

## Mineralogy of Tagish Lake: An ungrouped type 2 carbonaceous chondrite

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Abstract—In this paper we describe the recovery, handling and preliminary mineralogical investigation of the Tagish Lake meteorite. Tagish Lake is a type 2 carbonaceous chondrite which bears similarities to CI1 and CM chondrite groups, but is distinct from both. Abundant phyllosilicates as well as chondrules (however sparse) and common olivine grains in the matrix preclude any other classification. The bulk density of Tagish Lake (1.67 g/cc) is far lower than CI or CM chondrites (2.2–2.3 and 2.6–2.9 g/cc, respectively), or any other meteorite for that matter. We have identified two lithologies: a dominant carbonate-poor lithology and a less-abundant carbonate-rich lithology. The meteorite is a breccia at all scales.

We have noted similarities between Tagish Lake and some clasts within the enigmatic meteorite Kaidun; possibly there are genetic relationships here worth exploring. In the paper we describe a clast of CM1 material within Tagish Lake which is very similar to a major lithology in Kaidun.

#### TAGISH LAKE FALL AND RECOVERY

Over the past three decades the U.S. has placed a constellation of Department of Defense (DOD) satellites (called "capabilities") in orbit whose primary duty is to watch for sinister launches of ICBMs (Tagliaferri et al., 1994). Over the past five years this coverage has become global and constant (before this sporadically-active ground-based camera networks also recorded this information). In 30 years these satellites have detected hundreds of large meteoroids entering the Earth's atmosphere. Unfortunately, although we have had the satellite detections, and earlier ground-based observations, we have never successfully recovered fireball dust in the atmosphere or meteorites on the ground after a satellite detection. In the past, claims were made that dust samples from the Revelstoke and Allende fireballs were sampled (Carr, 1970), but these claims have never been substantiated, and our efforts to uncover any of these dust samples have been completely unsuccessful.

On 2000 January 18, sensors aboard two DOD "capabilities" detected the entry of a huge meteoroid into Earth's atmosphere over western Canada. Based on the luminosity of the fireball, Brown *et al.* (2000) calculated that this was the largest object to enter our atmosphere that year, and the largest

over land in a decade. Residents of Whitehorse in Yukon Territory were treated to an intense dawn fireball, thunderous booming and sizzling sounds, a sulfurous odor, and a luminous cloud that persisted for up to an hour. A synthesis of satellite optical data, features in the infrared satellite record, and ground-based dust cloud photos suggests an initial entry mass for the meteoroid of  $2 \times 10^5$  kg. Taking the measured bulk density of the recovered samples of ~1.7 g cm<sup>-3</sup>, this corresponds to an initial body 4 to 6 m in diameter (Brown *et al.*, 2000). The huge dust cloud that had been produced suggested that this was a very friable meteoroid—the sort of material that almost never reaches the ground or survives there for long.

For this exceptional event we made every possible effort to recover material for analysis. Within 72 h we sent a NASA stratospheric ER-2 aircraft over western Canada in an effort to collect any residual dust that might remain from the fireball, although the chances of success were slim. Realizing this sad fact, through the resident Geological Survey of Canada (GSC) geologist Charley Roots, we requested that local residents collect snow samples from frozen lakes, in the faint hope that fallout from the event would be present. Dr. Roots managed the collection of snow from several local lakes, and these samples are now curated in a frozen state (in air) at the Johnson

Space Center (JSC). There is no dust in these samples visible to the naked eye.

Because of the time of year (winter) and sparsely populated nature of the target area we did not expect that any meteorites would be recovered, but we were wrong. One week after the fireball, local resident Jim Brook noticed some dark objects scattered across the frozen surface of Tagish Lake, which he realized were probably meteorites. Over the next day he collected 1 kg of samples, and he had the unique foresight to keep the samples frozen. A snowfall brought a sudden close to his collecting. Successful collection activities were not resumed until the May spring thaw. These recovery efforts are covered in much more detail in a companion paper (Hildebrand *et al.*, 2002, unpubl. data).

At the urging of Peter Brown and Charley Roots, Jim Brook sent half of his cosmic harvest to one of us (M. E. Z.), by frozen express air shipment, for initial characterization. These samples were, and are, stored in a freezer in class 10 000 air. We call these samples "pristine". They have only been removed from the freezer for brief periods to weigh and photograph them. The results of these preliminary characterizations are summarized in Table 1. All of these sampling and characterization activities were done in a specially-cleaned nitrogen cabinet in the JSC Meteorite Processing Lab, with the samples being maintained in a frozen state using dry ice. One sample appeared to have thawed while on the ice, as it displayed patches of rust on the

TABLE 1. Characteristics of the 0.5 kg of frozen Tagish Lake samples sent to JSC in January 2000.

Sample	Description	Mass
1	One large piece with fusion crust knocked off one face, exposing interior.	159 g
2	Almost totally fusion-crusted individual. Sample was thawed and sampled by R. Clayton, D. Mittlefehldt, P. Brown and M. Lipschutz.	56 g
3	Five individuals stones, one fully fusion crusted, and <1 g of fines.	22 g
4	Almost totally fusion-crusted individual; oriented stone.	60 g
5	Five individual stones, and <1 g of fines.	27 g
6	Almost totally fusion-crusted individual. Contaminated by RCMP dog nasal fluids.	34 g
7	Totally fusion-crusted individual.	45 g
8	Seven individuals stones, and <1 g of fines. Sampled by S. Pizzarrelo and I. Gilmour.	33 g
9	Three pieces of one stone.	55 g

fusion crust. We have permitted this sample to come to room temperature. During this time the sample evolved a distinct odor of sulfur, probably indicating the presence of volatile sulfur-bearing organic compounds.

The samples that are currently at JSC represent approximately half of the pristine samples; the remaining half remain in Canada with the finder. The samples at JSC remain the property of the finder as of this writing. The frozen samples that came to JSC for preliminary characterization were packaged in polyethylene bags. We repackaged them in cleaned teflon bags, and have retained the original polyethylene bags for future contamination studies.

With the spring thaw in May, Alan Hildebrand and Peter Brown led a triage meteorite recovery effort in the rapidly-melting, ice-capped Tagish Lake (Hildebrand *et al.*, 2002, unpubl. data). By the time the rapidly disintegrating lake ice had become too dangerous to cross, their brave, wet team had collected hundreds of additional samples. Many of the samples were found frozen into lake ice. Most of the samples had turned to mud upon becoming wet, and were unrecoverable. If Jim Brook had not recovered his samples so quickly, and before the first snow, they would have never survived intact.

The stones recovered in May were in a variety of preservational states; we call these samples "degraded". All are suspect of having been in contact with liquid lake water and snow melt. Indeed many of these samples were recovered in powdered form, and as small disaggregated chips. We have been gradually thawing out most of these samples. During this process many of the stones fall to pieces, although some come through this operation largely undamaged. From what we have been able to determine there is no mineralogic difference between the samples that easily disaggregate from those that remain whole.

The thaw process proceeds in the following way. We place samples in clean aluminum foil in a large walk-in freezer with a low humidity. We continually reweigh each sample as ice is slowly sublimated in the freezer. For many samples this step may take only a week or two. For samples encased in solid blocks of ice it takes many months. When the masses have stabilized we relocate the stones to cleaned, room-temperature nitrogen cabinets. In ~24 h the stones are at room temperature and are ready for final weighing, preliminary visual inspection, and storage in another nitrogen cabinet. At the time of the writing of this paper, only a small percentage of the degraded samples have been brought to room temperature. We plan to maintain some of the samples frozen, in the event that we are losing some information during the thaw process.

# SAMPLE PREPARATION AND ANALYTICAL PROCEDURES

We were faced with several unique challenges with this meteorite. Some stones were frozen and had to be maintained frozen during subsampling. All of the samples were especially

friable, and would immediately turn into mud if they came in contact with abundant fluids. We were hopeful of finding fluid inclusions, or water-soluble sulfates or halides, in the samples and our experience with those necessitated the use of a special epoxy (Struers Epofix) and isopropyl alcohol as the only fluid. The epoxy cures at room temperature, is easily removable, and does not fluoresce in the Raman microprobe beam. All cutting and polishing operations were done slowly by hand (to prevent heating) using isopropyl alcohol. Samples had to be repeatedly re-impregnated with epoxy after every cut or round of polishing. We have noticed that our initial sections display a great deal of large pores and cavities which we now believe to have resulted from immature skill level in section preparation. Our later sections, made in the most careful manner and after we had gained some experience, have fewer large cavities. We believe that these later sections are representative of the actual physical character of the meteorite.

The majority of the large constituent mineral grains in the Tagish Lake chondrite were analyzed for major element composition using a Cameca Camebax microprobe operated at 15 kV and 20 nA. Carbonates were analyzed at 15 kV and 4 nA. A focused beam ( $\sim 2 \mu m$ ) was used for mineral analyses. Natural mineral standards were utilized, and corrections were applied for absorption, fluorescence, and atomic number effects using the Cameca on-line PAP program. The precision of the microprobe analyses is better than 1% relative for major elements. Element maps were also made using this instrument. Secondary electron and backscattered electron images were made using a JEOL JSM-6340F field emission (FEG) scanning electron microscope (SEM), operating at 15 kV, which we found to offer optimum values of resolution vs. electron penetration (and excitation) of the samples, and a Hitachi S 4500 FEG-SEM.

Due to the fine-grained nature of the material, we also prepared carefully selected Tagish Lake samples for more detailed characterization by transmission electron microscope (TEM) in three different ways. We made most observations using ultramicrotomed sections of grains embedded in EMBED-812 low-viscosity epoxy. In order to better examine some mineral species, especially phyllosilicates, we also employed two other mounting techniques for TEM analysis. First, a sample suspended in ethanol was set onto a Cu grid with a collodion film coated with carbon, and dried in air. In the final method a fragment of Tagish Lake embedded in EPON812 epoxy was vacuum impregnated at 60 °C for 1 h, and then polymerized at 60 °C for 48 h. After sectioning by ultramicrotomy, thin films were collected on Cu grid coated with collodion and carbon. We observed the samples using a JEOL 2000FX scanning transmission electron microscope (STEM) equipped with a LINK energy dispersive x-ray spectrometer (EDX) analysis system at JSC, and a JEOL 2010 with a Kevex Sigma system EDX at the University of Tokyo, both operating at 200 kV. We used natural mineral standards and in-house determined k-factors for reduction of compositional data; a Cliff-Lorimer thin-film correction procedure was employed (Goldstein, 1979). The TEM-EDX analyses are precise to within 1–3% relative. Additional TEM investigation of carbon was carried out at Kobe University using a JEOL 2010 high-resolution (HR)TEM, operating at 200 kV equipped both an EDX and an electron energy loss (EELS) spectroscopies. A Noran Voyager EDX analysis system, equipped with a Noran ultrathin window, was used to obtain *in situ* mineral analyses with the Cliff-Lorimer thin film approximation for data reduction.

#### **DENSITY AND POROSITY**

Because of the extremely short survival period of Tagish Lake samples on the ground, the obvious low density (readily apparent to anyone holding a thawed sample), and high porosity visible in the sections, we resolved to measure the bulk density of the meteorite. The problem we faced was that the stones become mush upon wetting, sometimes even by alcohol. Therefore we resorted to density determination by a less-accurate, dry technique.

We obtained a quantity of sieved  $(500-700 \,\mu\text{m})$ , clean, dry, purified quartz sand. We then weighed a known volume of this sand, submerged a Tagish Lake stone in the sand and reweighed the same total volume, and made the appropriate calculation to determine the weight and volume of the stone. We performed this same operation on a similarly-sized solid mass of quartz as a measure of the measurement error. Our measured density for Tagish Lake is  $1.66 \pm 0.08$  g/cc.

The density of Tagish Lake is significantly lower than that for any meteorite we have previously analyzed (or found in the literature), and most chondrtic interplanetary dust particles (Corrigan *et al.*, 1997). There is a recent measurement of Orgueil reporting a similarly low density (Consolmagno and Britt, 1998). However, Gounelle and Zolensky (2001) show that this Orgueil measurement is much too low.

#### **PETROGRAPHY**

The recovered stones are quite black, with small visible chondrules, pores and olivine crystals present in varying abundance (Figs. 1 and 2). We have now prepared and examined 17 thin sections cut from nine different stones, of both pristine and degraded samples. We believe this sampling gives us an almost unprecedented view of such a recent fall. The meteorite is a breccia at all scales. We have noted two major lithologies, carbonate-poor and carbonate-rich. We have also characterized the largest "foreign" clast, which resembles CM1 chondrites. Smaller clasts of other materials remain for future characterization. Additional major lithologies may be present among the uninvestigated stones (of which there are many), but probably not in high relative abundance. Although the carbonate-rich and -poor lithologies are generally distinct, and the meteotite is a breccia, one lithology can occasionally

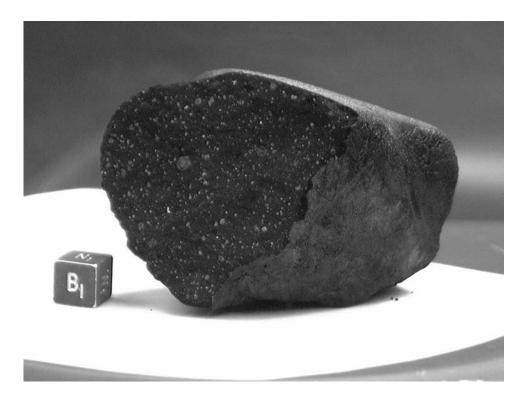


FIG. 1. The largest pristine Tagish Lake stone. The scale cube measures 1 cm on a side.

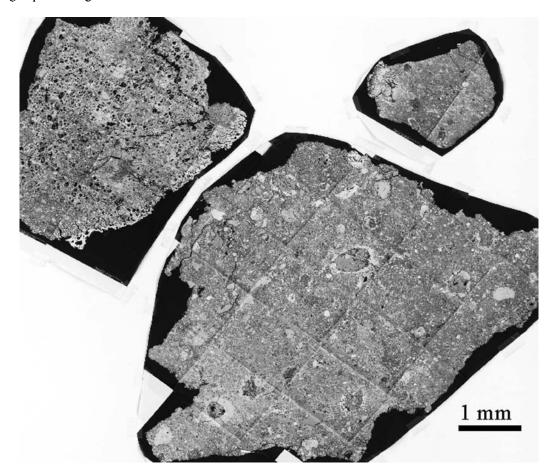


Fig. 2. Backscattered electron image of a polished thin section of a degraded sample.

be observed grading smoothly into the other, indicating that they are genetically related. We estimate that the carbonate-poor lithology is slightly more abundant than carbonate-rich one, approximately by a ratio of 3:2. This estimate is very tentative, due to the somewhat gradational differences between these materials.

The CM1 clast we describe below is quite distinct from the main lithologies of Tagish Lake. This lithology has been found so far in only one stone and so is probably rather rare.

The major constituent of all lithologies is fine-grained, opaque matrix consisting mainly of phyllosilicates, sulfides and magnetite (especially the latter mineral). Set within this matrix are rather sparse chondrules, aggregates of olivine, pyroxene and/or phyllosilicates, fine-grained aggregates, magnetite, carbonates, Fe-Ni sulfides, and rare calciumaluminum-rich inclusions (CAIs). Many of these larger components are surrounded by rounded, fine-grained, rims. Many of the rims contain high concentrations of very fine-grained Fe-Ni sulfides.

All samples are very porous, with pores commonly ranging up to  $50 \, \mu \text{m}$  in size, and a few as large as  $100 \, \mu \text{m}$ . These pores

are typically very irregular in shape, and commonly coated with carbonates. The pores are common to both of the major lithologies, but are somewhat more abundant in the carbonaterich one. The pore-filling carbonates are apparent as whitish, dusty crusts on broken surfaces of the meteorite, and can exceed 1 mm in length. We have not attempted a porosity determination, because of the extremely friable nature of the meteorite, and the uncertain political environment surrounding the larger samples.

We will describe the three lithologies separately. However to avoid repetition we describe the features common to the entire meteorite immediately below in the description of the first lithology.

#### The Carbonate-Poor Lithology

This dominant lithology is a phyllosilicate-rich, matrix-supported assemblage of fine- to coarse-grained, phyllosilicate-rich clasts (Fig. 3), sparse chondrules (Figs. 3 and 4), aggregates, sparse CAIs (Fig. 5) and a variety of loose grains of olivine (Fig. 6), magnetite, Fe-Ni sulfides, Cr-Ni phosphides and Ca-Mg-Fe carbonates (uncommon). The matrix consists

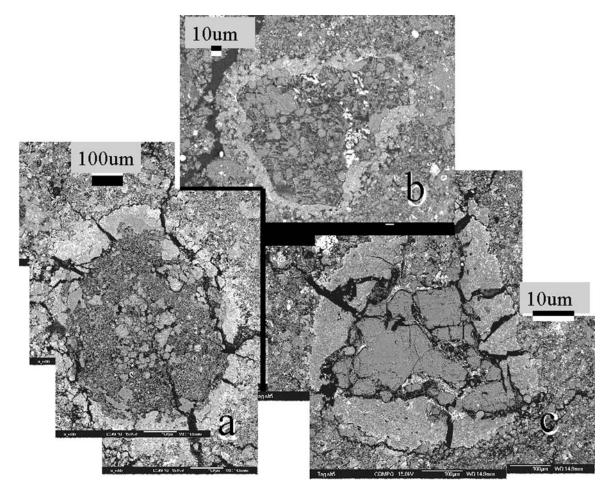


FIG. 3. Backscattered electron images from the carbonate-poor lithology of (a) an altered porphyritic chondrule, showing coarse-grained phyllosilicates (light grey) replacing olivine and pyroxene crystals (dark grey); (b) and (c) altered coarse-grained aggregates. Note the fine-grained rims which grade into matrix.

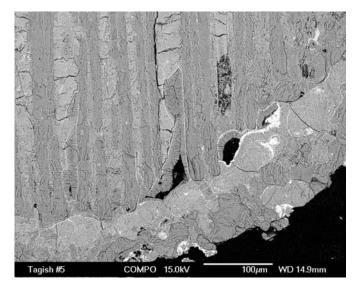


FIG. 4. Backscattered electron image of a quadrant of a large barred olivine chondrule from the carbonate-poor lithology, with serpentine replacing mesostasis. A fine-grained rim is apparent.

mainly of saponite, serpentine and Fe-Ni sulfides. Matrix-supported clasts consist of fine-grained phyllosilicates with minor sulfides and/or magnetite. Most clasts range from 50 to  $500 \,\mu\text{m}$  in average diameter, and are rounded. Pores are very common; many of these have walls covered by Ca-carbonates.

Chondrules, Aggregates and Calcium-Aluminum-Rich **Inclusions**—The majority of rounded, chondrule-like objects in the carbonate-poor lithology are partly to heavily aqueouslyaltered to coarse- and/or fine-grained phyllosilicates, but a few olivine (Fo<sub>99</sub> mode) or pyroxene (En<sub>99</sub> mode) grains remain to betray their origin as chondrules (Figs. 3 and 4). The textures that we see suggest that original chondrules were predominantly the olivine aggregate type, with a few porphyritic and rare barred oliving types thrown in the mix. In fact we found only one very large (1 mm in diameter) barred olivine chondrule, wherein the glass has been completely replaced by Mg-rich serpentine (mg# = Mg/(Mg + Fe) = 0.99), and the olivine is beginning to alter to saponite (mg# = 0.82). The majority of the chondrules have the apparent size range 0.25-1 mm, which is about the same size as those found in CM chondrules, but unlike those found in CR2 chondrites. Magnetite and/or sulfides are common both inside and rimming chondrules. All of these chondrules have rounded, rather thick, fine-grained,

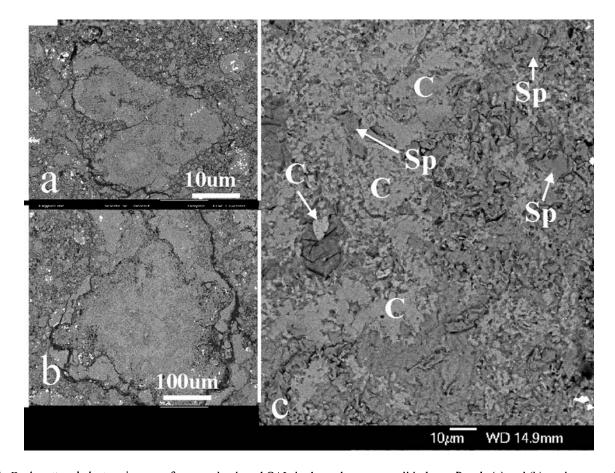


FIG. 5. Backscattered electron images of aqueously altered CAIs in the carbonate-poor lithology. Panels (a) and (b) are low-magnification views. (c) Close-up of the interior of the altered CAI in (b), showing the mineralogy typical of all. The dark phase throughout is phyllosilicate. The lighter grains are a Ca carbonate (C). White grains to the right are magnetite. There are also a few spinel grains (Sp).

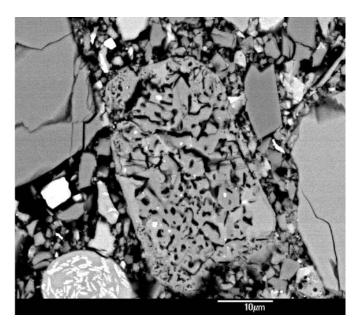


FIG. 6. Matrix olivine grain hideously disfigured by arrested dissolution. Scale bar measures  $10 \,\mu\text{m}$ . This grain is in the carbonate-poor lithology, but is typical of much olivine in Tagish Lake.

phyllosilicate-rich rims up to  $200\,\mu\mathrm{m}$  thick, which is about as thick as these ever get to be in carbonaceous chondrites. There are rounded aggregates of this rim material only, which may actually be larger rimmed chondrules sectioned through the rims. The most notable thing about Tagish Lake chondrules is their almost complete absence from both carbonate-poor and carbonate-rich lithologies. We have observed chondrule

frequencies on the order of ~1 cm<sup>-2</sup>; there are entire thin sections without any at all. Despite their rarity, the distribution of chondrule types in Tagish Lake is still approximately what was observed by Gooding (1983) for ordinary chondrites.

Aggregates in the carbonate-poor lithiology have a mineralogy similar to that of porphyritic chondrules, and most contain at least some olivine with or without pyroxene. We distinguish these aggregates from chondules because they do not have a circular shape. Aggregates outnumber chondrules by a factor of 5–10. The majority of the aggregates have apparent diameters ranging from 100 to 500  $\mu$ m; a few are up to 1 mm in diameter. Coarse-grained phyllosilicates are typically replacing the anhydrous silicates; in some instances very few of the latter remain. A typical type is aggregate #6, which is large (500  $\mu$ m), ovoid, and consists of an assemblage of saponite, serpentine, chlorite, olivine and iron oxyhydroxides enriched in phosphorus. Clinochlore appears to replace some of the serpentine and saponite.

Several altered CAIs (200–300 µm in size) were found with sinuous textures and fine-grained phyllosilicate-rich rims (Fig. 5). So far no two of these are exactly alike. A typical altered CAI has a phyllosilicate-rich rim with variable Al<sub>2</sub>O<sub>3</sub> content (see the phyllosilicate analyses in Table 2), speckled with magnetite, and poorly-characterized iron oxyhydroxides enriched in P, pyroxene (En<sub>94</sub>Wo<sub>4</sub>), olivine (Fo<sub>99</sub>), and siderite grains. The interior has the same mineralogy except that the iron oxyhydroxides are absent and porosity is far higher. Titanium-rich grains are present in the interior, which may have been fassaite, but analytical totals are now only 90%. Some altered CAIs consist largely of carbonates (dolomite and minor

TABLE 2. Microprobe analyses of minerals in Tagish Lake (wt%).

	Mn-olivine*	Phyllosilicates		Spinel	Ca-carbonate Dolomite		Andradite
Na <sub>2</sub> O	0.00	0.31	0.31	0.00	0.05	0.07	0.00
MgO	55.42	25.61	24.07	27.65	0.02	20.88	2.97
$Al_2O_3$	0.05	11.54	4.75	72.22	0.02	0.06	1.69
$SiO_2$	42.16	37.83	40.46	0.08	0.12	0.08	33.82
$P_2O_5$	nd	0.11	0.16	0.00	0.57	0.36	0.03
S	nd	0.12	0.13	0.00	0.01	0.01	0.22
K <sub>2</sub> O	0.01	0.09	0.10	0.01	0.00	0.01	0.01
CaO	0.17	0.07	0.14	0.07	64.19	31.60	29.93
TiO <sub>2</sub>	0.04	0.14	0.15	0.27	0.01	0.01	0.29
$V_2O_3$	0.01	nd	nd	nd	nd	nd	nd
$Cr_2O_3$	0.89	0.27	0.36	0.15	0.00	0.02	0.65
MnO	1.77	0.07	0.06	0.02	0.02	5.27	0.08
FeO	1.51	9.75	10.47	0.30	0.51	6.43	23.51
CoO	0.00	nd	nd	nd	nd	nd	nd
NiO	0.04	0.38	0.73	0.02	0.03	0.00	0.29
Total	102.06	86.29	81.89	100.80	65.55	64.67	93.74

<sup>\*</sup>Typical analysis. nd = not detected.

calcium carbonate), spinel and phyllosilicates (saponite and serpentine) that are homogeneous in composition (Table 2). Rare perovskite grains are enclosed within dolomite, and it is this mineral we have principally relied on to identify these objects as altered CAIs, since perovskite is generally confined to CAIs and weathers so slowly. Two of these CAIs are well analyzed. CAI1 is quite large (300  $\mu$ m) and irregular is shape, whereas CAI2 is smaller, and round. Both have fine-grained phyllosilicate-rich rims. CAI1 consists of a well-defined sequence of minerals: dolomite-enclosing perovskite is surrounded by spinel and phyllosilicates with a homogeneous composition (mg# = 0.81). In CAI2 the same minerals are present, plus minor calcium carbonate and saponite. For these CAIs there is no difference between dolomite composition and the general dolomite composition. The calcium carbonate grains are too small to obtain reasonable probe analyses. In spinel grains, the sum of tetravalent and divalent cations correlates well with the sum of trivalent cations, indicating substitutions similar to that observed in Mighei CAIs (MacPherson and Davis, 1994). Phyllosilicates in CAIs are generally a mixture of Mg-rich serpentine and saponite rather than the usual Fe-rich serpentine observed in CM2 chondrites (MacPherson and Davis, 1994; Lee and Greenwood, 1994; Greenwood et al., 1994; Zolensky et al., 1993, 1997). Some clinochlore is also present. These phyllosilicates are compositionally indistinguishable from those in the matrix of Tagish Lake. The saponite in these CAIs is similar to that in Kaidun (about the only place where high-quality saponite compositions have been obtained), although relatively depleted in Ca. We have not seen any unaltered CAIs in Tagish Lake sections. Recently, Simon and Grossman (2001a) have reported on hibonite recovered from powdered Tagish Lake samples. We have not yet observed this mineral in thin section, although we did experience a false alarm concerning blue silicon carbide (SiC) grains "contributed" to a thin section during its preparation.

Olivine and Pyroxene-Olivine grains are common within chondrules of the carbonate-poor lithology, aggregates and isolated in the matrix (Fig. 6). Olivine has the compositional range  $Fo_{71-100}$  (percent mean deviation (PMD) = 2%) with a peak at Fogg (Fig. 7). Iron-rich olivines are commoner in matrix than in chondrules or aggregates, which is similar to the occurrence in CM2 chondrites (McSween, 1977). Some of the olivines contain beads of Fe-Ni metal. Most of the olivines are deeply embayed, evidence of arrested dissolution (Fig. 6) (Nahon, 1991) and replacement by phyllosilicates (mg#=0.80). Pyroxene is rare, occurring mainly as isolated grains in matrix and within a few aggregates; it is generally iron- and calciumpoor  $(En_{93-99} (PMD = 2\%)$ , with a peak at  $En_{98}$ ). We have observed only one high-Ca pyroxene, a diopside grain sitting in matrix. Mn-rich olivines are very rare; but we have analyzed four (Table 2), and observed a maximum MnO content of 1.77 wt%. We have not so far found any refractory olivines (Fo > 99.5), although some have been found by Simon and Grossman (2001b).

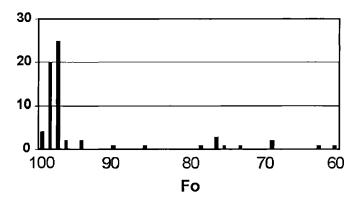


FIG. 7. Compositional range displayed by Tagish Lake olivines, for both lithologies (which have the same compositional distribution).

The meteorite shock stage for the entire meteorite, as determined from olivine and pyroxene textures, is S1, which is typical for carbonaceous chondrites (Scott *et al.*, 1992). Of course, it is always possible that deformed olivines which might have once recorded a higher shock stage have been preferentially replaced by phyllosilicates. This problem is universal with type 1 and 2 carbonaceous chondrites.

Secondary and Accessory Minerals–Phyllosilicates in the carbonate-poor lithology are mostly confined to the fine-grained matrix but are also found as coarser grains associated with olivine, magnetite or carbonate. They mainly consist of magnesium-rich serpentine and saponite, as identified by both compositions and TEM imaging (Table 3). Clinochlore is also present, and is recognized most easily by its high  $Al_2O_3$  content (Table 3). This mineral is frequently intergrown with serpentine.

Magnetite is very abundant in the matrix but also in clasts, in association with olivine, carbonates and phyllosilicates, and is commonly intergrown with sulfides. Framboids and placquettes are the dominant magnetite lithologies, but euhedral single grains are also present (Fig. 8). Unfortunately, it has often been impossible to determine the order of precipitation (see Fig. 9g). However, in many places in the matrix, magnetite forms a thin rim around sulfides, indicative of a late-stage oxidation event. Also, we have seen perfect pseudomorphs of framboidal and placquette magnetite aggregates after pyrrhotite crystals (Fig. 8a), clearly indicating that magnetite is replacing the sulfide. This oxidation event/replacement has apparently not affected the aggregates and chondrules to the same degree, possibly because of lower permeabilities in these objects. We have noted iron oxyhydroxides having a composition similar to that of the so-called "COPS" phase (carbon, oxygen, phosporus, sulfur) (Engrand, 1993) within and in close proximity to aggregates, in a manner similar to that observed in CR2-type microclasts in howardites (Gounelle et al., 2002, unpubl. data). The COPS phase was first described from Antarctic micrometeorites (Engrand, 1993). However, we have recently determined that this material is not a single, crystalline phase (Zolensky, Gounelle and Ohsumi, unpubl. data), so we have not investigated this confusing phase further.

TABLE 3. Microprobe analyses of phyllosilicates in Tagish Lake (wt%).\*

	Tagish Lake lithologies					CM clast		
Analyses	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
Na <sub>2</sub> O	0.18	0.05	0.39	0.13	0.26	0.12	0.02	0.05
MgO	22.96	32.18	15.73	28.26	16.97	26.33	34.00	30.74
$Al_2O_3$	11.97	1.07	13.66	3.56	19.24	3.27	1.26	1.70
$SiO_2$	34.02	42.33	30.62	39.35	26.42	38.66	41.39	40.50
$P_2O_5$	0.08	0.04	0.00	0.04	1.11	0.03	0.00	0.02
S	0.06	0.06	0.06	0.09	0.32	0.18	0.05	0.07
K <sub>2</sub> O	0.05	0.02	0.06	0.05	0.11	0.10	0.00	0.03
CaO	0.65	0.09	0.60	0.14	0.10	0.12	0.07	0.09
$TiO_2$	0.10	0.08	0.09	0.09	0.09	0.09	0.03	0.07
$Cr_2O_3$	0.33	0.71	0.31	0.56	0.08	0.62	0.52	0.52
MnO	0.38	0.15	0.35	0.20	0.23	0.16	0.10	0.14
FeO	16.61	10.72	27.14	14.34	20.47	12.69	11.52	11.63
NiO	0.05	0.05	0.05	0.15	0.03	0.17	0.04	0.06
Total	87.43	87.57	89.06	86.96	85.45	82.54	88.99 <sup>,</sup>	85.62

<sup>\*</sup>Only analyses with S < 0.05 wt%,  $P_2O_5 < 1$  wt%, and  $Cr_2O_3 < 1$  wt% are considered. Analyses: (1) typical analysis; (2) lowest Mg content; (3) highest Mg content; (4) average of 48 analyses; (5) typical clinochlore; (6) lowest Mg content; (7) highest Mg content; (8) average of 50 analyses.

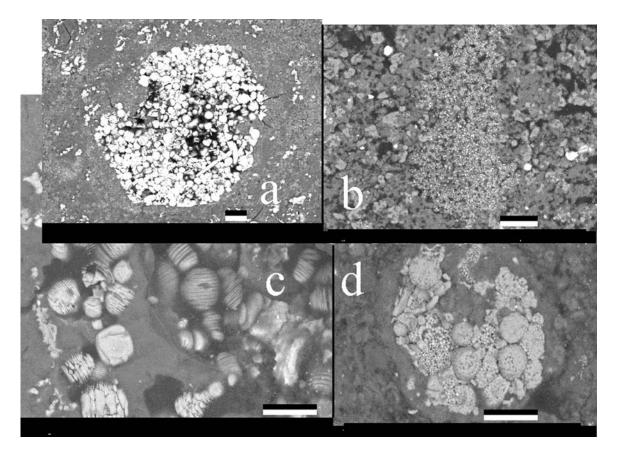


FIG. 8. Common morphologies of magnetite in Tagish Lake (both major lithologies). (a) Magnetite framboids and placquetes pseudomorph after a pyrrhotite crystal, (b) patch of matrix incredibly rich in very fine-grained magnetite, (c) placquetes, and (d) framboids. Scale bars measure  $10 \, \mu m$ .

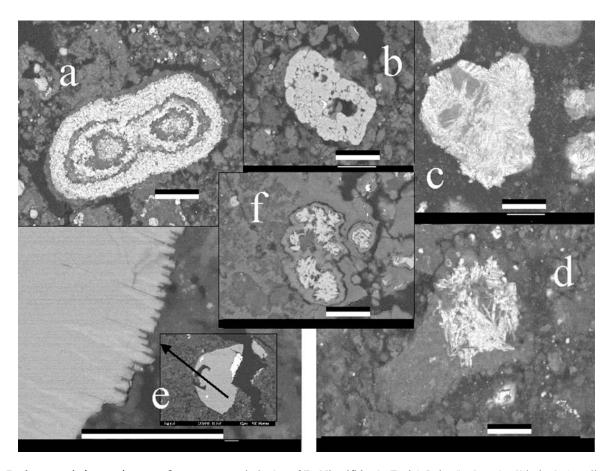


FIG. 9. Backscattered electron images of common morphologies of Fe-Ni sulfides in Tagish Lake (both major lithologies). All scale bars measure  $10 \mu m$ . (a) Pyrrhotite aggregate indicating multiple precipitation episodes. (b) Typical aggregate of intermediate composition sulfide. (c) and (d) Aggregates of acicular pyrrhotite surrounded by phyllosilicate. (e) Large white grain from the carbonate-rich lithology is a mixture of pyrrhotite (dark) and sulfide of intermediate composition (light). The small white grain is a sulfide. The pale grains are Fe-Mg carbonates. The rest is matrix, a mixture of carbonates and phyllosilicates. (f) Acicular pyrrhotite in a zoned phyllosilicate aggregate, possibly a pseudomorph after a CAI.

The sulfides present in all of Tagish Lake are pyrrhotite, pentlandite, and the oft-encountered phase intermediate in composition between them (Zolensky and Di Valentin, 1998). This latter phase has never been satisfactorily characterized, yet it appears to be real. Sulfides are typically very fine-grained (submicron) and display a wide variety of morphologies including rounded, elongated, irregular, and acicular. This acicular morphology for pyrrhotite is rather unusual, and is otherwise only known from a CM1 lithology in the enigmatic chondrite Kaidun (Zolensky et al., 1996a). A notable aspect of this lithology is the rarity of coarse-grained sulfides in the matrix—those observable at petrographic scales. The bulk of the opaques visible at this scale are magnetite. For the most part the coarse-grained sulfides are confined to aggregates and chondrules.

Despite their paucity at large scales, SEM and TEM observations show that sulfides are very abundant as a fine-grained component of the matrix (Fig. 9). These sulfides are present as both well-developed prismatic crystals (up to 200 nm long) and smaller rounded grains scattered within

phyllosilicates. The selected area electron diffraction (SAED) pattern of the Ni-poor Fe-sulfide suggests that it is a hexagonal pyrrhotite (a = 3.45 Å, c = 5.8 Å), although satellite reflections betray the presence of a superstructure cell. This phase is consistent with the microprobe analysis (Mikouchi *et al.*, 2001). Occasional grains of pentlandite are also present.

Some of the matrix sulfides in the carbonate-poor lithology have thin rims (Fig. 10). Dark-field TEM imaging in conjunction with TEM grid tilting reveals that these rims consist of carbon, and have layer lattice spacings of 3.5–3.8 Å. These data suggest that the rims are poorly-graphitized carbon (PGC), which is formed by the thermal processing of graphitizable carbon compounds including complex macromolecular hydrocarbons such as kerogen (Fischbach, 1971; Brearley, 1990). Graphitization is a thermally-activated, irreversible process that is sluggish and involves the progressive loss of H, N and O from the organic material as temperature is increased. The basal lattice spacing of the Tagish Lake PGC is in the range 3.5–3.8 Å, indicating that this lithlogy probably never witnessed metamorphic temperatures higher than 200–375 °C

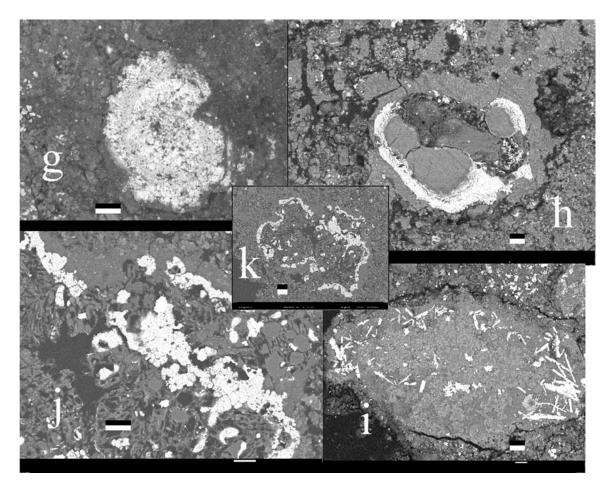


FIG. 9. Continued. Backscattered electron images of common morphologies of Fe-Ni sulfides in Tagish Lake (both major lithologies). All scale bars measure  $10 \mu m$ . (g) Complexly intergrown aggregate of very fine-grained magnetite (grey) and pyrrhotite (white). (h) to (k) Phyllosilicate aggregates with pyrrhotite-rich rims. (j) is a closeup of (k).

(Rietmeijer and Mackinnon, 1985). This temperature is consistent with the presence of fully hydrated serpentine (Akai, 1988, 1990a,b), which begins to dehydrate at ~400 °C.

Another notable feature of both the carbonate-poor and carbonate-rich lithologies is the clear petrographic evidence for multiple episodes of sulfide precipitation. Phyllosilicate aggregates often show sulfide enrichments towards grain boundaries, or sport actual sulfide rims, indicating that sulfidation episodes followed the bulk of the aqueous alteration reactions. Matrix grains and small phyllosilicate aggregates display up to three distinct sulfide layers (Fig. 9a), alternating with phyllosilicates, suggesting that the sulfur content and other critical aspects of the aqueous fluids responsible for alteration fluctuated through time.

In many places we can actually observe the incomplete transformation of pyrrhotite into the intermediate sulfide phase (Figs. 9 and 11). In Fig. 12 we show the sulfide compositions from typical CM2 chondrites spanning the range of aqueous alteration. The intermediate sulfide compositions observed in Tagish Lake are typical of the most heavily aqueously-altered CM2 chondrites, such as Allan Hills (ALH) 83100, while pure

pyrrhotite is most abundant in the least altered CM2 chondrites (Fig. 12). The presence of both phases in Tagish Lake implies an intermediate degree of aqueous alteration.

A Cr-Ni phosphide has been found in the carbonate-poor lithology, in association with schreibersite, as scattered submicrometer-thick plates within a coarse-grained phyllosilicates clast (Fig. 13). This phase and occurrence is reminiscent of similar phosphides associated with florenskyite (FeTiP), found only in a Kaidun C1 clast (Ivanov *et al.*, 2000). Because of the extremely small grain size, we have not determined if these occurrences are identical. This is clearly a new mineral, but it may be too fine-grained and rare to characterize further at this time.

Calcium carbonate is by far the most abundant carbonate (Table 4) in the carbonate-poor lithology, but compared with the carbonate-rich lithology it is relatively uncommon, occurring as sparse very fine polycrystalline grains ( $<5~\mu m$ ) scattered throughout the matrix, lining pores, and forming associations with phyllosilicates and magnetite. It contains no detectable magnesium or manganese, though the iron content can be as high as 1.74 wt% FeO. Dolomite is the second most

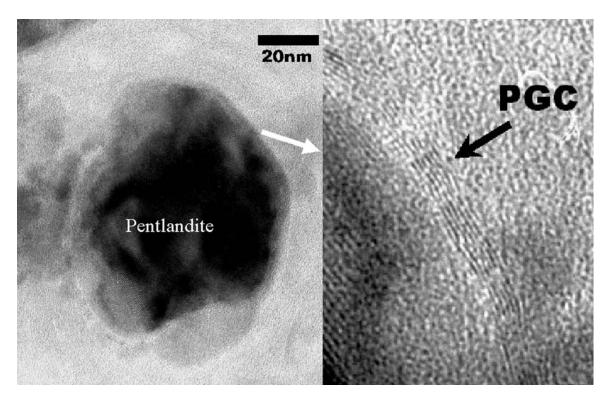


FIG. 10. (a) Transmission electron microscope (TEM) image of a matrix pentlandite particle rimmed with poorly-graphitized carbon (PGC), from the carbonate-poor lithology. (b) High-resolution TEM image from the rim of (a) showing characteristic PGC (002) interlayers spaced at 3.5–3.8 Å.

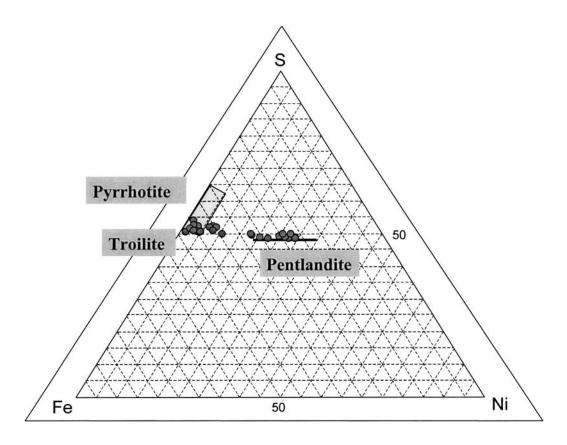


FIG. 11. Compositions of Tagish Lake Fe-Ni sulfides (both major lithologies) displayed in a Fe-Ni-S atom% ternary diagram.

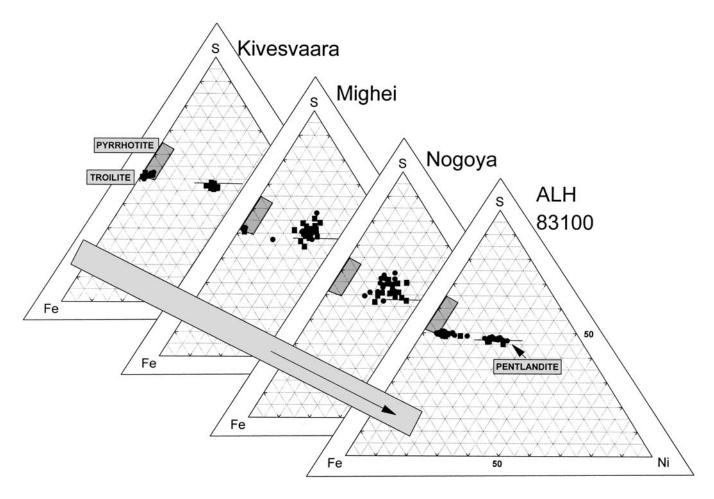


FIG. 12. Sulfide compositions from CM chondrites displayed as in Fig. 11. The chondrites are arranged from least aqueously-altered CM2 (Kivesvaara) to most-altered CM2 (ALH 83100). In general, the amount of intermediate sulfides (lying between pyrrhotite and pentlandite) is directly related to the relative degree of aqueous alteration, and the amount of stoichiometric troilite varies indirectly. Tagish Lake fits into this trend.

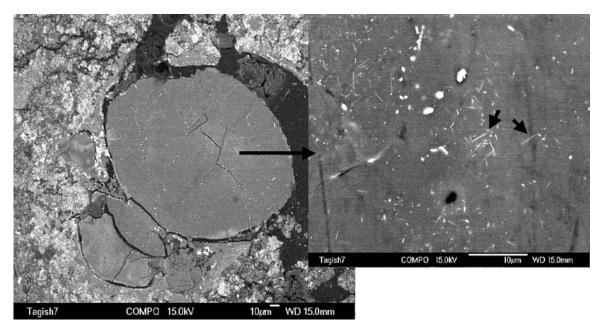


FIG. 13. (a) Backscattered electron image of a coarse phyllosilicate grain containing submicrometer Cr-Ni phosphides (bright dots), from the carbonate-poor lithology. (b) Closeup of the phosphide laths (white). Scale bars measure  $10 \, \mu \text{m}$ .

TABLE 4. Microprobe analyses of carbonates in Tagish Lake (wt%).

Analyses	(1)	(2)	(3)	
MgO	0.00	9.91	14.21	
$SiO_2$	0.03	0.09	0.02	
CaO	54.37	1.80	4.12	
MnO	0.00	0.00	2.91	
FeO	0.32	44.87	31.71	
Total	54.72	56.67	52.97	

Analyses: (1) Ca carbonate from carbonate-poor lithology; (2) and (3) siderites from carbonate-rich lithology.

important carbonate; it is found in some of the CAIs, and rarely in matrix. It is slightly coarser grained (<10  $\mu$ m) and usually embayed with phyllosilicates. It can contain up to 7.79 wt% FeO and up to 5.27 wt% MnO. Siderite is present in one ironrich aggregate, and some CAIs. The CaO content of this is up to 3.46 wt% and the MnO content can reach 0.60 wt%. In many places carbonates are lining walls of large pores, obviously a late-stage precipitate.

We found no sulfates in any Tagish Lake lithology, despite the fact that we took great care in thin section preparation to preserve water-soluble phases. It will be interesting to reobserve these same samples in a few years to determine whether terrestrial sulfates appear, as they have for most CI falls (Gounelle and Zolensky, 2001).

TEM observation of microtomed sections reveals that the matrix of the carbonate-poor lithology consists mainly of phyllosilicates, with an entangled ribbon-like structure (Fig. 14).

This material consists mainly of two phases, which we identified on the basis of layer lattice fringes in HRTEM images and by EDS composition. A phase with 10–12 Å layer lattice fringes, which we took to be saponite (Fig. 14a), and serpentine, identified by 7 Å layer lattice fringes. Serpentine also occurs as parallel fine-grained crystallites of several tens of layers thick. A few povlen and cylindrical forms characteristic of chrysotile were also observed (Fig. 14b). A few examples of more iron-rich serpentine (Table 3) were also noted. We also observed areas of coherent- to disordered intergrowths of serpentine with saponite, and intergrowths of serpentine with a chlorite-type phase (probably clinochlore) with a basal spacing of ~14 Å (Fig. 15). In most matrix areas saponite is definitely present, identified by basal spacings together with EDX composition, and is more abundant than serpentine. Saponite is present in clumps and flakes of fine crystallites only a few sheets thick, and also as coarser grained sheaths. Also present in some coarse sheaths are the intergrowths with serpentine. Low-magnification images of some areas show a weak foliation to the phyllosilicates; they are complexly piled on one another. These textures are typical of CI1 chondrites, but completely unlike CM chondrites (Zolensky et al., 1993; Brearley, 1997).

EDS analysis of the matrix phyllosilicate gives compositions intermediate between serpentine and saponite solid solutions, that generally match with the microprobe analysis of the most Fe-poor phyllosilicates (Fig. 16). The atomic Fe/(Fe + Mg) ratio (fe#) is ~0.3, which is more comparable to Ivuna rather than Orgueil (0.15) (Brearley, 1997).

An unusual feature of both the carbonate-poor and -rich lithologies is the presence of an unprecedented amount of irregularly-shaped open spaces. These could be called pores

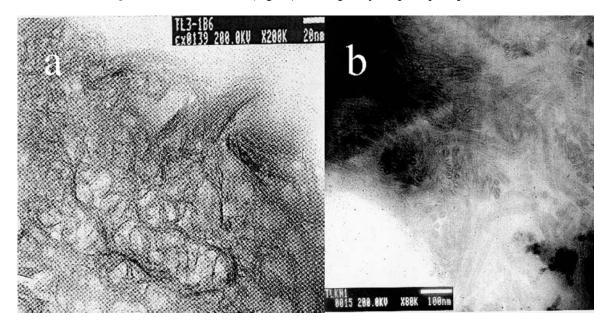


FIG. 14. (a) Transmission electron microscope (TEM) image of saponite with an entangled, ribbon-like structure from the matrix of the carbonate-poor lithology. Scale bar measures 20 nm. (b) TEM image of serpentine with platy to cylindrical structures from the matrix of the carbonate-poor lithology. Scale bar measures 100 nm.

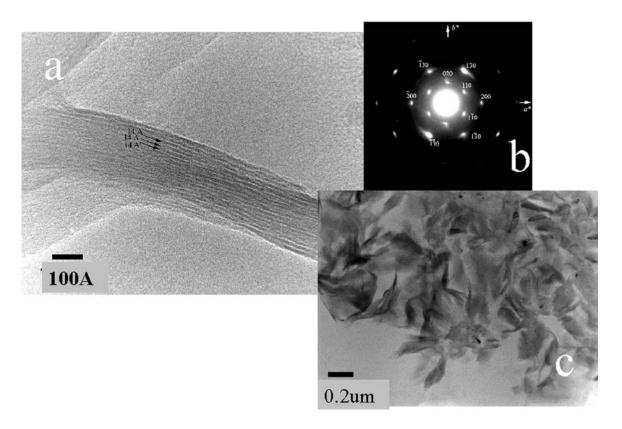


FIG. 15. Coherently interlayered saponite, clinochlore and serpentine flakes from the matrix of the carbonate-poor lithology. (a) High-resolution TEM image showing the 14 Å interlayer spacings characteristic of the intergrowth of serpentine and clinochlore; scale bar measures 100 Å. (b) Electron diffraction pattern from (a). (c) Low-magnification TEM image of matrix phyllosilicate flakes, including (a); scale bar measures  $0.2 \, \mu \text{m}$ .

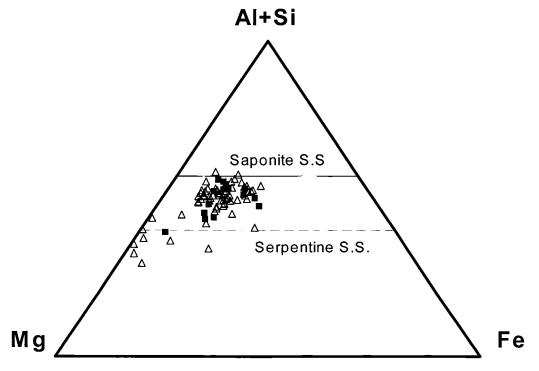


FIG. 16. Composition of phyllosilicates in Tagish Lake lithologies showing an intermediate composition between serpentine and saponite solid solutions (WDS data). Triangles represent the carbonate-poor lithology, squares are from the carbonate-rich lithology.

or cavities, but are clearly not vesicles since they have granular, irregular walls and the meteorite shows no evidence of having once been molten. The huge number of these pores undoubtedly contributes to the extreme friability of the bulk meteorite, dooming it to a short survival time on the ground. These pores also explain the uniquely low-density of this meteorite.

Carbon-Rich Phases—One sample of the carbonate-poor lithology (and so far one sample only) contains numerous hollow, bubble-like hydrocarbon globules set within the phyllosilicate matrix (Nakamura *et al.*, 2001). We have also found within matrix one irregularly-shaped graphite grain with fairly sharp SAED reflections (Fig. 17). It is not associated with any other phases, but is present as one individual grain. This may be a presolar graphite grain, although cauliflower or onion-skin texture typical for presolar graphites (Zinner *et al.*, 1995) is not obvious here.

#### The Carbonate-Rich Lithology

This lithology is similar to the carbonate-poor lithology in most respects, and actually grades into it. However, there are significant differences which must be due to significantly different conditions of alteration. We will discuss only these differences here. Although the abundance of chondrules is about the same in the carbonate-rich and -poor lithologies, fine-grained clasts and CAIs are almost absent from the former—we have only observed one very altered CAI in a single section. In general, this lithology is notably finer grained than any other common lithology in carbonaceous chondrites, excepting the finest grained CV3 dark inclusions (Krot *et al.*, 1997).

Phyllosilicates in the carbonate-rich lithology consist almost entirely of saponite, and are almost universally very fine-grained. In fact only one in three of the TEM grids we prepared of this material displayed any serpentine at all. Although the compositions of these appeared to be essentially the same as those observed in the carbonate-poor lithology, the finer grained nature of phyllosilicates in the carbonate-rich lithology makes this conclusion rather uncertain. Andradite is observed to be present in association with phyllosilicates and magnetite in one aggregate of uncertain origin (Fig. 18).

Magnetite is rare in the carbonate-rich lithology. This is a major difference from the carbonate-poor lithology, where magnetite is perhaps more common than in any other meteorite we have ever observed, including Bells (Zolensky *et al.*, 1993).

The micrometer-sized and smaller-sized sulfides so common in the carbonate-poor lithology are far less abundant in the carbonate-rich lithology. However, coarser grained

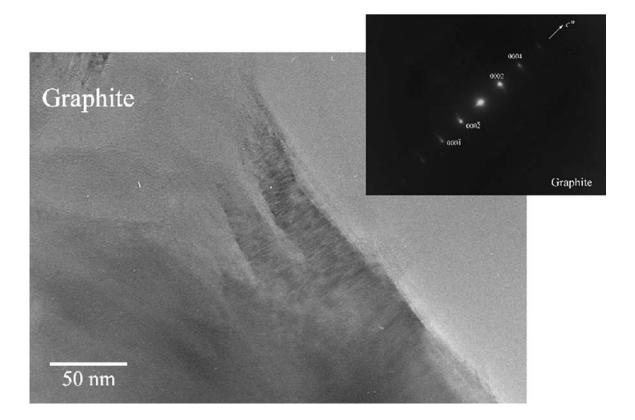


FIG. 17. Transmission electron microscope image of a graphite grain from the matrix of the carbonate-poor lithology. Scale bar measures 50 nm.

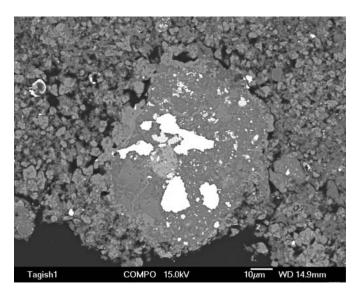


FIG. 18. Backscattered electron image of a phyllosilicate (dark grey)-magnetite (white)-andradite (light grey in the center) aggregate in the carbonate-rich lithology. A rim of carbonates (light grey) surrounds the aggregate. Scale bar measures  $10 \, \mu m$ .

sulfides are a little more abundant. The compositional range of the sulfides in the two Tagish Lake lithologies are identical.

Fe-Mg-Ca-Mn carbonates are *very* abundant in the carbonaterich lithology (Fig. 19), and are apparent in hand specimens as whitish grains. Grains consisting solely of calcium carbonate are rare here, as they are in the carbonate-poor lithology. This Ca carbonate is present mainly as dispersed grains in the matrix, and is also intimately associated with phyllosilicates in small aggregates (Fig. 20). These separate Ca carbonate grains contain no detectable MgO or MnO, and the iron content is below 0.34 wt% FeO.

The great majority of the carbonate in the carbonate-rich lithology first appeared, on the basis of microprobe analyses, to be breunnerite, with a MnO content that reached 2.91 wt%, and a CaO content up to 4.46 wt%. However, after we made numerous high-resolution element maps we realized that these grains are actually complex intergrowths of different generations of carbonates of wildly varying composition (Figs. 20-22). The associations are all the same: cores of Ca carbonate have thin  $(1-2 \mu m)$  rims and separations of Fe-Mg-Mn carbonates (siderite) with essentially no Ca. The element maps reveal that these siderites follow irregular fractures in the Ca carbonates as well as rimming them (Fig. 21). The fact that the fractures are irregular, and curve, rather than following cleavage or parting planes suggests that the fractures could have been produced by impacts. We have observed similar curved fractures in optical-grade calcite from the Harding mine, New Mexico, USA (M. Zolensky, unpubl. observation; Northrop, 1959), apparently caused by overzealous blasting during mining operations (C. T. Smith, pers. comm., 1975). Mary Sue Bell has also observed curved fractures developed in calcite experimentally shocked to a minimum of 14 GPa (Bell, 1997). We hypothesize that fluids percolating through the Tagish Lake

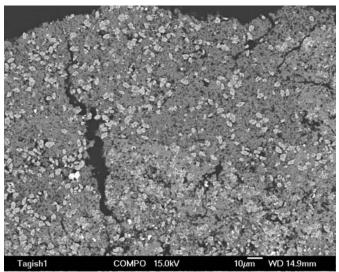


FIG. 19. Backscattered electron image of typical carbonate-rich lithology. Essentially all of the light colored grains are complex carbonate grains. Scale bar measures  $10 \,\mu\text{m}$ .

parent asteroid, after the fracturing event, replaced and coated existing Ca carbonate surfaces with siderite. It is unfortunate that these Mn-enriched siderite separations are so narrow, as this may preclude any efforts to date their formation by Mn-Cr systematics.

These complex carbonates have a variety of associations (Fig. 20). In some places they are grouped into loose aggregates, and many constituent carbonate grains have cores of olivine with irregular boundaries. In these cases calcium carbonate has clearly replaced olivine in amoeboid olivine aggregates. The carbonates are also complexly intergrown with magnetite and phyllosilicates into rounded aggregates; these probably also originated as olivine aggregates. Carbonates rim phyllosilicate grains; obviously a partial replacement texture. Finally, carbonates partially fill practically all of the large pores in this lithology, as mentioned above.

#### CM1 Clast

Present in two sections (KN1-1 and KN1-2) cut from a single stone, one large clast is strikingly dissimilar to the remainder of the meteorite. The clast consists of abundant completely-altered olivine aggregates and chondrules (barred and porphyritic), surrounded by very fine-grained matrix (Fig. 23). These objects are up to 0.5 mm in diameter, and are quite remarkable. The aggregates and chondrules are almost completely altered to phyllosilicates and opaques, in absolutely perfect pseudomorphs. Olivine, pyroxene and glass has been replaced by light brown, transparent to translucent phyllosilicates (Table 2); only a couple of olivine grains remain in each section. Metal has been completely replaced by pyrrhotite and magnetite. Some of the pseudomorphs after olivine consist of completely transparent grains of

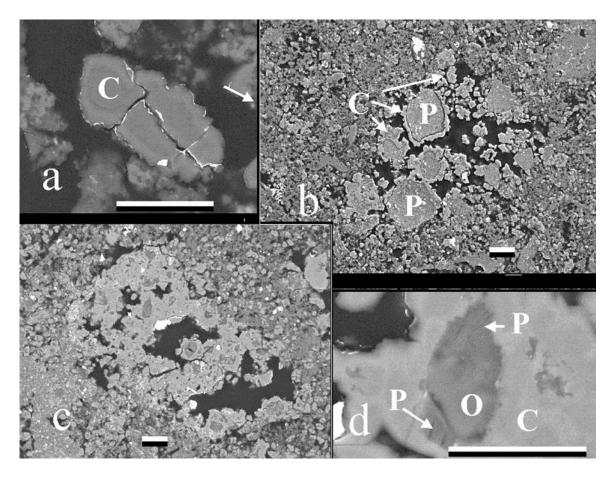


FIG. 20. Backscattered electron images illustrating typical morphologies of carbonates (C) in Tagish Lake. Phyllosilicates are indicated by "P". Scale bars measure  $10 \mu m$ . (a) Ca-carbonate grain from the carbonate-poor lithology. (b) Typical carbonate occurrences in the carbonate-rich lithology. Phyllosilicates (P) are rimmed (partially replaced) by Ca-Mg-Fe carbonates (C). The very small white grains are magnetite. (c) and (d) Olivine aggregate partially replaced by phyllosilicate, which in turn is largely replaced by carbonate.

phyllosilicate, which may be single crystals. The replacement is so perfect that the section gives the appearance of merely being a rather thin section of an ordinary unaltered carbonaceous chondrite. Only a general light-brown color in transmitted light suggests that something is unusual. Some of the aggregates and chondrules have igneous rims; all have finegrained rims. In some cases the fine-grained rims have a layered aspect, as in most CM2 chondrites (Metzler *et al.*, 1992), due to varying quantities of submicron-sized sulfide grains.

Coarse-grained sulfides are very common, in marked contrast to the bulk of Tagish Lake. There are also pyrrhotite veins (!) up to 1 cm in length, which cut completely across chondrules and aggregates (Fig. 23). The clast is carbonate-poor. Magnetite is abundant, and is complexly intergrown with the sulfides. As in Tagish Lake proper, it appears that there were several generations of sulfide and magnetite deposition. Abundant sulfide-magnetite aggregates appear to record a final stage of magnetite deposition (at least there are fine rims of magnetite present in most cases). However, the sulfide veins appear to lack these magnetite rims. How could this happen? This clast appears identical to material we have previously

observed as clasts in Kaidun, and which we have called CM1 (Zolensky *et al.*, 1991).

#### DISCUSSION

### **Alteration History**

There are several extraordinary features of this meteorite, one of which bears on its remarkably well-preserved alteration history. The two different lithologies are sufficiently similar in basic mineralogy to suggest a genetic relationship, although such a relationship is not required. The mineralogical and isotopic (Engrand *et al.*, 2001) similarities between the anhydrous components of the two lithologies demonstrate that the two lithologies had similar anhydrous precursors; therefore, the differences between the secondary minerals can help to trace back different alteration histories. However, each lithology records a different stage of alteration, and petrographic textures are sufficiently well preserved to permit us to decipher the alteration sequence as is rarely possible with other chondrites.

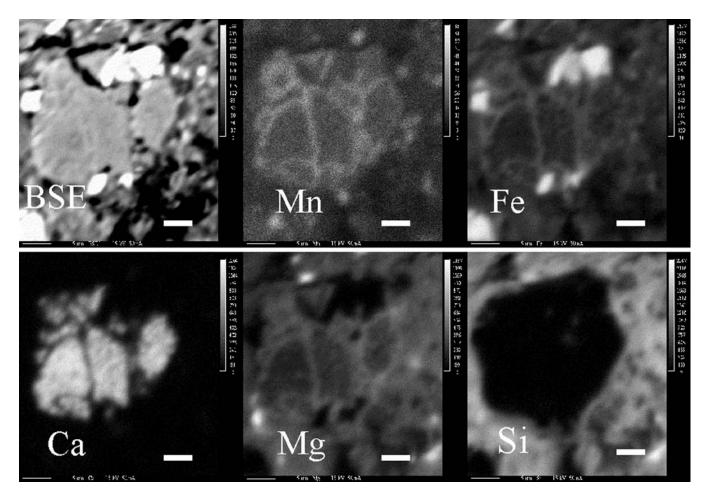


FIG. 21. Backscattered electron and element maps of a complex carbonate grain from the carbonate-rich lithology. Elements being mapped are Mn, Fe, Ca, Mg and Si. Scale bars measure  $5 \mu m$ .

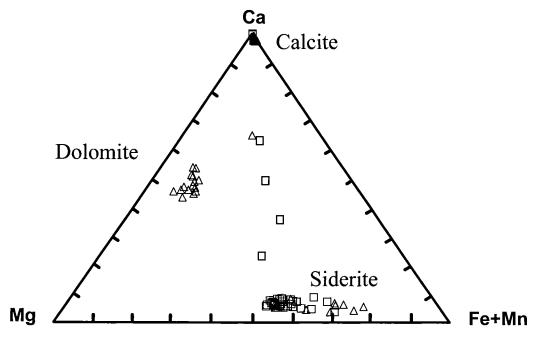


FIG. 22. Composition of carbonates in Tagish Lake (WDS data), plotted onto a Ca, Fe, Mg atom% ternary diagram. Triangles are analyses from the carbonate-poor lithology, squares are from the carbonate-rich lithology.

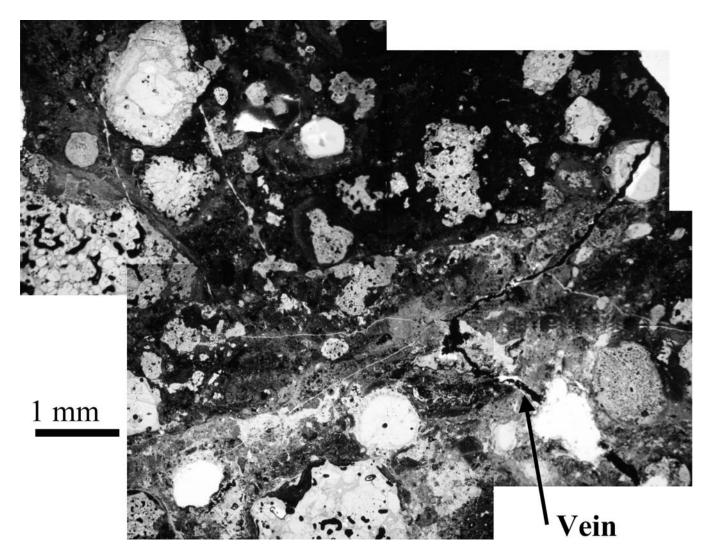


FIG. 23. Transmitted light image of the CM-like foreign clast. Abundant chondrules are evident. A thin, sinuous pyrrhotite vein (black, arrowed) cross-cuts the clast.

Both the carbonate-poor and carbonate-rich lithologies have been affected by a significant amount of aqueous alteration, but the reactions have not proceeded to completion. Olivine and pyroxene grains are commonly found partially transformed into phyllosilicates (Fig. 20), and all of them show evidence of partial dissolution (Fig. 6). Mesostasis in chondrules is almost entirely replaced by phyllosilicates (Fig. 4). CAIs are almost unrecognizable, which indicates a more pervasive alteration than any other CAI-bearing C2 chondrite. Both saponite and serpentine are ubiquitous, in both coarse- and fine-grained crystals. The rather Mg-rich compositions of the serpentines suggest an advanced level of aqueous alteration (Browning *et al.*, 1996). Porosity is high, and all of the pores have partial (in the carbonate-poor lithology) to complete (in the carbonate-rich lithilogy) linings of Ca-carbonate.

Sulfides are rather rare, compared to other type 2 chondrites, and they are generally present only as very small grains in

matrix. The small, rounded sulfides are generally present only among the finely-crystalline saponite clumps, while the larger and euhedral pyrrhotites are usually surrounded by the coarser grained saponite and saponite-serpentine flakes. This suggests a genetic relationship. Perhaps the coarser minerals are growing at the expense of the finer grained assemblage; or perhaps the presence of serpentine indicates a varying pH of the aqueous solution on the asteroid (Zolensky *et al.*, 1993). The majority of the sulfides are pyrrhotite, but pentlandite and intermediate composition sulfides are also present. As we have seen, this compositional range is consistent with incomplete aqueous alteration.

The incomplete extent of aqueous alteration witnessed by the sulfides and phyllosilicates in Tagish Lake tells us that the general lack of chondrules and CAIs is not due to extensive aqueous alteration having erased them, rather they were simply never present in significant numbers. It is clear that coarse-grained sulfides were at one time present, as pseudomorphs of magnetite after pyrrhotite bear witness (Fig. 8a). Obviously, oxidation of these sulfides has occurred, transforming them into magnetite framboids and placquettes. Such an event would clearly have also destroyed very fine-grained sulfides in the matrix. The presence of these therefore indicates that a later stage of sulfide deposition has occurred. Despite this late event, sulfur has clearly been removed, at least from the inorganic phases. It is likely that this sulfur was incorporated into the evolving organic phases. When we thawed one frozen stone in the lab there was a pronounced sulfur smell, indicating that a sulfur-bearing volatile phase, probably an organic material, was being lost.

There are clear differences between the dominant Tagish Lake lithlologies. Unusual Fe-Cr phosphides (Fig. 13) of unknown origin are present in at least one phyllosilicate aggregate in the carbonate-poor lithology. Magnetite is the most abundant opaque phase in this same lithology, at all sizes. The magnetite and sulfides are often intergrown on such a fine scale it is impossible to judge which formed first (see Fig. 9g). However, many altered chondrules and phyllosilicate aggregates in the carbonate-poor lithology have sulfideenriched rims (Fig. 9h-k), which is consistent with the sulfides being a late-stage mineral. Carbonates in the carbonate-poor lithology are mainly Ca-rich, and have been deposited mainly in open pores, and replaces some of the phyllosilicates (Fig. 20). Figure 20c,d shows one instance where olivine is incompletely replaced by phyllosilicates, and that material has in turn been incompletely replaced by Ca carbonate.

The carbonate-rich lithology records further deposition of carbonates, first Ca-rich, but changing to more Fe-Mg-Mn rich siderite. These carbonates are deposited into open pores as well as replacing phyllosilicates. In one place andradite is present in the core of a phyllosilicate aggregate. Compared with the carbonate-poor lithology magnetite is less abundant, and in places is clearly being overgrown by a late-stage generation of sulfides.

The apparent sequence of secondary (alteration) mineralization displayed in the carbonate-poor lithology is: phyllosilicates/pyrrhotite replace anhydrous silicates and metal, then magnetite replaces pyrrhotite, then fine-grained sulfides precipitate in matrix, then carbonates grow in matrix and open pores. The composition of the precipitating phases requires the presence of an abundant quantity of CO- and COS-rich fluids, present for prolonged periods at relatively low temperatures.

Mineral associations reveal the following order of alteration for the carbonate-rich lithology: aqueous alteration producing phyllosilicates and magnetite (although the latter is present in lower abundance than in the other lithology), deposition of Ca carbonates including replacement of some silicates, impact fracturing, deposition of siderite including replacement of some Ca-carbonates and possibly other silicates. Coarse-grained sulfides are deposited. Finally, andradite precipitates, although there are insufficient observations to permit us to suggest why

or how it appeared. The wide extent and variable chemistry of the aqueous fluid suggested by this mineral paragenesis indicates that the fluid was more abundant than for most other carbonaceous chondrites asteroids, and highly mobile. This conclusion is supported by the results of the oxygen isotopic analysis (on a bulk sample) by Clayton and Mayeda (2001), which requires a water/rock ratio of ~2, a factor of 2 over typical CM or CI chondrites.

We note again that this discussion assumes that the two dominant lithologies of Tagish Lake are related, and that the carbonate-rich lithology succeeded the carbonate-poor one. Both of these assumptions may be wrong. Perhaps the two lithologies have no genetic relationship. In this case they would share a common precursor mineralogy, but record different alteration processes marked by different ice/water ratios, and duration of aqueous alteration. It is also interesting to consider the case that the carbonate rich-lithology may precede the carbonate-poor one (rather than the opposite way around). Future workers should keep this possibility in mind.

The large CM-like clast described above records replacement of magnetite by pyrrhotite and pentlandite, often in coarse-grained crystals, veins, and clast rims. Carbonates are essentially absent from this lithology—either they were dissolved or never precipitated here in the first place. There is no reason to require this clast to be related to Tagish Lake proper. In fact, the phyllosilicate compositions in this clast have much lower Al<sub>2</sub>O<sub>3</sub> contents than that generally present in Tagish Lake (Table 2). However, the presence of mineralogically similar clasts within Kaidun, and the similarity of the oxygen isotopic composition of one C1 Kaidun lithology with Tagish Lake may not be a coincidence. More *in situ* oxygen isotope measurements are envisioned for this clast, in order to assess its link with the Kaidun lithology.

The presence of poorly-graphitized carbon, and the hydrocarbon globules, among other features requires that aqueous alteration occurred at low temperatures, probably <100 °C. The oxygen isotopic composition suggests a temperature near 0 °C (Clayton and Mayeda, 2001). Nevertheless, there is a small amount of chlorite present in Tagish Lake, which could indicate locally higher temperatures. Also, the apparent dissolution of magnetite in the carbonate-rich lithology could indicate higher temperatures (>300 °C), for the formation of the carbonate-rich lithology. We unfortunately do not know exactly which mix of lithologies were in the sample analyzed for oxygen isotopes by Clayton and Mayeda (2001).

Figure 24 diagrams our first appraisal of the sequence of mineralization in Tagish Lake. It should be profitable to compare this sequence with what is observed in other type 2 chondrites (CR, CM), to determine whether there were unique aspects of Tagish Lake's fluid composition evolution, or whether the same trends are observed and, indeed, are generic to hydrous asteroids. The exciting thing about Tagish Lake is that the alteration sequence is fairly readable, and further work will undoubtedly clarify the record to a great degree.

# Mineral Paragenesis

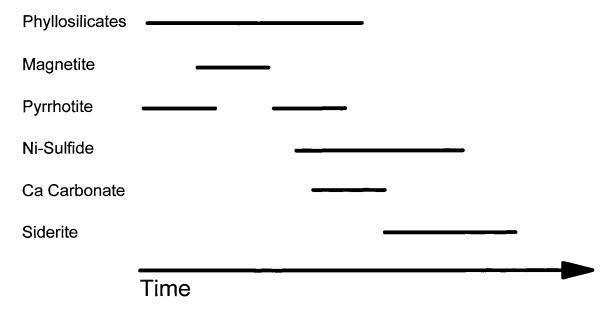


FIG. 24. Diagram summarizing the timing of secondary mineralization in Tagish Lake.

#### What is Tagish Lake, and Why is it so Unusual?

With its pervasive though incomplete aqueous alteration, presence of remnant chondrules, and high carbon and water content Tagish Lake is undoubtedly a type 2 carbonaceous chondrite, but our certainties end there. Abundant phyllosilicates as well as the presence of sparse chondrules and common olivine grains in the matrix preclude any other classification. The rarity of glass and chondrules, and the small size of the latter, distinguish it from available CR2 chondrites (Weisberg et al., 1993), which actually have a similar matrix mineralogy (serpentine, saponite and abundant oxides). Altered chondrules and CAIs, olivine and spinel compositions, and the relative abundance of sulfides are reminiscent of CM2 carbonaceous chondrites. However, the relative abundance and extreme compositional range of carbonates (see Fig. 22), the composition of phyllosilicates, the presence of andradite and the absence of tochilinite are strong differences with the well-known CM2 carbonaceous chondrites (Zolensky et al., 1993). CAIs are also a lot more altered than in CM2 chondrites, and the diopside rims typically present on CAIs have disappeared, if they were in fact ever present in this meteorite. The foreign clast described above appears to be identical to CM1 material found in the Kaidun chondrite, but Kaidun is such a confusing and complex meteorite that this does not advance our understanding. However, the presence of this clast does suggest a possible link to CM asteroids. As more Tagish Lake samples are examined we will learn more about how abundant this foreign clast material really is.

Clayton and Mayeda (2001) have reported on the results of bulk and component oxygen isotope measurements of pristine and degraded Tagish Lake samples, with these yielding  $\delta^{18}O = +18$  to 19.0%,  $^{17}O = +8.3$  to 9.2%. This rather large difference apparently reflects actual sample heterogeneity, and is not due to terrestrial weathering. More detailed oxygen isotopic analysis of component materials in Tagish Lake has been presented by Engrand et al. (2001). The measured isotopic composition for bulk Tagish Lake is far removed from the fields of CM and CR chondrites, and is more similar but still distinct from those of CI chondrites and of the group of metamorphosed carbonaceous chondrites such as Belgica 7904, Yamato 82162, and Yamato 86720. If these data on Tagish Lake are interpreted in terms of Clayton's model of exchange of minerals with liquid water on a parent asteroid, the oxygen isotopic data suggests a higher water/rock ratio than for the CM group, and a lower temperature of aqueous alteration (near 0 °C) than the CI group (Zolensky et al., 1989; Brown et al., 2000). The Tagish Lake bulk oxygen isotopic composition is essentially identical to that of one poorly characterized Kaidun C1 lithology (Robert Clayton and Andrei Ivanov, pers. comm., 2001). We are currently examining this Kaidun material, which does not appear to be identical to Tagish Lake. Again, the possible link of Tagish Lake to Kaidun is interesting.

Large excursions in isotopic composition reported by Grady and coworkers indicate the presence of interstellar grains within Tagish Lake (Brown *et al.*, 2000). They clearly detected nanodiamonds and silicon carbide in Tagish Lake (based upon carbon combustion temperatures), and the presence of both components was matched by similar excursions in  $\delta^{15}$ N during nitrogen release. The bulk elemental composition of Tagish Lake, taken together with its higher concentration of interstellar materials, is consistent with its being more primitive than other C2 chondrites (although these criteria do not require this).

In this paper we have noted similarities between Tagish Lake and some clasts within the enigmatic meteorite Kaidun; possibly there are genetic relationships here worth exploring. In fact the oxygen isotopic composition of the bulk Tagish Lake meteorite is identical to a previously analyzed "C1" clast in Kaidun (Robert Clayton, pers. comm., 2001). We are currently reassessing the nature of this Kaidun material. In any case, Tagish Lake's oxygen isotopic composition is clearly not like any recognized group of meteorites.

Because hydrous phyllosilicates are common in Tagish Lake and the oxygen isotopes are different from both CI1 and CM chondrites (Brown et al., 2000) we propose that Tagish Lake represents a new kind of type 2 carbonaceous chondrite. This is supported by the bulk density of Tagish Lake (1.67 g/cc), which is far lower than CI or CM chondrites (2.2–2.3 and 2.6– 2.9 g/cc, respectively), or any other meteorite for that matter (Wasson, 1974). This low density, extremely high porosity. and the meteorite's extreme friability (as best witnessed by its destruction in the atmosphere into a huge cloud of dust and numerous rather small stones) suggest that even if this type of chondrite were to fall on the Earth rather frequently, it inevitably rapidly perishes in this alien environment. In fact the mineralogy of Tagish Lake is rather similar to Antarctic micrometeorites, and submillimeter-sized clasts found within howardite-eucrite-diogenite (HED) achondrites (Zolensky et al., 1996b; Gounelle et al., 2002, unpubl. data). Gounelle et al. (2002, unpubl. data) have recently suggested that these latter materials are the most abundant materials falling onto Earth throughout its history. Viewed in this light, Tagish Lake may be more representative of extraterrestrial material falling onto Earth than the bulk of recovered meteorite falls. If this is correct then the bulk of the water and organics on Earth could well have arrived in Tagish Lake type materials. Current isotopic studies of the water and organics in Tagish Lake are exploring this intriguing possibility.

Results of the bulk composition determinations by Friedrich et al. (2002) and Mittlefehldt (2002) indicate that the Tagish Lake refractory lithophile element (Zr–Sr) abundances are similar to those of CM chondrites, while the moderately volatile and volatile lithophile element (Mn–Br) abundances are intermediate between CM and CI chondrites. The refractory siderophile element (Re–Pd) abundances are like those of both CI and CM chondrites, whereas the moderately volatile and volatile siderophile element (Au–Tl) and volatile chalcophile

element (Zn–Cd) abundances are generally between those of CI and CM chondrites. In summary, Tagish Lake has a higher bulk volatile content than the other type 2 chondrites (Friedrich *et al.*, 2002; Mittlefehldt, 2002).

Hiroi et al. (2001) suggest that Tagish Lake may be the first meteorite to originate from a D-class asteroid. Certainly, the high carbon (~5 wt%; Brown et al., 2000) and water content of this meteorite, the absence of evidence for high-temperature alteration, the record levels of preserved interstellar materials, low bulk density and extreme porosity are all consistent with an origin far from the Sun. In addition, the strikingly low abundance of chondrules, particularly high-temperature barred olivine and microporphyritic types, suggests an origin unusually far from the chondrule-forming region. If we assume that chondrules were formed by some solar process (Sorby, 1877; Hewins et al., 1996; Shu et al., 2001), then accretion of the Tagish Lake parent asteroid far from the Sun is reasonable. If this suggestion is correct then Tagish Lake may be the most mineralogically primitive meteorite available for study. Certainly its mineralogy is more primitive than that of the thoroughly-altered CI chondrites, despite having a slightly lower bulk volatile content (Friedrich et al., 2002). While CO, CV and CH chondrites are significantly less aqueously altered than Tagish Lake, they also contain lots of chondrules which formed by the wholesale destruction of earlier generations of solids. We contend that some of these earlier, chondruleprecursor materials may be better preserved in Tagish Lake.

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