Lake in insoluble carbon content. The spectra (Fig. 2) reveal that the Tagish Lake insoluble carbon is composed of predominantly aromatic material and differs from those of the Orgueil, Murchison, and Allende meteorites, in which both aromatic and aliphatic features are observed (*7*, *17*, *18*). A δD of +930 ± 6‰ was obtained for the residue (*19*), a value consistent with an origin from interstellar aromatic species (*20*) and corresponding to the upper limits of δD values measured for Murchison (+483‰ to +1066‰) (*21*).

The acid-digested material also released fullerenes when further extracted (*22*). Laser desorption mass spectrometry (LDMS) revealed a limited series of higher fullerenes between $C_{60}{}^{+}$ and $C_{160}{}^{+}$, as compared with the Murchison distribution (*23*), which displays a broader range of molecules ($C_{60}{}^{+}$ to $C_{250}{}^{+}$) dominated by the higher fullerenes (Fig. 3). This may be attributed to different compositions of the original nebular condensates and/or to different aqueous and thermal processing. The isotopic composition of both helium and argon in Tagish Lake fullerenes resembles that of the "planetary" gases with $^{3}He/^{36}Ar$ ratios that approach ~0.01, as opposed to the solar value of ~1. Similar isotopic composition and temperature release patterns of the gases were obtained upon combustion of the unextracted macromolecular carbon, suggesting that fullerenes may be the major carriers of planetary noble gases in Tagish Lake (*24*).

These results, when compared with the analytical data available for other carbonaceous chondrites, show that the Tagish Lake meteorite is as unique in its organic chemistry as it appears to be in its mineralogy and petrology (*3*). Although most of its organic constituents are not exclusive, their selective distribution is, and it presents an example of chemical evolution not seen before. The main features of these organics are the dominance of water-soluble carboxyl and dicarboxyl compounds, the paucity of amino compounds, and the aromatic character of the insoluble carbon.

Fig. 2. Solid-state ^{13}C variable amplitude cross-polarization–magic angle spinning NMR spectra of insoluble macromolecular material from (**A**) Orgueil and (**B**) Tagish Lake meteorites. 1, protonated and nonprotonated aromatic C, carboxyl C, and carbonyl C (100 to 250 ppm); 2, aliphatic and aromatic-linked CH_3, CH_2, and aliphatic C adjacent to heteroatom (0 to 100 ppm). Tagish Lake aliphatic ≤7.0%, from repeat analyses (*16*).



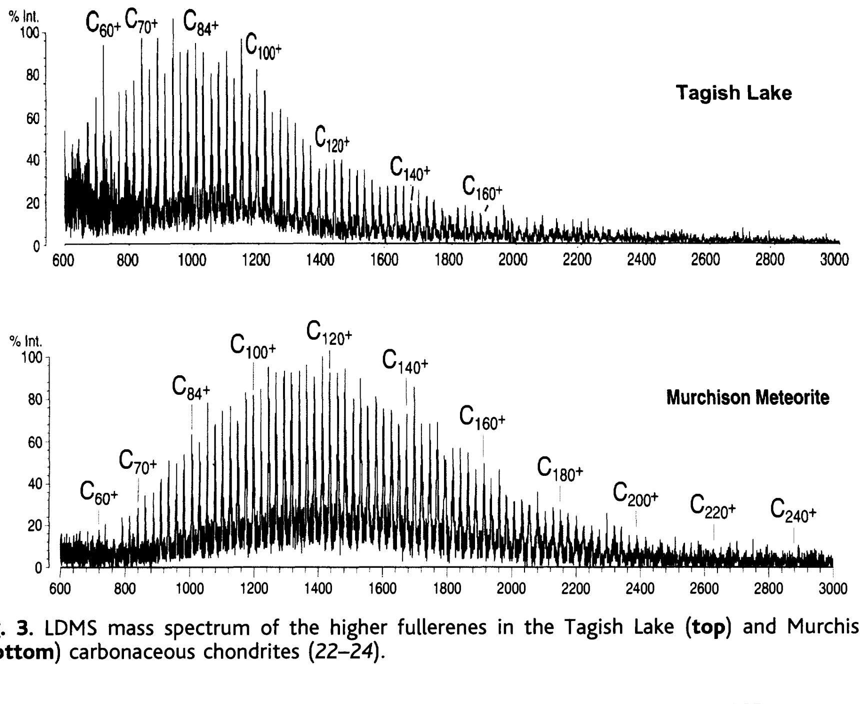


Fig. 3. LDMS mass spectrum of the higher fullerenes in the Tagish Lake (**top**) and Murchison (**bottom**) carbonaceous chondrites (*22–24*).

What can this composition tell us about the origin of its organic matter and the distinct origin of this meteorite? Soluble organics in meteorites have been generally viewed as products of parent body aqueous (and perhaps thermal) processing of reactive, volatile precursors such as water, HCN, NH_3, and ^{13}C-, ^{15}N-, and deuterium-rich interstellar organics (*1*). In the Tagish Lake suite, we find no clear examples of such chemistry, as would be the abundant Murchison amino and hydroxy acids believed to have formed through a Strecker synthesis (*25*). Moreover, some aliphatic compounds, e.g., carboxylic acids and n-alkanes, display a distinct linear chain preference, suggesting catalytic surface processes rather than the random combination of radicals predicted for interstellar molecules (*1*). However, Tagish Lake mineralogy shows evidence of water alteration (*3*), and the $\delta^{13}C$ values of the meteorite aliphatic and aromatic dicarboxylic acids suggest that primitive interstellar materials were incorporated into organics of this meteorite as well (*5*). In view of the compositional and petrological heterogeneity of carbonaceous chondrites, the findings are not mutually exclusive but rather point to a similar, although somewhat broader scenario. In fact, the Tagish Lake organic content would be consistent with an origin from a primitive parent body that, before aqueous processing, was depleted or never acquired volatile compounds and accreted both nebular and interstellar larger molecules such as hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), nitriles, and dinitriles. These latter compounds are common in the interstellar clouds and would account, upon parent body accretion, water exposure, and subsequent hydrolysis, for the carboxylic and dicarboxylic compounds found in Tagish Lake. Support for this chemistry comes from the "prominence of nitriles and other cyano species" found in Tagish Lake extracts (*26*) and our identification of specific dinitriles as dicyanobenzene and dicyanotoluene.

Tagish Lake macromolecular carbon also has features suggestive of an interstellar origin, i.e., aromaticity, elevated δD, and the pyrolytic release of mostly PAHs (*27*). It can reasonably be proposed that PAHs were predominant precursors to this material, because the compounds are abundant and ubiquitous in both circumstellar and interstellar gas phase environments (*20*) and, we may assume, were also abundant in the solar nebula. The alternative possibility exists that aliphatic hydrocarbons in the meteorite underwent condensation and aromatization as in terrestrial kerogens. However, such thermal alteration would not be consistent with the hydrogen abundance of the material, which implies an incomplete condensation of rings, the finding in the extracts of labile molecules, as nitriles, and the presence of a substantial aliphatic phase in more petrologically processed chondrites, as CVs (*7*).

Far from disappointing, the relative simplicity of Tagish Lake organic content provides insight into an outcome of early solar system chemical evolution not seen so far. In particular, the finding of just one suite of organic compounds matching those of Murchison and of some (but not all) the carbonaceous phases and compounds seen in other chondrites demonstrates the presence of distinct organic synthetic processes in primitive meteorites. It also implies that the more complex organic matter of heterogeneous chondrites may result from multiple, separate evolutionary pathways.

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9. GC-C-IRMS: HP 6890 GC, Finnigan high-temperature conversion interphase III, and Mat Delta$^+$-XL MS. GC conditions were as for GC-MS (*8*). Oxidation was at 940°C with a ceramic oxidation reactor bearing NiO/CuO/Pt wires. Standard CO_2 (six pulses) $\delta^{13}C$: −10.07‰ (VPDB). Data were analyzed with Finnigan ISODAT software, with $\sigma < \pm 0.3‰$ for peaks >0.5 V.
10. −85‰ to −5‰, up to +5‰ for methanogenic bacteria.
11. These acids were analyzed with DB-17 column (60 m by 0.25 mm; Agilent Technologies). Other $\delta^{13}C$ values were +7.7 ± 0.3‰ for a methyl-phthalimide and +5.9 ± 0.7‰, +10.5 ± 0.8‰, and +17.5 ± 0.4‰ for three dimethyl-phthalimides.
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31. $\delta‰ = (R_{sample} - R_{standard}/R_{standard}) \times 10^3$. For carbon, $R = {}^{13}C/{}^{12}C$ and the standard is VPDB. $\delta^{13}C$(VPDB) of isopropanol: −29.3‰ (EA-IRMS). $\delta^{13}C$ mass balance equations: $\delta^{13}C_{diacid} = [\delta^{13}C_{der.\ diacid} - a(-29.3)]/b$ (*a* and *b* are fractional carbon abundances of isopropanol and diacid, respectively). A procedural succinic acid standard gave no substantial carbon fractionation.

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Origin of Whales from Early Artiodactyls: Hands and Feet of Eocene Protocetidae from Pakistan

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Partial skeletons of two new fossil whales, *Artiocetus clavis* and *Rodhocetus balochistanensis*, are among the oldest known protocetid archaeocetes. These came from early Lutetian age (47 million years ago) strata in eastern Balochistan Province, Pakistan. Both have an astragalus and cuboid in the ankle with characteristics diagnostic of artiodactyls; *R. balochistanensis* has virtually complete fore- and hind limbs. The new skeletons are important in augmenting the diversity of early Protocetidae, clarifying that Cetacea evolved from early Artiodactyla rather than Mesonychia and showing how early protocetids swam.

Whales are marine mammals grouped in the order Cetacea (*1*). Most mammals live on land and the fossil record of early mammals is terrestrial. Thus, it has long been reasonable to infer that the origin of whales involved an evolutionary transition from land to sea. This is one of the most profound changes of adaptive zone amenable to study in the fossil record. Living whales are so distinctive, and intermediate fossils sufficiently rare, that a half-century ago Simpson regarded Cetacea as "on the whole the most peculiar and aberrant of mammals," inserting them arbitrarily in his classic 1945 classification of mammals (*2*). In response, Boyden and Gemeroy (*3*) applied innovative immunological precipitin tests and found much higher cross-reactivity of Cetacea to Artiodactyla than to other living orders of mammals. Van Valen (*4*) attempted to reconcile close relationship of whales and artiodactyls with the then-known fossil record by proposing that whales originated from Paleocene mesonychid condylarths,

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