The Meteoritical Society

Meteoritics & Planetary Science 44, Nr 1, 95–106 (2009)
Abstract available online at http://meteoritics.org

The origin of the Brunflo fossil meteorite and extraterrestrial chromite in mid-Ordovician limestone from the Gärde quarry (Jämtland, central Sweden)

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(Received 24 April 2008; revision accepted 09 August 2008)

Abstract–The Brunflo fossil meteorite was found in the 1950s in mid-Ordovician marine limestone in the Gärde quarry in Jämtland. It originates from strata that are about 5 million years younger than similar limestone that more recently has yielded >50 fossil meteorites in the Thorsberg quarry at Kinnekulle, 600 km to the south. Based primarily on the low TiO_2 content (about 1.8 wt%) of its relict chromite the Brunflo meteorite had been tentatively classified as an H chondrite. The meteorite hence appears to be an anomaly in relation to the Kinnekulle meteorites, in which chromite composition, chondrule mean diameter and oxygen isotopic composition all indicate an L-chondritic origin, reflecting an enhanced flux of meteorites to Earth following the disruption of the L chondrite parent body 470 Ma. New chondrule-size measurements for the Brunflo meteorite indicate that it too is an L chondrite, related to the same parent-body breakup. Chromite maximum diameters and well-defined chondrule structures further show that Brunflo belongs to the L4 or L5 type. Chromites in recently fallen L4 chondrites commonly have low TiO_2 contents similar to the Brunflo chromites, adding support for Brunflo being an L4 chondrite. The limestone in the Gärde quarry is relatively rich (about 0.45 grain kg^{-1}) in sediment-dispersed extraterrestrial chromite grains (>63 μm) with chemical composition similar to those in L chondrites and the limestone (1–3 grains kg^{-1}) at Kinnekulle, suggesting that the enhanced flux of L chondrites prevailed, although somewhat diminished, at the time when the Brunflo meteorite fell.

INTRODUCTION

In 1952, in the Gärde quarry near Brunflo in Jämtland (Fig. 1), the quarry men found a strange black clast in a slab of Middle Ordovician red limestone (Thorslund et al. 1984). The clast was interpreted as an altered terrestrial ultramafic rock that had been transported with floating algae to the deeper parts of the Ordovician sea. The slab was forgotten until 1979 when Per Thorslund realized that the clast may be a fossil meteorite. New studies were initiated showing that the clast was, at the time, the oldest known fossil meteorite (Thorslund and Wickman 1981). Fossil meteorites are extremely rare, and by the end of the 1980s less than a handful of fossil meteorites had been proposed for the entire geological record, with the Brunflo meteorite being the perhaps only really convincing case (Schmitz and Tassinari 2001). Then a second mid-Ordovician meteorite was found in the Thorsberg quarry near Österplana at Kinnekulle in southern Sweden (Fig. 1; Nyström et al. 1988). This find triggered a systematic search for fossil meteorites in the Thorsberg quarry, and soon about four to five meteorites per year were being delivered by the quarry workers (Schmitz et al. 1997, 2001). Forty fossil meteorites had been recovered by the end of 2000, and additionally about forty undescribed finds have been made after that (Schmitz et al. 2001, Forthcoming). The abundance of meteorites on the mid-Ordovician sea floor is far too high to be explained by a meteorite flux similar to that of today. Schmitz et al. (2001) hence suggested a one to two orders-of-magnitude increase in the flux rate following the disruption of the L chondrite parent body in the asteroid belt. This was a major event in solar system history, discovered initially based on low (about 450–500 Ma) gas retention ages in a major fraction of the L chondrites that fall on Earth today (Anders 1964; Turner et al. 1966; Keil et al. 1994). New high-precision Ar-Ar dates of recently fallen L chondrites have constrained the age of the L chondrite parent disruption event to 470 ± 6 Ma, which coincides within error with the age of the meteorite-rich beds at Kinnekulle according to the current geological time scale (Cooper and Sadler 2004; Korochantseva et al. 2007). The origin of the fossil meteorites at Kinnekulle from this event is supported by the trace element and oxygen isotopic composition of relict chromite

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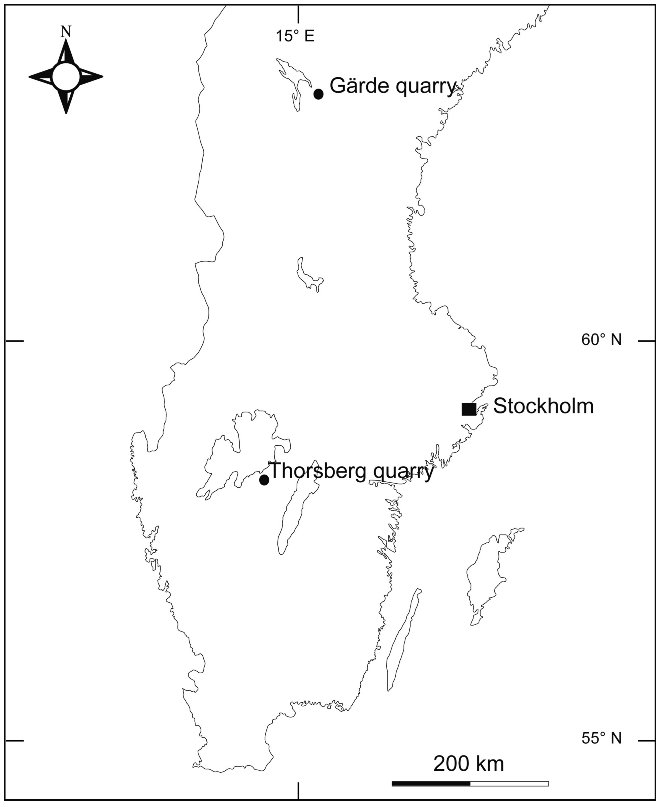

Fig. 1. Map of southern and central Sweden with the locations of the Gärde and Thorsberg quarries.

and chondrule size measurements, indicating that most or all of the meteorites are pseudomorphosed L chondrites (Schmitz et al. 2001; Bridges et al. 2007; Greenwood et al. 2007). The meteorites represent a range of petrographic types, 3/4 to 6, and are definitely not just one fall that has been reworked in some way (Bridges et al. 2007). Additional support comes from sediment-dispersed extraterrestrial (ordinary chondritic) chromite (EC) grains (>63 µm) from decomposed micrometeorites and meteorites in condensed (i.e., very slowly deposited) limestone in southern Sweden and central China (Schmitz et al. 2003; Schmitz and Häggström 2006; Häggström and Schmitz 2007; Heck et al. 2008; Schmitz et al. 2008). Condensed limestone of mid- to late Arenigian age contains only rare EC grains, on the order 0.01–0.02 grain kg^{-1}, whereas latest Arenigian–early Llanvirnian limestone, formed at the same rate, contains 1–3 EC grains kg^{-1}. This represents a strong case for a two orders-of-magnitude increase also in the flux of micrometeorites to Earth after the asteroid breakup event.

The Brunflo meteorite was tentatively classified as an H chondrite by Thorslund et al. (1984), based on the relatively low TiO_2 (1.8 wt%) concentrations in its relict chromite and the relative abundance of different textural and petrological chondrule types. This meteorite hence appears to be an anomaly relative to the abundant L chondrites found in the about 5 million years older rocks at Kinnekulle. Here we try to further constrain the origin of Brunflo by re-examining its chromite chemistry and some of its petrological characteristics in light of a more recent understanding of features that can be used to classify meteorites. For example, the mean chondrule size differs between H and L chondrites (Grossman et al. 1988; Rubin 2000) and the maximum diameter of chromite differs with petrographic type, circumstances used by Bridges et al. (2007) to classify fossil meteorites from Kinnekulle. As regards the general petrography of Brunflo and the diagenesis and formation of secondary minerals in the meteorite we refer to the detailed studies by Thorslund et al. (1984), Nyström and Wickman (1991) and Hofmann et al. (2000). Here we also determine the distribution and composition of sediment-dispersed EC grains in the limestone strata from which the Brunflo meteorite originates in the Gärde quarry. The aim is to determine if the increased flux of L-chondritic matter to Earth was sustained at the time Brunflo fell. This can be anticipated according to model simulations for major parent body breakup events (Zappalà et al. 1998).

THE GÄRDE QUARRY

The quarry in which the Brunflo meteorite was found is known as the Gärde quarry, Rödbrottet or Rödberget, and is situated at Gärde, 2–3 km east of Brunflo in the central part of Sweden (63°5′ N, 14°53′ E) (Thorslund et al. 1984). The quarry exposes a 7 m thick section of condensed Orthoceratite Limestone of the same type that yields the abundant fossil meteorites at Kinnekulle. The entire section at Gärde in terms of conodont zonation belongs to the *Eoplacognathus suecicus* Zone, with the lower ~3 m assigned to the *E. suecicus—Scalpellodus gracilis* Subzone and the upper about 4 m to the *E. suecicus—Panderodus sulcatus* Subzone (Löfgren 1978). The limestone plate containing the Brunflo meteorite yields conodonts of the *E. suecicus-P. sulcatus* Subzone, i.e., the meteorite originates from the upper part of the Gärde quarry.

The strata in the Gärde quarry are clearly younger than the meteorite-yielding strata at Kinnekulle, but assigning an absolute age difference is difficult (Fig. 2). According to the latest geological time scale, the upper part of the *E. suecicus* Zone has an age of about 464 Ma, whereas the meteorite-rich lower part of the *E. variabilis* Zone, i.e., the conodont zone that immediately underlies the *E. suecicus* Zone, is dated to about 467 Ma (Cooper and Sadler 2004); this would then indicate an about 3–4 Myr time difference. These numbers, however, should be treated with caution because they imply significantly higher sedimentation rates for the Orthoceratite Limestone than generally assumed (Lindström 1971), and by extension also higher meteorite flux rates for the mid-Ordovician than presently estimated. The vertical distance in the rock record between the lower *E.*

variabilis Zone and the lower *E. suecicus–P. sulcatus* Subzone is about 16 m (data for Kalkberget and Gusta sections in Löfgren 1978), with the *E. variabilis* Zone representing 12 m. At Kinnekulle the *E. variabilis* Zone is cut after about 11 m by a hiatus and the entire *E. suecicus* Zone is lacking. All the subzones of the *E. variabilis* Zone occur at this locality and probably only a minor part of the uppermost zone is absent (Schmitz and Häggström 2006). Assuming sedimentation rates of 2 mm per 1000 years, i.e., the sedimentation rates used to estimate mid-Ordovician meteorite flux (Schmitz et al. 2001), implies that the Brunflo meteorite fell on the order of 8–10 million years after the oldest meteorites at Kinnekulle, and 6–8 million years after the youngest meteorites. Admittedly there is an uncertainty in the sedimentation rates and, considering both the new geological time scale and the actual rock record, we suggest an about 5 Myr time difference between the meteorites at Kinnekulle and the Brunflo meteorite. It is clear, however, that the Gärde section represents an upward continuation of the 22 m thick section previously searched for extraterrestrial chromite at Kinnekulle (Schmitz and Häggström 2006). The Gärde samples are clearly 1–2 million years younger than the highest sample, 8.5 m above the base of the lowest meteorite-carrying bed, the Arkeologen bed, studied at Kinnekulle (Fig. 2).

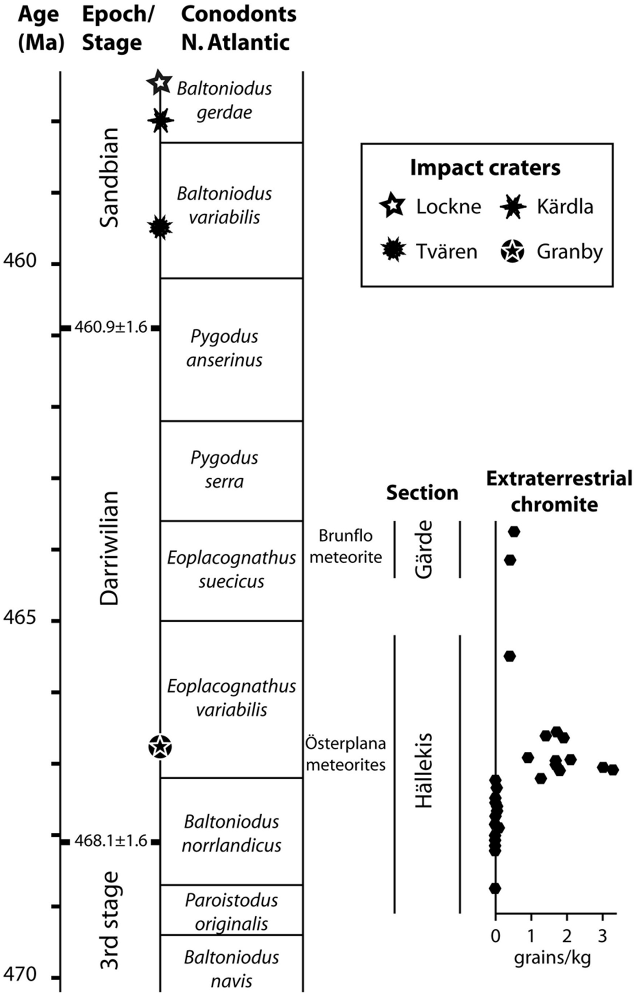

Fig. 2. General stratigraphic scheme for the mid-Ordovician (after Cooper and Sadler 2004). Stratigraphic position of four mid-Ordovician Baltoscandian impact craters after Grahn et al. (1996). Distribution of extraterrestrial chromite in the Gärde quarry from this study and from the Hällekis section after Schmitz and Häggström (2006).

MATERIALS AND METHODS

Small pieces from the Brunflo meteorite were collected with a chisel and hammer from the backside of the meteorite-carrying limestone slab, now deposited at the Natural History Museum in Stockholm. The samples were dissolved in 6 M hydrochloric acid, and chromite grains were recovered for chemical analyses. Chondrule sizes in the meteorite were measured on photos of the meteorite originally taken for the article by Thorslund et al. (1984), and now stored with the meteorite in Stockholm. These photos are of excellent quality but lack scales. Some of the photos were used in the 1984 publication with a millimeter scale, and from these photos the scale for the remaining photos could be established. For chondrules that were fragmented or non-circular, the maximum diameter was measured. The photos used by R. Hutchison for chondrule size measurements of the fossil meteorite Österplana 011 (synonym name Ark 011) from the Thorsberg quarry, in Bridges et al. (2007), were remeasured in order to check for consistency. We measure and compare apparent chondrule sizes, i.e., in a two dimensional section. The measurement of chromite grain size was carried out on thin sections of the meteorite, according to the methods developed by Bridges et al. (2007), using a scanning electron microscope with a backscatter detector. Chromite identity was established by element analysis (see below).

From the Gärde quarry two samples of limestone, with weights of 23.5 and 27.4 kg, were collected at 2.70–3.00 and 5.62–5.75 m, respectively, above the base of the 7 m section described in Löfgren (1978). Both samples are within the *E. suecicus—P. sulcatus Subzone*, i.e., from the same subzone that yielded the Brunflo meteorite. The samples were split with a sledge-hammer into pieces smaller than 8 cm and then further crushed to a fragment size of 5 mm using a Retsch Jaw Crusher BB 200. The crushed material was decalcified in 6 M hydrochloric acid at room temperature. After sieving the residue at 32 μm, the remaining fraction was leached with 18 M hydrofluoric acid at room temperature with occasional stirring. The residue was recovered with a 32 μm sieve. The acid-insoluble fraction in the size range 63 to 355 μm was searched under the binocular microscope and suspected chromite grains were picked with a fine brush for element analyses. The acid-insoluble fractions in this study differ from samples in previous studies by containing abundant black, chromite-resembling iron oxide grains, creating difficulties in the chromite search. For detailed chromite quantifications, we avoid in most cases magnetic or heavy liquid separation techniques because this adds uncertainties. Usually it is enough to scan a residue under the light microscope two or three times for a complete recovery of grains, but as long as a repeated search gives new grains, another search is warranted. The grain residues in this study were searched five to eight times for chromite grains. In total five samples had been collected from the Gärde quarry, but because of the tedious nature of the chromite searches only two samples were studied.

All chromite grains for chemical analyses were mounted in epoxy resin and polished flat using a 1 µm diamond slurry. Quantative element analyses were performed with an energy dispersive spectrometer (Inca X-sight from Oxford instruments) with a Si detector, mounted on a Hitachi S-3400 scanning electron microscope. Cobalt was used for standard; the acceleration voltage was 15 kV, the sample current about 1 nA and the counting live-time 80 s. Precision (reproducibility) of analyses was typically better than 1–4%. Analytical accuracy was controlled by repeated analyses of the USNM 117075 (Smithsonian) chromite reference standard (Jarosewich et al. 1980). The concentration of ferric iron was calculated using the formula XY_2O_4 and based on the assumption of perfect stoichiometry, where X = Fe^{2+}, Ni, Mn, Mg, Zn and Cu, and Y= Cr, Al, V and Fe^{3+}; Ti is calculated as being in an ulvöspinel component.

RESULTS

Brunflo Chromite Composition

The chemical results for 36 polished chromite grains from the Brunflo meteorite are shown in Table 1. Our results are very similar to the results by Thorslund et al. (1984) for their grains with low ZnO content (Table 2). These authors divided their grains into two categories, grains with more and less than 1 wt% ZnO. Grains with up to 6.8 wt% ZnO were found in thin sections and this was attributed to diagenetic alteration of the chromite grains. Only five of our grains have ZnO concentrations significantly higher than 1.0 wt% and the highest value is 2.7 wt%. Test analyses, however, of the surfaces of unpolished grains gave some higher ZnO concentrations, up to 11%, indicating that Zn is preferentially bound to the surfaces of the grains. Schmitz et al. (2001) showed for the meteorites at Kinnekulle that there is an inverse correlation between FeO and ZnO, and explained this by substitution of Zn for Fe in the chromite lattice during diagenesis. The Brunflo chromite differs slightly in composition from the "average" chromite of the Kinnekulle meteorites in so far that it has higher FeO and Al_2O_3 and lower TiO_2 (Table 2). The TiO_2 content of extraterrestrial chromite is particularly indicative of its origin from a specific type of ordinary chondrite (Bunch et al. 1967; Schmitz et al. 2001). The observation by Thorslund et al. (1984) that the Brunflo chromite has a TiO_2 content (1.8 wt%) more similar to the average chromite of recently fallen H (2.2 wt%) than L (2.7 wt%) chondrites is corroborated here (Table 2; see also Bunch et al. 1967 and Wlotzka 2005). However, as discussed in a later section, we do not think this feature is evidence of that the Brunflo meteorite is an H chondrite.

The new analyses presented here show that in relation to the more than 1000 extraterrestrial chromite grains studied in our lab during the past decade, the Brunflo chromites, apart from minor ZnO replacement, appear very well preserved. They show none of the features of altered grains from the Lockne impact crater resurge deposits, where hydrothermal activity has affected grains, or from a few of the fossil meteorites at Kinnekulle that show signs of severe corrosion (Schmitz et al. 2001; Alwmark and Schmitz 2007). The TiO_2 content is one of the most stable features of the chromite chemistry during diagenesis. The replacement of Fe and sometimes Mg, with Zn, and in some cases also Mn, can dramatically alter the Fe content of a grain but does not have any significant effect on the Ti and Al contents. Another effect seen in some cases is a small increase, typically from 57 to 60–61 wt% in Cr_2O_3 at the expense of FeO, which may cause a slight lowering of the TiO_2 concentration (Schmitz et al. 2001; Alwmark and Schmitz 2007), but there is no indication of this in the Brunflo chromites. On the contrary, the Cr_2O_3 concentrations of the Brunflo chromites are persistently low, on average 56.4 wt%, and FeO is high, on average 29.2 wt% (Tables 1–2). Only in the few grains with slightly enhanced ZnO does the FeO fall to concentrations of 27.5 wt%, which is still slightly higher than the mean of 26.9 wt% for 26 fossil meteorites from Kinnekulle. The results show that, except minor Zn replacement, the Brunflo chromites are well preserved (in terms of major and trace elements) and the low TiO_2 concentrations represent an original, relict feature.

Brunflo Chondrule and Chromite Sizes

A total of 140 chondrules from photos of the Brunflo meteorite were measured (Fig. 3), giving a mean diameter of 0.49 mm (Table 3). This corresponds well with the average size of chondrules in L group chondrites (0.5 mm) but not in H chondrites (0.3 mm) (Grossman et al. 1988; Rubin 2000; Bridges et al. 2007). When the measurements are plotted as frequency in phi units ($-\log_2 d$) they show a near log-normal distribution (Fig. 4), which is the typical distribution of chondrule sizes in recently fallen chondrites (e.g., King and King 1979; Eisenhour 1996), thus confirming the validity of our measurements. Furthermore, the remeasurement of 162 chondrules from Österplana 011 (Ark 011) gave a mean chondrule diameter of 0.52 mm, which is in good agreement with the previous mean of 0.49 mm (Bridges et al. 2007).

Chromite grains (>5 µm) were identified and measured

Table 1. Element concentration (wt%) of chromites from the Brunflo meteorite. Each value is a mean of three different analysis points of each individual grain.

Grain no.	Cr_2O_3	Al_2O_3	MgO	TiO_2	V_2O_3	FeO_{tot}	FeO	Fe_2O_3	MnO	ZnO	Total	Fe#	Cr#
1	56.81	6.64	2.00	1.85	0.89	29.60	29.60	0.00	1.18	1.01	99.98	89.3	85.2
2	56.28	6.77	2.40	1.91	0.67	28.97	28.97	0.00	0.84	0.88	98.72	87.1	84.8
3	56.69	6.28	2.50	2.07	0.65	29.28	29.28	0.00	0.74	b.d.	98.21	86.8	85.8
4	56.83	6.85	2.17	1.90	0.75	29.41	29.41	0.00	1.08	0.66	99.65	88.4	84.8
5	56.83	6.75	2.26	1.98	0.71	29.26	29.26	0.00	0.92	0.65	99.36	87.9	85.0
6	57.34	7.00	2.18	1.87	0.72	29.75	29.75	0.00	0.58	0.38	99.82	88.5	84.6
7	56.42	7.09	2.23	1.79	0.69	29.26	29.26	0.00	0.87	0.66	99.01	88.0	84.2
8	55.63	6.85	2.29	1.80	0.73	29.48	29.47	0.01	0.70	0.51	97.99	87.8	84.5
9	56.20	6.65	2.33	2.01	0.64	29.26	29.26	0.00	0.89	0.64	98.62	87.6	85.0
10	56.91	6.69	2.34	1.96	0.74	29.52	29.52	0.00	0.75	0.83	99.74	87.6	85.1
11	55.85	6.59	2.28	2.15	0.72	29.44	29.44	0.00	0.34	1.05	98.42	87.9	85.0
12	55.72	6.66	1.78	1.75	0.81	28.56	28.56	0.00	0.66	2.29	98.23	90.0	84.9
13	56.73	7.22	2.40	1.71	0.81	28.68	28.68	0.00	0.86	0.66	99.07	87.0	84.0
14	56.50	6.56	2.42	1.91	0.76	29.60	29.60	0.00	b.d.	0.81	98.56	87.3	85.2
15	56.53	6.61	2.28	1.91	0.64	29.19	29.19	0.00	0.84	0.47	98.47	87.8	85.2
16	56.66	6.06	2.00	2.26	0.63	28.81	28.81	0.00	1.05	1.78	99.25	89.0	86.2
17	57.15	6.71	1.92	1.91	0.67	28.30	28.30	0.00	0.54	2.11	99.31	89.2	85.1
18	55.90	6.88	2.33	2.07	0.77	29.81	29.55	0.29	1.13	0.73	99.65	87.7	84.5
19	55.78	6.65	2.25	1.90	0.74	29.26	29.26	0.00	0.93	0.68	98.19	87.9	84.9
20	56.12	6.95	2.26	1.69	0.77	29.30	29.29	0.02	0.93	0.66	98.68	87.9	84.4
21	56.47	7.08	2.16	1.85	0.81	29.09	29.09	0.00	0.65	0.92	99.03	88.3	84.2
22	54.83	7.39	2.25	1.65	0.86	29.16	29.15	0.01	0.81	0.60	97.55	87.9	83.3
23	57.07	6.37	2.03	1.89	0.78	29.88	29.88	0.00	0.63	0.33	98.98	89.2	85.7
24	56.16	6.91	1.99	1.66	0.60	29.53	29.53	0.00	0.91	0.65	98.41	89.3	84.5
25	55.39	7.31	1.71	1.82	0.58	27.47	27.47	0.00	0.54	2.73	97.55	90.0	83.6
26	55.82	7.06	2.08	2.02	0.83	28.93	28.93	0.00	0.64	1.73	99.11	88.6	84.1
27	57.34	6.87	2.36	2.01	0.73	29.13	29.13	0.00	0.75	0.87	100.06	87.4	84.8
28	56.26	6.73	2.43	2.01	0.66	29.00	29.00	0.00	0.97	0.67	98.73	87.0	84.9
29	55.93	6.72	2.15	1.50	0.62	28.44	28.44	0.00	1.00	0.85	97.21	88.1	84.8
30	57.51	6.48	2.08	1.94	0.89	29.30	29.30	0.00	1.16	0.49	99.85	88.8	85.6
31	56.66	6.97	2.17	1.91	0.84	29.35	29.35	0.00	0.47	0.71	99.08	88.4	84.5
32	56.14	7.04	2.44	1.65	0.61	28.50	28.50	0.00	0.87	0.68	97.93	86.8	84.2
33	57.17	6.84	2.16	1.88	0.77	29.14	29.14	0.00	0.63	0.52	99.11	88.3	84.9
34	56.38	6.96	2.27	1.87	0.71	29.97	29.88	0.10	0.94	0.48	99.59	88.1	84.5
35	55.41	6.74	2.36	1.93	0.82	29.93	29.68	0.28	0.52	0.59	98.33	87.6	84.6
36	57.14	7.09	2.35	1.75	0.64	29.60	29.60	0.00	0.79	0.71	100.07	87.6	84.4
Mean	56.40	6.81	2.21	1.88	0.73	29.20	29.18	0.02	0.78	0.86	98.88	88.1	84.8
Std dev. (1σ)	0.62	0.28	0.18	0.15	0.08	0.51	0.49	0.07	0.24	0.57	0.75	0.8	0.6

b.d: below detection limit; Fe#: mol% $Fe^{2+}/(Fe^{2+} + Mg)$; Cr#: mol% $Cr/(Cr + Al)$.

Table 2. The average element concentration (wt% and standard deviation 1σ) of the Brunflo chromite grains in comparison with other studies.

	Cr_2O_3	Al_2O_3	MgO	TiO_2	V_2O_3	FeO	MnO	ZnO	Fe#	Cr#
The Brunflo chondrite, 36 grains (this study)	56.40 ± 0.62	6.81 ± 0.28	2.21 ± 0.18	1.88 ± 0.15	0.73 ± 0.08	29.20 ± 0.51	0.78 ± 0.24	0.86 ± 0.57	88.1 ± 0.8	84.8 ± 0.6
The Brunflo chondrite, 53 grains with ZnO < 1% (Thorslund et al. 1984)	57.14	6.45	1.84	1.75	0.74	30.86	0.45	0.56	90.4	85.6
26 meteorites from Thorsberg quarry, Kinnekulle, 594 grains (Schmitz et al. 2001)	57.60 ± 1.30	5.53 ± 0.29	2.57 ± 0.83	2.73 ± 0.40	0.73 ± 0.03	26.94 ± 3.89	1.01 ± 0.33	1.86 ± 2.43	85.3 ± 5.0	87.5 ± 0.6
Sediment dispersed grains from Thorsberg quarry, Kinnekulle, 276 grains (Schmitz and Häggström 2006)	57.61 ±1.58	6.07 ± 0.76	2.58 ± 0.79	3.09 ± 0.33	0.75 ± 0.07	27.36 ± 2.63	0.78 ± 0.20	0.53 ± 0.50	85.6 ± 3.9	86.5 ± 1.4
Sediment dispersed grains from Gärde quarry, 23 grains (this study)	57.59 ± 1.41	5.65 ± 0.36	2.78 ± 0.68	3.01 ± 0.29	0.72 ± 0.07	28.40 ± 1.31	0.80 ± 0.29	0.47 ± 0.67	85.2 ± 3.2	86.7 ± 0.8
12 recent H5/6 group chondrites (Schmitz et al. 2001)	56.64 ± 0.37	6.44 ± 0.14	2.98 ± 0.23	2.20 ± 0.17	0.73 ± 0.02	29.27 ± 0.67	1.00 ± 0.08	0.33 ± 0.05	84.7 ± 1.2	85.5 ± 0.3
12 recent L5/6 group chondrites (Schmitz et al. 2001)	56.00 ± 0.65	5.97 ± 0.43	2.93 ± 0.97	2.68 ± 0.40	0.75 ± 0.02	30.22 ± 2.23	0.83 ± 0.10	0.30 ± 0.07	85.3 ± 4.9	86.3 ± 0.8

Fe#: mol% Fe/(Fe + Mg); Cr#: mol% Cr/(Cr + Al).

Table 3. Chondrule diameter measurements.

Meteorite	No. of chondrules	Mean chondrule diameter[1] (mm)
Brunflo[2]	140	0.49
Österplana 011[2]	162	0.52
Österplana 011[3]	132	0.49
Average H chondrites[4]	–	0.3
Average L chondrites[4]	–	0.5
Average LL chondrites[4]	–	0.6

[1] All data refer to apparent chondrule diameter, measured on a planar surface, see main text.

[2] This study.

[3] Bridges et al. (2007).

[4] Grossman et al. (1988); Rubin (2000).

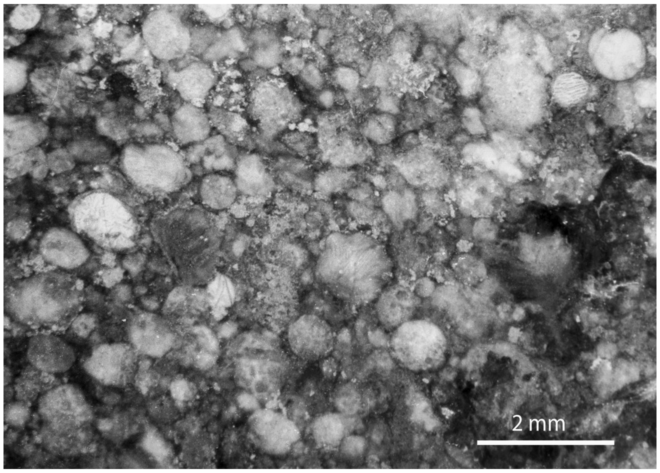

Fig. 3. Well-defined relict chondrules in the Brunflo meteorite; reflected light. The photo was taken for the paper by Thorslund et al. (1984).

in an area of 130 mm^2 on thin sections from the meteorite (Table 4; Fig. 5). This yielded 146 grains or 1.1 grain mm^{-2}. The mean chromite grain size is 37 μm with the largest grain having a diameter of 130 μm. According to Bridges et al. (2007), the largest grain diameter (D_{max}) is the most reliable factor in distinguishing between petrographic types of L chondrites. A comparison with their compiled data for D_{max} in the different types of L chondrites places the Brunflo meteorite in petrographic type 4 or 5 (Table 4). This is consistent with the relatively sharp definition of the chondrules in the meteorite, indicating petrographic type 3 to 5 (Fig. 3). It is also notable that the number of chromite grains (>5 μm) per area unit for Brunflo is comparatively low and within the range of recently fallen L4 chondrites (Table 4). This is significant because a clear trend of increasing spinel abundance with petrologic type was observed also for recently fallen LL chondrites in the study by Kimura et al. (2006).

Sediment-Dispersed Grains

The samples at 2.70 m and 5.62 m contain 9 and 17 extraterrestrial chromite grains (EC), respectively, representing average contents of 0.4 and 0.5 grain kg^{-1}. These numbers probably represent a full recovery of grains, but we cannot entirely exclude the possibility that they rather represent minimum values. A few other terrestrial or unidentified chrome-rich spinel (OC) grains were also found in the two samples (Table 5). The classification of chromite grains into EC and OC groups follows that of Schmitz and Häggström (2006). The EC grains are characterized in the first hand by high Cr_2O_3 contents of ~55–60 wt%, FeO concentrations in the range of ~25–30%, low Al_2O_3 at ~5–8 wt%, and MgO concentrations of ~1.5–4 wt%. The EC grains also plot in narrow ranges of V_2O_3, ~0.6–0.9 wt%, and TiO_2, ~2.0–3.5 wt%, concentrations. For a grain to be classified as an EC grain it has to have a composition within the defined ranges for all the elements listed above. All other chrome spinel grains are classified as OC grains.

The sediment-dispersed EC grains from the Gärde quarry have the typical L (or LL) chondritic composition with, for example, an average TiO_2 concentration of 3.0 wt%, very constant V_2O_3 contents around 0.7 wt%, and low Al_2O_3 (~5–6 wt%). The average composition of the grains is very similar to the average composition of 276 sediment-dispersed grains from the meteorite-rich interval at Kinnekulle (Table 2). Standard deviations are low, indicating a homogeneous composition and a single origin of the grains.

The average content of extraterrestrial chromite in the limestone in the Gärde quarry can be compared with Kinnekulle, where the meteorite-rich zone, i.e., 0.0–3.0 m above the base of the Arkeologen bed, typically contains 1–3 grains kg^{-1} limestone whereas only 5 grains were found in 379 kg of rock from 14 levels throughout the 10 m interval of Orthoceratite Limestone immediately below the base of this bed (Schmitz and Häggström 2006). At Kinnekulle we also searched for extraterrestrial chromite in a single sample of 28 kg at 8.5 m above the base of the Arkeologen bed, i.e., this is the sample that stratigraphically (upper part of *M. ozarkodella* Subzone) lies the closest to the Gärde section. The content of extraterrestrial chromite in this sample, 11 grains, i.e., 0.4 grain kg^{-1}, is more or less identical compared to the two samples from the Gärde section (Fig. 2).

DISCUSSION

Our data suggest that the Brunflo meteorite is an L chondrite, most likely of the L4 petrographic type. Thorslund et al. (1984) emphasized that their assignment of the Brunflo meteorite, based on its relative abundances of different chondrule textural types and low TiO_2 in relict chromite, to the H chondrite group was tentative. The only difference in relative abundances of chondrule types between the three (H, L, LL) ordinary chondrite groups is a suspected slight trend in an increase of porphyritic olivine chondrules from H to L to LL chondrites (Gooding and Keil 1981). Whether the porphyritic chondrules in Brunflo were of olivine type can not be established due to the altered state of the meteorite, thus one cannot use the relative abundance of chondrule types for classification of Brunflo.

The chromite of recently fallen H and L chondrites differs in that the latter group has on average about 0.5 wt% higher TiO_2 content, a fact shown in different compilations for recently fallen meteorites (petrographic types 5 and 6) in Bunch et al. (1967), Schmitz et al. (2001) and Wlotzka (2005) (Table 2). Chromite TiO_2 concentration is thus a strong discriminant between these groups. However, it must be stressed that, although in different populations of L and H chondrites, the difference in average TiO_2 concentration is a very robust

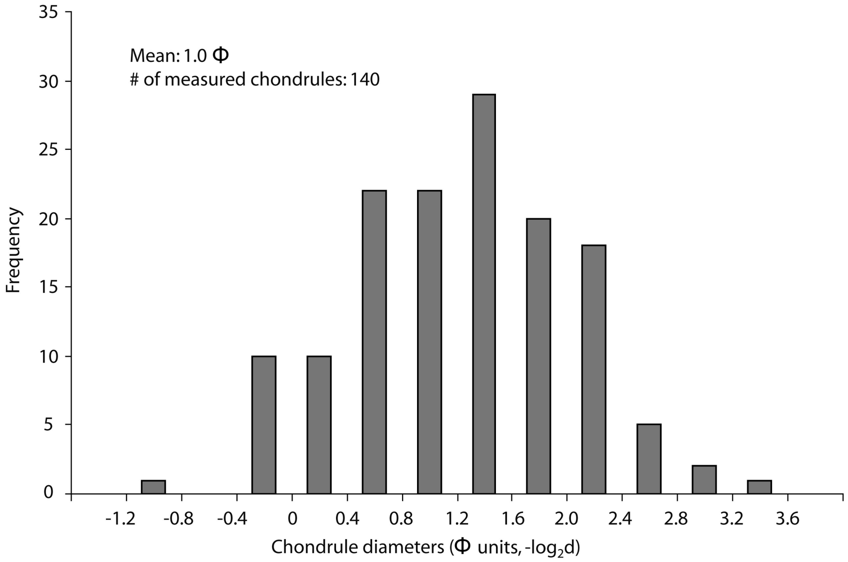

Fig. 4. Chondrule diameter size in phi-units plotted against frequency, showing a near log-normal distribution.

Table 4. Chromite measurements.

	Number of meteorites	Area (mm^2)	No. of grains >5 µm	D$_{max}$ µm	Mean D >5 µm	Grains >5 µm (mm^{-2})
Brunflo	1	130	146	130	37	1.1
Recent L3[1]	2	92	46	34–50	15	0.2–0.7
Recent L4[1]	3	225	292	87–150	22	0.2–4.3
Recent L5[1]	2	59	184	76–158	25	2.8–3.3
Recent L6[1]	2	64	217	253–638	34	2.8–5.6

[1] Bridges et al. (2007).

feature, the chromite composition of single meteorites from respective groups can overlap. For a single meteorite with low chromite TiO$_2$, the probability is higher that it is an H than an L chondrite, but it may still be an L chondrite. On the other hand, for a group of fossil meteorites with an average TiO$_2$ content identical to the average for recently fallen L chondrites, the bulk of the fossil meteorites must be L chondrites, as suggested for the Kinnekulle meteorites (Schmitz et al. 2001), and supported by oxygen isotope and chrondrule-size measurements (Bridges et al. 2007; Greenwood et al. 2007).

A more careful evaluation of the TiO$_2$ content of chromites from ordinary chondrites shows that there is a trend of increasing TiO$_2$ not only from H to L to LL, but also from type 3 to 4 to 5/6 (Fig. 6). In the compilations of Bunch et al. (1967), this trend is particularly pronounced for the L chondrites. Chromite from three L3 chondrites show TiO$_2$ in the range 0.25–0.6 wt%, and five L4 chondrites show values between 1.4 to 2.1 wt%, in marked contrast to eight L5/6 chondrites with 2.5 to 3.4 wt% TiO$_2$. Compilation of all available literature data for TiO$_2$ in chromite in L4 chondrites plus results for three new meteorites analysed for the present study are given in Table 6 and Fig. 6. We show that 11 of 13 L4 chondrites have TiO$_2$ values lower than 2.1 wt%, and most have values around 1.8 wt%, similar to chromite in the Brunflo meteorite. Only two give values similar to the L5/6 meteorites around 2.7 wt%. This is analogous to the results of Kimura et al. (2006) who showed that spinel group minerals in LL chondrites display a relation between TiO$_2$ content and petrographic type, particularly for the range of type 3 chondrites, a fact they interpreted as being the result of increased diffusion of TiO$_2$ into spinels from groundmass glass in chondrules and high Ca-pyroxene. This explanation is likely to be valid for the L chondrites as well. Although L4 chondrites are generally considered to be "equilibrated" meteorites there are subtle, gradational trends near the border between "unequilibrated" and "equilibrated" meteorites. For example, many type 4 meteorites have highly unequilibrated low-Ca pyroxene (Dodd et al. 1967). In summary, chondrule size measurements clearly show that Brunflo is an L chondrite, and the data on the D$_{max}$ and abundance of its chromite, the sharp definition of its chondrules and the information on the similar chromite TiO$_2$ concentration in recently fallen L4 chondrites, add support also for an L4 classification for Brunflo.

The distribution of sediment-dispersed extraterrestrial chromite grains at Kinnekulle (Schmitz and Häggström 2006)

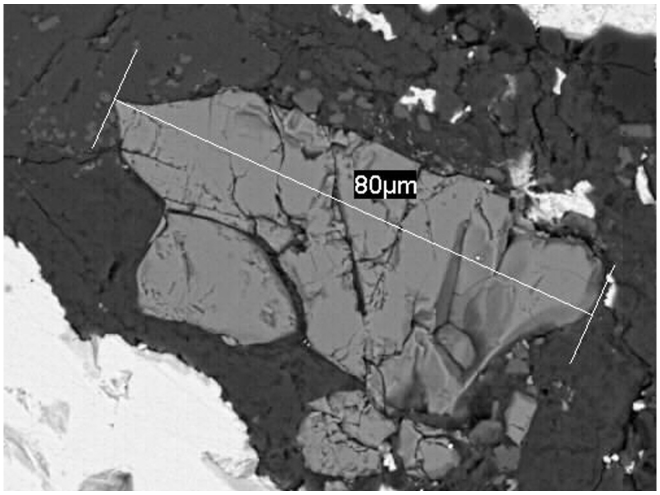

Fig. 5. Backscatter image of chromite grain in the Brunflo meteorite.

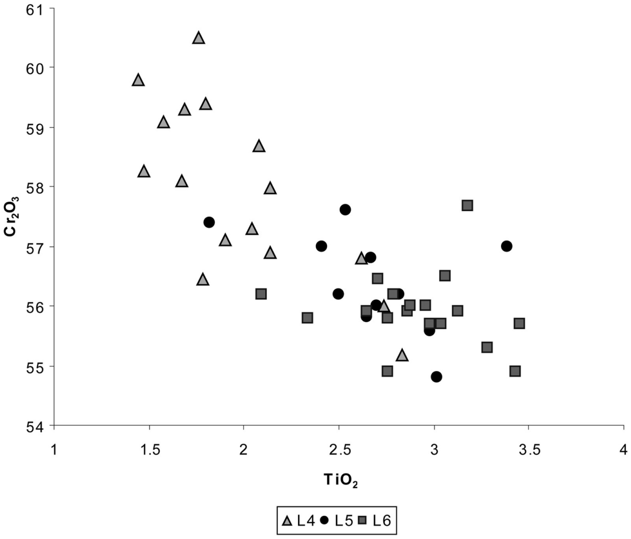

Fig. 6. TiO_2 versus Cr_2O_3 in chromite from L chondrites of petrographic types 4, 5, and 6. Data sources: Bunch et al. (1967); Schmitz et al. (2001); this study; Wlotzka (2005).

and cosmic ray exposure ages of chromite grains from the Kinnekulle meteorites (Heck et al. 2004) indicate that the disruption in the asteroid belt of the L chondrite parent body occurred shortly before the formation of the Arkeologen bed at Kinnekulle, close to the base of the *E. variabilis* Zone. Fossil meteorites and extraterrestrial chromite are abundant in most of the 3–4 meters of section directly overlying the base of the Arkeologen bed (Fig. 2). At 8.5 m above the basal Arkeologen bed, the content of extraterrestrial chromite has tailed off from initial values of typically 1–3 grains kg^{-1} to 0.4 grain kg^{-1}. The samples from Gärde, representing a time 1–2 million years after the highest sample at Kinnekulle, yield an average value of 0.45 grain kg^{-1}, consistent with the tail of the Kinnekulle trend. This value represents an order-of-magnitude enrichment in extraterrestrial chromite compared to the 0.01–0.03 grain kg^{-1}, in condensed limestone formed during the Ordovician before the asteroid breakup event or in similarly condensed pelagic limestone from the latest Cretaceous and Paleocene part of the famous Bottaccione Gorge section in Italy (Cronholm and Schmitz 2007). The results for the Gärde section agree with modelling simulations showing that an increase in the flux of extraterrestrial material to Earth may prevail for 5–30 million years after a major parent body breakup event in the asteroid belt (Gladman et al. 1997; Zappalà et al. 1998). The small decrease in EC content of the limestone over about 5 million years may reflect a slight tailing-off in the flux of micrometeorites to Earth, however, it is difficult to determine variations by a factor of a few in sedimentation rates, and uncertainty exists as to the significance of the small decline in EC content. Measurements of Ne isotopes in sediment-dispersed EC grains from the mid-Ordovician show high amounts of solar-wind implanted gases, indicating that most grains arrived on Earth as parts of unmelted micrometeorites (Heck et al. 2008).

The sediment-dispersed EC grains in the Gärde section have a mean TiO_2 content of 3.0 ± 0.3 wt%, which is similar to the mean TiO_2 content of chromite in recently fallen L5/6 meteorites, 2.7 ± 0.4 wt%, and in the abundant fossil meteorites, 2.7 ± 0.4 wt%, from Kinnekulle (Table 2). The Gärde sediment-dispersed EC grains are significantly higher in TiO_2 than chromite in recently fallen L4 chondrites, commonly <2.1 wt% TiO_2, the Brunflo meteorite, 1.9 wt%, and recently fallen H5/6 chondrites, 2.2 ± 0.2 wt% (Tables 2 and 6). This suggests that the sediment-dispersed EC grains in the Gärde quarry, just like the sediment-dispersed grains in the older limestone at Kinnekulle, originate predominantly from decomposed micrometeorites and meteorites of the L5/6 type. This supports a sustained rain of L chondritic material to Earth at the time the Brunflo meteorite fell. The higher average TiO_2 content of the Gärde sediment-dispersed EC compared with the Brunflo chromites suggests that micrometeorites and meteorites of the L4 type represented a comparatively small fraction of the total flux of chondritic matter to Earth, just like today. Among the recent falls, the L4 meteorites make up about 7% of all L chondrites (Grady 2000). The chromite grains of L3 chondrites are usually smaller than 63 µm (Bridges et al. 2007) and are not recovered in our approach.

CONCLUSIONS

The Orthoceratite Limestone in the mid-Ordovician Gärde quarry is rich (0.45 grain kg^{-1}) in sediment-dispersed extraterrestrial chromite grains (>63 µm). This chromite shows similar chemical composition as the extraterrestrial chromite in limestones and fossil L chondrites at Kinnekulle, suggesting that the enhanced flux of L chondrites prevailed at the time the Brunflo meteorite

Table 5. Element concentration (wt%) of sediment-dispersed extraterrestrial chromite grains (EC) and other chromite grains (OC) from the Gärde quarry. Each value is a mean of three analysis points of each individual grain.

m.	#	Cr_2O_3	Al_2O_3	MgO	TiO_2	V_2O_3	FeO_{tot}	FeO	Fe_2O_3	MnO	ZnO	Total	Fe#	Cr#
2.70 EC grains														
	1	55.15	5.80	1.68	3.93	0.74	31.25	31.25	0.00	0.78	0.26	99.59	91.3	86.4
	2	58.77	5.76	2.80	3.12	0.73	26.69	26.69	0.00	0.86	0.55	99.28	84.2	87.2
	3	57.33	5.55	2.28	3.19	0.68	28.06	28.06	0.00	1.05	0.53	98.67	87.4	87.4
	4	60.06	4.84	2.25	2.75	0.76	26.07	26.07	0.00	0.93	0.68	98.34	86.7	89.3
	5	57.09	6.10	1.02	2.82	0.72	28.57	28.57	0.00	0.55	2.99	99.86	94.0	86.3
	6	57.29	5.71	2.87	2.93	0.70	28.92	28.92	0.00	1.03	b.d.	99.45	85.0	87.1
	7	57.44	5.77	2.66	2.98	0.74	28.02	28.02	0.00	1.13	0.57	99.31	85.5	87.0
	8	57.58	5.66	2.42	2.99	0.70	29.79	29.79	0.00	0.92	0.36	100.42	87.4	87.2
	(9[1]	45.36	4.86	0.39	2.18	0.58	40.86	–	–	0.57	3.76	98.56	–	86.2)
	Mean	57.59	5.65	2.25	3.09	0.72	28.42	28.42	0.00	0.91	0.74	99.37	87.7	87.2
	Std dev. 1σ	1.41	0.36	0.62	0.37	0.03	1.65	1.65	0.00	0.18	0.93	0.65	3.3	0.9
	OC grains													
	1	34.92	26.38	6.51	b.d.	b.d.	30.88	25.45	5.43	0.48	b.d.	99.17	68.7	47.0
	2	52.09	11.19	7.62	b.d.	b.d.	27.54	21.08	6.46	0.75	b.d.	99.18	60.8	75.7
	3	50.70	12.94	5.28	b.d.	b.d.	29.41	24.70	4.71	0.55	0.61	99.49	72.4	72.4
	4	37.89	22.03	7.63	b.d.	b.d.	30.73	22.36	8.37	1.23	b.d.	99.52	62.2	53.6
5.62 EC grains														
	1	59.37	6.27	2.06	3.09	0.70	25.53	25.53	0.00	b.d.	1.31	98.33	87.4	86.4
	2	57.29	5.67	2.77	3.03	0.75	29.78	29.78	0.00	0.82	b.d.	100.11	85.8	87.1
	3	56.78	5.63	2.91	3.14	0.72	29.16	29.16	0.00	0.93	b.d.	99.27	84.9	87.1
	4	57.16	5.54	2.63	3.19	0.65	29.21	29.21	0.00	0.88	b.d.	99.26	86.2	87.4
	5	58.24	6.09	3.49	2.29	0.81	27.42	27.42	0.00	0.96	b.d.	99.30	81.5	86.5
	6	56.85	6.02	3.44	2.92	0.73	27.23	27.23	0.00	0.93	0.77	98.89	81.6	86.4
	7	56.52	6.34	2.81	2.82	0.87	28.60	28.60	0.00	0.83	0.63	99.42	85.1	85.7
	8	56.02	6.29	3.16	3.11	0.79	29.55	29.55	0.00	0.89	b.d.	99.81	84.0	85.7
	9	56.14	5.89	3.08	2.91	0.70	28.58	28.58	0.00	0.84	0.64	98.78	83.9	86.5
	10	56.79	6.01	3.08	3.05	0.61	28.32	28.32	0.00	0.87	0.63	99.36	83.8	86.4
	11	56.24	6.12	3.62	2.90	0.64	27.83	27.83	0.00	0.93	0.96	99.24	81.2	86.0
	12	56.75	5.99	4.21	3.04	0.71	27.99	27.99	0.00	0.70	b.d.	99.39	78.9	86.4
	13	57.06	5.89	2.65	2.91	0.78	28.49	28.49	0.00	0.79	b.d.	98.57	85.8	86.7
	14	57.07	6.10	2.58	2.76	0.53	29.67	29.67	0.00	b.d.	b.d.	98.71	86.6	86.3
	15	56.84	6.25	3.45	3.27	0.73	28.46	28.46	0.00	0.74	b.d.	99.74	82.2	85.9
	(16[1]	44.01	6.11	1.17	1.34	0.47	39.33	–	–	b.d.	1.85	94.28	–	82.8)
	(17[1]	34.13	6.87	2.33	0.52	b.d.	38.94	–	–	b.d.	5.63	88.42	–	76.9)
	Mean	57.01	6.01	3.06	2.96	0.71	28.39	28.39	0.00	0.74	0.33	99.21	83.9	86.4
	Std dev. 1σ	0.84	0.25	0.53	0.23	0.08	1.11	1.11	0.00	0.31	0.45	0.48	2.4	0.5
OC grains														
	1	45.37	20.89	10.72	b.d.	b.d.	20.22	18.04	2.18	b.d.	b.d.	97.20	48.6	59.3
	2	49.40	13.83	7.56	b.d.	b.d.	28.18	22.28	5.90	b.d.	b.d.	98.97	62.3	70.5
	3	53.20	10.49	4.11	0.70	0.51	29.46	27.15	2.31	0.60	b.d.	99.07	78.8	77.3
	(4[1]	36.77	28.00	9.17	b.d.	b.d.	23.86	–	–	b.d.	1.05	98.85	–	46.8)
	(5[1]	45.54	16.84	7.65	b.d.	b.d.	20.18	–	–	b.d.	1.23	91.44	–	64.5)

[1] Semi-quantitative analysis on unpolished grain, not included in the mean and standard deviation.

b.d: below detection limit; Fe#: mol% $Fe^{2+}/(Fe^{2+} + Mg)$; Cr#: mol% $Cr/(Cr + Al)$.

Table 6. Element concentration (wt%) of chromites from recent L chondrites of petrographic type 4.

Meteorite	Type	Cr_2O_3	Al_2O_3	MgO	TiO_2	V_2O_3	FeO	MnO	ZnO	Total	Fe#	Cr#
Atarra (Bunch et al. 1967)	L4	59.4	5.1	1.43	1.80	0.72	31.9	0.55	–	100.90	92.6	88.7
Atarra (This study)	L4	58.3	6.0	2.04	1.47	0.80	30.1	0.95	0.77	100.43	89.2	86.6
Awere (This study)	L4	57.1	6.3	1.98	1.90	0.68	30.8	0.50	–	99.28	89.7	85.9
Bald Mountain (Bunch et al. 1967)	L4	58.1	5.5	1.83	1.67	0.74	32.3	0.67	–	100.81	90.8	87.6
Bjurböle (Wlotzka 2005)	L4	56.9	5.7	2.18	2.14	0.67	30.7	0.65	0.33	99.29	88.8	87.0
DaG 323 (Wlotzka 2005)	L4	57.3	6.2	2.35	2.04	0.65	30.7	0.74	0.38	100.32	88.0	86.2
Floyd (This study)	L4	58.0	6.2	2.51	2.14	0.80	30.0	0.93	–	100.52	87.0	86.2
Goodland (Bunch et al. 1967)	L4	59.8	3.7	1.64	1.44	0.81	32.8	0.80	–	100.99	91.8	91.6
HaH 219 (Wlotzka 2005)	L4	60.5	4.5	5.08	1.76	0.64	26.7	0.66	0.15	99.94	74.7	90.1
Haxtun (Wlotzka 2005)	L4	59.3	4.9	3.49	1.69	0.63	28.4	0.69	0.23	99.29	82.0	89.1
McKinney (Bunch et al. 1967)	L4	58.7	4.0	2.55	2.08	0.70	32.4	0.55	–	100.98	87.7	90.8
McKinney (This study)	L4	56.5	6.8	3.03	1.78	0.74	28.4	1.19	–	98.42	84.0	84.7
NWA 767 (Wlotzka 2005)	L4	56.8	5.6	1.55	2.62	0.83	31.7	0.74	0.38	100.24	92.0	87.2
Saratov (Schmitz et al. 2001)	L4	59.1	3.9	1.75	1.58	0.80	30.3	0.93	0.53	98.90	90.7	91.1
Waltman (Schmitz et al. 2001)	L4	55.2	5.9	2.97	2.83	0.74	29.7	0.77	0.35	98.38	84.9	86.3
Waltman (This study)	L4	56.0	6.3	3.21	2.74	0.70	28.9	0.69	–	98.63	83.5	85.6

Fe#: mol% Fe/(Fe + Mg); Cr#: mol% Cr/(Cr + Al).

fell. The Brunflo meteorite is most likely a part of this increased flux of L chondritic matter. This is favored by the size measurement of chondrules, giving a mean diameter of 0.49 mm, which is in good agreement with data from recently fallen L chondrites. Furthermore, the D_{max} of relict chromite and the relatively sharp chondrule definition imply that Brunflo is of petrographic type 4 or possibly 5. The low TiO_2 (1.8 wt%) of the Brunflo chromites, previously used as the main argument for an H-chondritic origin, is also consistent with an origin from an L4 chondrite. Recently fallen L4 chondrites typically have chromite TiO_2 values very similar to that of the Brunflo chromite.

Acknowledgments–We thank A. Löfgren for enlightened discussions on conodont stratigraphy, D. Holtstam for help with sampling the Brunflo meteorite, J. O. Nyström for providing us with photos of Brunflo, M. Tassinari and J. Schmitz for assistance during sampling of the Gärde quarry, and M. Nolvi and N. Thorelli for help with picking chromite grains from the Gärde samples. Helpful reviews were provided by J. Grossman, J. Bridges and F. Wlotzka. This study was supported by the National Geographic Society, Swedish Research Council and Crafoord Foundation.

Editorial Handling—Dr. Christine Floss

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