



Invited review

Extraterrestrial spinels and the astronomical perspective on Earth's geological record and evolution of life

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ABSTRACT

Relict spinel grains ($\sim 25\text{--}250 \mu\text{m}$ in diameter) from decomposed extraterrestrial material in Archean to Recent sediments can be used to reconstruct variations in the flux of different types of meteorites to Earth through the ages. Meteorite falls are rare and meteorites weather and decay rapidly on the Earth surface, making it a challenge to reconstruct ancient fluxes. Almost all meteorite types, however, contain a small fraction of spinel minerals that survive weathering and can be recovered by acid-dissolution of large samples (100–1000 kg) of slowly deposited sediments of any age. The spinel grains originate from either micrometeorites, meteorites or asteroids, and can give detailed information on the types of extraterrestrial matter that fell on Earth at specific times in the geological past. Inside the spinels, synchrotron-light X-ray tomography can identify 1–30 μm inclusions of most of the other minerals that made up the original meteorite. With advanced microanalyses of the spinels, such as Ne isotopes (from solar wind, and produced by cosmic rays), oxygen isotopes (meteorite class and group) and cosmic ray tracks, it may be possible to unravel from the geological record fundamental new information about the solar system at specific times through the past ~ 3.5 Gyr. Variations in flux and types of meteorites may reflect large-scale perturbations of the orbits of planets and other bodies in the solar system, as well as the sequence of disruptions of the parent bodies for the meteorite types known and not yet known. Orbital perturbations may be triggered by near-by passing stars, giant molecular clouds, the galactic gravitational field, supernova shock waves or unusual planetary alignments.

The spinel approach has so far been primarily applied to the middle Ordovician Period. In sediments of this age the breakup of the L chondrite parent body at ~ 466 Myr ago manifests itself by a two orders of magnitude increase in L chondrite material. A total of 99 fossil meteorites (1–21 cm in diameter), of which all or almost all are L chondrites, have been found in a small quarry in marine limestone of mid-Ordovician age in southern Sweden. The identification of the meteorites as L chondrites relies primarily on chemical and isotopic analyses of relict spinel (chromite). In addition, coeval slowly formed marine limestone from Sweden, China, and Russia is extremely rich in chromite grains ($>63 \mu\text{m}$) with L chondrite composition. Based on a high content of solar wind Ne these spinels are interpreted as originating primarily from micrometeorites. Typically 1–10 grains are found per kg of rock, compared to background concentrations of 1–3 grains per 100 kg of similarly slowly deposited rock from other time periods. The elevated flux of L chondrite material to Earth in the mid-Ordovician coincides with important biotic changes, known as the Great Ordovician Biodiversification Event, as well as global volcanism and tectonic reorganizations. This indicates a possible primary or secondary connection between astronomical and terrestrial perturbations. Further evidence for a relation between perturbations of the asteroid belt and the Earth comes from a more general, long-term correlation of common breakup events in the asteroid belt, and repeated major ice ages as well as environmentally driven biotic change on Earth. In essence, with the spinel approach described here it will be possible to systematically, in great detail and on a strictly empirical basis, relate major events in the larger astronomical realm to the sequence of biotic, tectonic and climatic events on Earth. A pioneer astrostratigraphy can be established for Earth's geological record, complementing existing bio-, chemo-, and magnetostratigraphies.

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1. Introduction

The tradition is that astronomers look up at the sky and geologists look down at Earth. Here, a new approach is discussed that can relate events in the skies to events on Earth during the past ~3.5 Gyr, by looking down deep into Earth's sedimentary record. Classical geologists have a tradition to look on Earth as a more or less closed system. It took them a long time to accept that many of Earth's craters are related to impacts of extraterrestrial objects rather than volcanism (Grieve and Stöffler, 2012). Only recently has it been accepted that astronomical events can play a crucial role in evolution of life on Earth (Alvarez et al., 1980; Alvarez, 2003). Here, I discuss a feasible way to reconstruct the detailed variations in the flux of meteorites and micrometeorites to Earth during most of its history. This approach has the potential to tie the paleo-astronomical realm to the paleo-geobiosphere of Earth through the ages. Conventionally, it has been believed that it would not be possible to reconstruct paleo-variations in the flux of different types of meteorites. Meteorite falls are rare and meteorites and micrometeorites weather and decay rapidly on the Earth surface. A change in our understanding of this came with the realization from studies of rare fossil ~462–466 Myr old meteorites, that most of the spinel group minerals in meteorites are extremely resistant to weathering in terrestrial sedimentary environments (Thorslund et al., 1984; Nyström et al., 1988), and can be recovered from slowly formed sediments of almost any age over the past 3.5 Gyr (Schmitz et al., 2003; Schmitz and Häggström, 2006; Cronholm and Schmitz, 2007, 2010). Once recovered the spinel grains can give a plethora of information of the original host meteorite or micrometeorite. Methods have been developed for determining the class, group and even petrologic type of the meteorite from which a sand-sized, sediment-dispersed extraterrestrial spinel grain originated (e.g., Schmitz et al., 2001, 2003; Alwmark and Schmitz, 2009a; Heck et al., 2010). Noble-gas measurements of such grains show that they contain a record of the meteorite's cosmic ray exposure age, exposure of solar wind during transport to Earth and regolith-derived gas signals acquired on asteroid parent bodies (Heck et al., 2004, 2008; Meier et al., 2010). In some spinel types and in silicate inclusions in spinels, cosmic ray tracks can be reconstructed, tracks that were acquired in space at a specific time in the history of the solar system, and preserved in an ancient sediment on Earth (Riebe, 2012).

Most of the development of methods in the spinel approach relies so far on studies of marine sediments that formed in the mid-Ordovician, shortly after the breakup of the L chondrite

parent body in the asteroid belt ~466 Myr ago (Schmitz et al., 2001, 2003, 2008). The approach, however, can be applied to any time in Earth's history for which there are slowly accumulated sediments available. Changes in the meteorite and micrometeorite flux, which may have a number of astronomical explanations, can be tied in detail, bed-by-bed in the geological strata, to the evolution of Earth's life and the record of, for example, volcanism, climate and sea-level. It is obvious that the atmosphere has a significant effect on Earth in the shorter perspective, like day, month, year, solar 11-year cycle and the 21–400 Kyr Milankovitch cycles, but in the longer perspective our understanding of any causal relationships is weak. Studies of the meteorite flux to Earth in the distant past will also add to our understanding of meteorites and asteroids of today's world. Did the same types of meteorites fall on Earth in about the same proportions as today? Or, do present meteorites only reflect a small fraction of the types of asteroid parent bodies that can deliver meteorites to Earth? How do large asteroid breakup events affect the meteorite flux to Earth? Can the material that reaches Earth from such events at different times after the breakup tell us something about how the largest asteroids are built up? These and many other new questions in meteoritics can be addressed empirically by searches for extraterrestrial spinels in ancient sediments.

More general questions are, what can variations in the meteorite flux tell us about astronomical events at a solar-system or even galactic scale? Did these events have any recognizable effects on Earth's environment and fauna? A few visionaries early realized that the evolution of animals on Earth could be affected by cosmic events. For example, Nininger (1942) suggested that faunal cataclysms in Earth's history could be related to impacts of large extraterrestrial bodies. Schindewolf (1954, 1963) argued that evolution is not gradual in the way Darwin proposed, but characterized by a series of coupled mass extinction and diversification events. He invoked gamma-ray bursts from exploding supernovae, triggering extinctions but also the mutations necessary for the following rapid faunal diversification. The idea that dinosaurs or other major groups of organisms were eradicated from Earth's surface because of asteroid or comet impacts was proposed repeatedly during the 20th century (De Laubenfels, 1956; McLaren, 1970; Urey, 1973), but these early claims, like numerous other extinction scenarios, were not based on any supporting physical evidence. A breakthrough came when Alvarez et al. (1980) showed the existence of a global iridium anomaly at the Cretaceous-Tertiary (K-T) boundary which they interpreted to reflect the impact of a major extraterrestrial body causing one of the largest species mass extinction

events in Earth history. This theory has now withstood thirty years of intense testing (Schulte et al., 2010). A 10 km-sized impactor hit the Yucatan peninsula 66 Myr ago forming the ~150–200 km large Chicxulub crater. The event eradicated the dinosaurs after their ~165 Myr successful existence on this planet (Lyson et al., 2011), but also seriously affected most groups of common marine invertebrates.

Many suggestions have been published the past decades on how evolution of life and astronomical events could be intertwined. For example, an observed 30 Myr cyclicity in life's extinction pattern has been related to the regular passage of the solar system at about this frequency through the galactic equatorial plane (Rampino and Stothers, 1984; Rampino and Haggerty, 1996). In the galactic plane, which has a higher concentration of interstellar dust, additional external gravitational forces perturb the orbits of solar system objects and catapult comets from the outer to the inner solar system, with enhanced risk of impacts and environmental perturbations on Earth. Until today, however, no other extinction event than that at the K-T boundary has been convincingly tied to an asteroid or comet impact (Alvarez, 2003; Racki, 2012). Recently, however, it has been shown that the onset of the main phase of the Great Ordovician Biodiversification Event in the Middle Ordovician coincides with the breakup of the L chondrite parent body, the largest documented breakup event in the asteroid belt the last ~3 Gyr (Schmitz et al., 2008). Frequent impacts on Earth of kilometer-sized asteroids from this event could have spurred evolution in accordance with the so called intermediate disturbance hypothesis, initially applied to diversity changes in coral reefs and tropical rain forests.

The accretion of extraterrestrial matter through Earth's history is a new cross-disciplinary research field (see Peucker-Ehrenbrink and Schmitz, 2001). The astrosphere can be tied to the geobiosphere by detailed searches for extraterrestrial signatures in Earth's sedimentary strata. This can be combined with studies of the astronomical skies, creating a true "astrogeobiosphere" perspective. Nesvorný et al. (2002, 2007, 2009) have used recently acquired information about abundant mid-Ordovician fossil L chondrites (see Section 2) in combination with astronomical data and simulations to try to trace the origin of L chondrites to the asteroid family that represents the residual signature in the asteroid belt of the exploded L chondrite parent body. Likely candidates are the Flora and the Gefion asteroid families. Likewise, based on dynamical simulations the Baptistina asteroid family was suggested to have broken up ~160 Myr ago, an event supposedly ejecting the asteroids that formed the Tycho crater on the Moon 109 Myr ago and the Chicxulub crater on Earth 66 Myr ago (Bottke et al., 2007). However, spectral data for the main Baptistina family asteroid rule out a relation to the K-T boundary impactor and suggest a rather young age, 80 Myr ago, for the breakup event (Masiero et al., 2011; Reddy et al., 2011).

Reconstructing astronomical history from extraterrestrial signatures in the sedimentary record mostly suffers from the "needle-in-the-haystack" problem. In a normal marine setting, like on the middle shelf, sedimentation rates are typically 10 cm per Kyr, so a complete record of the Earth's Phanerozoic Eon (the last 541 Myr) in this environment, would be represented by a 54 km thick sequence of strata. In the decades after the discovery of the iridium anomaly at the K-T boundary (Alvarez et al., 1980), a number of tools have been developed to systematically search for signatures of asteroid or comet impacts in the sedimentary record. The most commonly used proxies are osmium isotopic and iridium concentration anomalies (Paquay et al., 2008; Miller et al., 2010; Ravizza and VonderHaar, 2012). Chromium isotope anomalies (Kyte et al., 2011), impact spherule-beds (Glass and Simonson, 2012; Krull-Davatzes et al., 2012), shocked quartz (Bron and Gostin, 2012), and Ni-rich spinels formed on Earth in impact vapor clouds

(Robin and Molina, 2006), are other proxies used in this research. Studies of Earth's cratering record, with presently ~180 confirmed impact craters (Grieve, 2001; Reimold and Jourdan, 2012) also adds to describing the intertwined history of life, Earth and the astronomical realm (Fig. 1). Fewer studies have provided robust data on variations in the flux to Earth of extraterrestrial debris in the meteorite to dust size fractions. Most notably, ³He has been used for tracing variations in the flux of interplanetary dust particles (Farley et al., 2012). Recovery of fossil meteorites in Ordovician limestone (Schmitz et al., 2001) is the so far only method to reconstruct meteorite fluxes. In the following, focus will be on the new extraterrestrial spinel proxy, that while requiring tedious sediment processing, may add significantly to the understanding of the evolution of the astrogeobiosphere.

2. Mid-Ordovician extraterrestrial spinels and the breakup of the L chondrite parent body

2.1. Mid-Ordovician fossil meteorites

The spinel approach has its roots in a chance discovery of a fossil meteorite in middle Ordovician, ~462 Myr old marine limestone from the Brunflo quarry in central Sweden (Fig. 2). In 1952 the manager of the Brunflo quarry handed over to Per Thorslund, a paleontology professor at Uppsala University, a polished limestone plate with a dark ~10 cm clast in it. Thorslund was intrigued by the plate and asked a petrographer for advice, who identified the clast as a pseudomorph after some kind of terrestrial ultramafic rock. Thorslund concluded that the clast probably had become attached to algae at the shores of the large epicontinental sea that covered Baltoscandia during the Ordovician. The stone had drifted to the deeper, sediment-starved parts of the sea, where it sank to the sea floor and became embedded in the sediments. The plate was stowed away in Thorslund's office for 27 years. In the 70s mineralogy professor Franz-Erik Wickman was pursuing pioneering research identifying astrobromes in the Swedish bedrock, such as the Siljan crater in central Sweden. Wickman was introduced to the clast-bearing limestone plate in 1979 and realized that the clast was a fossil meteorite, the Brunflo meteorite. The meteorite shows original chondrule textures, but is completely replaced by diagenetic minerals, except for relict spinel in the form of chromite. The composition of this chromite, and earlier detailed work on the chemistry of the chromite phase of recent meteorite finds and falls by the group of Klaus Keil, allowed the identification of Brunflo as a fossil ordinary chondrite (Thorslund and Wickman, 1981; Thorslund et al., 1984; and chromite work: Keil, 1962; Bunch et al., 1967; Snetsinger et al., 1967). Based on the chromite composition Brunflo was assumed to belong to the H chondrite group, but see Section 2.2 on recent reclassification to the L chondrite group.

Relatively soon after the publication of the Brunflo find a second mid-Ordovician fossil meteorite, Österplana 001, was found on a dump pile in the Thorsberg quarry in southern Sweden (Nyström et al., 1988) (Figs. 2, 3). The meteorite is altered to phyllosilicates, calcite and barite but contains abundant relict chromite with ordinary chondritic composition. In the Thorsberg quarry, limestone ~4–5 Myr older than in the Brunflo quarry is quarried for the production of mainly floor plates, window sills and other building stone. A local amateur geologist and fossil collector, Mario Tassinari, read about the discovery in the local news paper, contacted the quarry workers and asked them to lay aside any further meteorite-resembling objects instead of throwing them away. This was the basis in 1992 for a scientifically controlled, systematic search project for fossil meteorites on the mid-Ordovician sea floor (Figs. 4–10). By 1996 thirteen meteorites, all L (or LL) chondrites, had been found in the Thorsberg quarry, and it became obvious

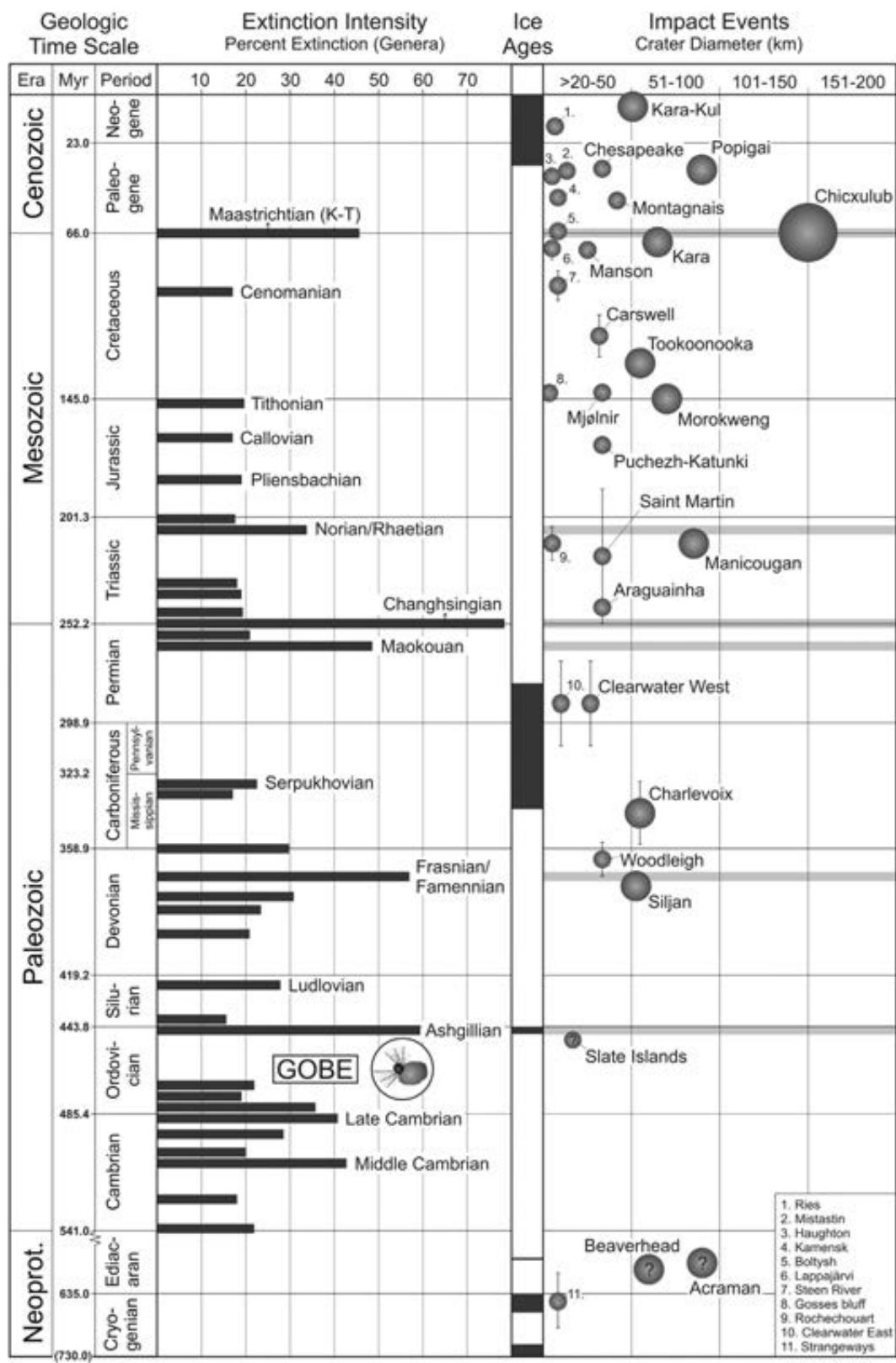


Fig. 1. Mass extinctions, ice ages and impact craters during the past 730 Myr. Stratigraphic subdivisions and numerical ages from the 2012 Geological Time Scale (Gradstein et al., 2012). The extinction events and their intensities are from a compilation in Keller (2005). Only extinction events with a higher than 15% intensity at the genus level are presented. Ages and sizes of impact craters larger than 20 km follows the Earth Impact Database, early 2013 (<http://www.unb.ca/pasc/ImpactDatabase/>). The approximate timing of the Great Ordovician Biodiversification Event (GOBE) and the breakup of the L chondrite parent body in the asteroid belt 466 Myr ago are also shown. The illustration is only intended to give a first-order overview, and many details can be debated.

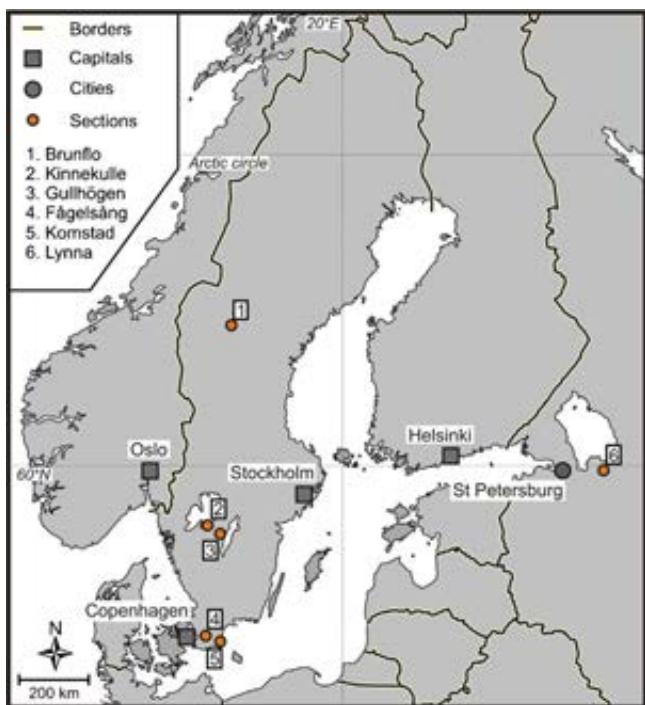


Fig. 2. Location of quarries and sections where fossil meteorites and micrometeorites have been found in Sweden and western Russia: Brunflo quarry in central Sweden; Kinnekulle with the Thorsberg and Hällekis quarries (see Fig. 3) and Gullhögen quarry, southern Sweden; Komstad quarry and Fågelsång section, southernmost Sweden; and Lynna River section near St. Petersburg, Russia.

that there were “too many” meteorites on the ancient sea floor compared to the expected abundances based on our understanding of the meteorite flux in today’s world (Schmitz et al., 1996, 1997). Literature studies soon gave a hint of a possible explanation. With the advances in the early 60s in U–He and K–Ar dating of recently fallen meteorites, it became apparent, that although most meteorites have gas retention ages going back to ~4 Gyr,

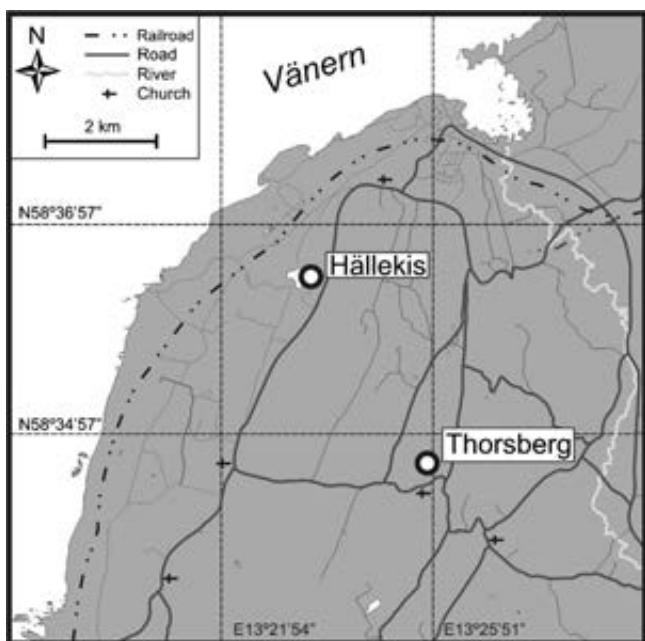


Fig. 3. Location of Hällekis and Thorsberg quarries at Kinnekulle, Västergötland, southern Sweden.



Fig. 4. The active Thorsberg quarry where 99 fossil meteorites have been found. The exposed section starts with the Botten bed, whereas the Golvsten (i.e. “floor stone”) and Arkeologen beds are under the floor surface of the quarry, see further Fig. 10.

there exists a large group of L chondrites with shock features and young gas retention ages, around 400–500 Myr (Kirsten et al., 1963; Anders, 1964; Hintenberger et al., 1964; Keil, 1964; Heymann, 1967). This had been attributed to a major asteroid disruption event, the breakup of the L chondrite parent body around 500 Myr ago (Anders, 1964; Bogard, 1995; Keil et al., 1995; Haack et al., 1996). Nowhere in the quite extensive literature had the suggestion been made that traces of this dramatic event possibly could be found in Earth’s geological record.

Both the Brunflo and the Österplana fossil meteorites have been recovered from so called Orthoceratite Limestone, that has a long quarrying history at several places in Sweden. This type of limestone formed over several hundred thousand km² of the Baltoscandian Shield during a ~20 Myr period in the late Early and Middle Ordovician, when a large epicontinental sea covered the area (Lindström, 1971; Jaanusson, 1972; Schmitz et al., 1996; Lindström et al., 2000). Distances to the shores at the meteorite-yielding localities in Sweden were large, and the limestone formed very slowly, at a rate of a few mm per Kyr. This is similar to deep-sea clays in the central Pacific Ocean today, however, water depths at meteorite-yielding localities on the Baltoscandian Shield were only on the order of 100–300 m (Chen and Lindström, 1991; Schmitz et al., 1996). The limestone is relatively pure with 80–90% calcite and is arranged in a series of massive beds mostly 2–20 cm thick, separated by iron-stained or marly surfaces, interpreted as submarine firm- or hard-grounds or very slowly deposited sediment layers (Lindström, 1962, 1979). Sediment deposition at the sea floor



Fig. 5. The systematic search since 1992 for fossil meteorites in the Thorsberg quarry was performed with the three quarry owners and brothers Göran Thor (upper photo), Sören Thor and Stig Thor (left and right in lower photo) and the local amateur geologist Mario Tassinari (middle of lower photo). In the upper photo Göran Thor shows the level for the find of a fossil meteorite in the Sextummen bed.

was intermittent, with short pulses of sedimentation during, for example, storm events followed by long periods, perhaps 0.1–1 Kyr, with very little deposition, and sea-floor dissolution and hard-ground formation. The abundant cone-shaped, elongated cephalopod shells in the limestone do not show any preferred orientation, indicating a tranquil sea-floor environment with no strong bottom currents; however, there may have been currents higher up in the water column preventing fine-grained material from settling.

2.2. Mid-Ordovician fossil meteorites by 2012

Twenty years (as of December 2012) after the initiation of the search project for fossil meteorites, 99 meteorites (1–21 cm in

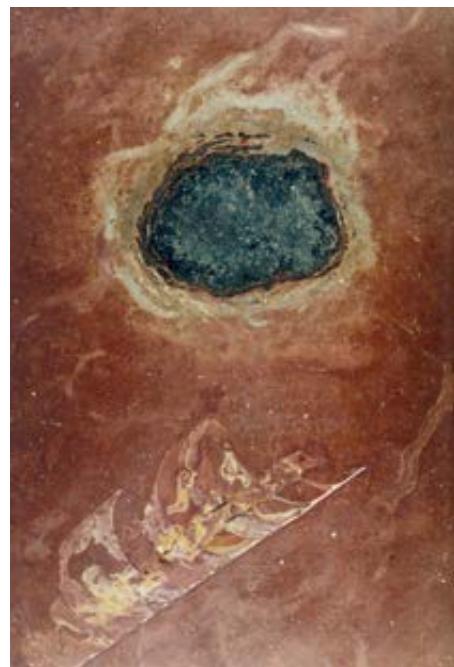


Fig. 7. The fossil meteorite Österplana 035 (informal name: Sex 001), that was found in the Sextummen bed in 1996. The meteorite is 8 × 6 cm in cross section. This is one of the meteorites with best preserved petrological textures, such as chondrules. The outer parts of the meteorite have been peeling off because of weathering on the sea floor. Similar weathering patterns can be seen in recent desert meteorites. The meteorite is surrounded by a grey worm track. In the lower part of the plate a piece of a fossil nautiloid shell is seen.

diameter) have been recovered in the Thorsberg quarry (Figs. 4–10). Although the meteorites can be readily identified by petrological criteria with trained eyes, the main criterion to identify a meteorite is by their content of relict spinels, almost exclusively chromite. All other common meteoritic minerals have been replaced by mainly calcite and clay minerals, but petrological textures are commonly preserved. All of the recovered meteorites have yielded chromite with the very characteristic ordinary chondrite elemental composition. Chromites of L and LL chondrites have too similar elemental compositions to be distinguished, whereas H and L chromite grains show markedly different average compositions, particularly for TiO₂, but some overlap occurs (see Section 2.4). From each fossil meteorite, several chromite grains are analyzed in order to obtain a good average elemental composition. Based on this approach, all or almost all fossil meteorites found by 2012 are L (or LL) chondrites



Fig. 6. The fossil meteorite Österplana 060 (informal name: Gla 2:004) still in situ in the quarry. The meteorite was found in the Glaskarten 2 bed in June 2009. The observers are Simonetta Monechi and Alan Hildebrand.

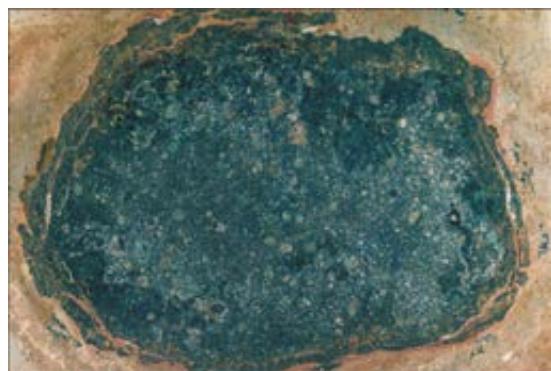


Fig. 8. Close up of Österplana 035. Note chondrule texture and a cm-sized clast in upper right part. Lower photo shows the chondrule texture. Scale at bottom is in mm.



Fig. 9. Upper photo: Göran Thor liberates fossil meteorite Österplana 032 (informal name: Bot 003) from the quarry wall in September 2000. Lower photo: The meteorite is the largest found and measures $21 \times 6.5 \times 4$ cm. Note the profound hard-ground surface a few cm below the base of the meteorite, but the meteorite has landed on soft sediment and sunken into it.

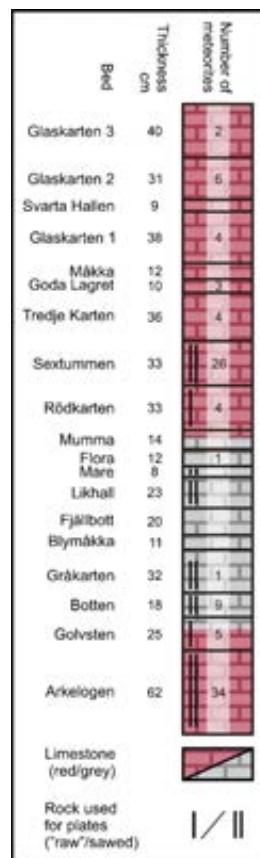


Fig. 10. Distribution of the 99 fossil meteorites found by September 2012 in the Thorsberg quarry. The division of the column in different beds is based on traditional use by quarry workers. Some of the names may be at least hundred years old.

(Schmitz et al., 2001). Average chondrule diameters for recent L and LL chondrite finds and falls are significantly larger than for H chondrites (~0.5, 0.6, and 0.3 mm, respectively). Chondrule sizes measured in six fossil meteorites by Bridges et al. (2007) indicate they are L chondrites. Most importantly, an L chondrite origin has been confirmed for some of the meteorites by analyses of chromite oxygen isotopes as well as elemental analyses of silicate inclusions in the chromite grains (see Sections 2.5 and 2.6).

The meteorites have been found throughout a limestone succession 4.7 m thick over a total area of less than 20,000 m² of the mid-Ordovician sea floor (Figs. 4–10). The quarried limestone sequence represents somewhere between 1.3 and 2.5 Myr of deposition. The limestone is tectonically undisturbed, with well-developed horizontal beds that can be traced for many hundreds of meters in the quarry. The systematic quarrying of these beds provides a near ideal setting for a detailed reconstruction of the distribution of meteorites on the ancient sea floor. However, only a fraction of the rock record over this area has been used for production of sawed plates and thus efficiently searched for fossil meteorites. The section is divided by the quarry workers into 19 beds, each with its characteristic lithological and sedimentological features and with different values for commercial purposes (Fig. 10). Meteorite recovery in a respective bed is related to the extent to which a particular bed is used for production of sawed slabs. This has to be considered in estimates of the paleoflux of meteorites (Schmitz et al., 2001). There is no doubt that meteorites from different beds and different hard-ground surfaces within a particular bed represent different falls. Meteorites from the same hard-ground surface or bed, but of different petrologic types, may also represent different falls. It is impossible that meteorites, for

example, from one major strewn field would have drifted on the sea floor for 1–2 Myr, and then became fixed at different levels throughout the section. Meteorites decompose, geologically speaking, rapidly (20–30 Kyr) in terrestrial environments (Bevan et al., 1998; Bland, 2001). There is no indication of a tailing off in the meteorite abundance upwards through the section. Meteorites in the upper beds can have as angular shapes as meteorites further down in the section. Bridges et al. (2007), using maximum chromite diameter as proxy, showed that there are different petrologic types among the fossil meteorites, ruling out an origin from a single or only a few fall events. Also the consistently different cosmic ray exposure ages of chromite from meteorites at different levels in the section indicate that many different falls are recorded (see Section 2.7). There is presently no way of distinguishing if meteorites of the same petrologic type on the same hard-ground surface represent fragments of the same fall or different falls. Based on this reasoning, a detailed study of the stratigraphic positions of the 40 meteorites that had been found by 2000 concluded that they represented at least 12 different falls (Schmitz et al., 2001). By that time ~6000 m² of the mid-Ordovician sea floor had been quarried. With the help of the quarry log book, with a detailed account of the total area of rock surface exposed through sawing, it could be confidently concluded that meteorites are *at least* one to two orders of magnitudes more abundant on the ancient sea floor than if the meteorite flux would have been the same as today. Recent meteorite searches in wet and cold desert areas or meteor sky-watch camera networks indicate that statistically on the order of one >10 g meteorite falls per 10,000 km² per year (Halliday et al., 1989; Huss, 1991; Bevan et al., 1998; Bland, 2001). With such a flux in the mid-Ordovician not even one meteorite would have been found in the rock pile quarried by 2000 at Thorsberg. A similar flux estimate is planned including also the 59 meteorites found after 2000, but the results will not differ significantly from those previously obtained. The quarrying of the sea floor has proceeded with about the same pace throughout the past twenty years and has typically yielded 4–6 meteorites per year, indicating that meteorites are relatively evenly distributed over the sea floor. In the past years 15 new meteorites have been found also in the 1.4 m interval immediately above the beds that had yielded meteorites by 2000 (Fig. 10). These are the youngest beds in the section and are of low quality for commercial purposes. They are basically just removed from the quarry in order to obtain access to the high-quality limestone underneath. The many meteorites from this interval are pure chance finds and attest to a very high content of meteorites in these strata as well.

One fossil meteorite has also been found in the Gullhögen quarry, 35 km to the southeast of the Thorsberg quarry (Tassinari et al., 2004) (Fig. 2). In this quarry, limestone is only quarried to be crushed for e.g. road fill, and systematic searches for meteorites cannot be performed. However, in a limestone block that had fallen off a truck, Mario Tassinari located a meteorite, Gullhögen 001, one cm in diameter, with chromite having the typical L chondrite composition. According to conodont analyses the block originates from the same stratigraphic level as the middle to upper part of the meteorite-yielding interval at Thorsberg. This chance find attests to the fact that meteorites are common in mid-Ordovician rocks outside of the Thorsberg quarry as well.

The Brunflo meteorite, from 600 km to the north of Thorsberg, was long an enigma. Having been tentatively classified as an H chondrite based, among other properties, on the lower TiO₂ values (1.9 wt%) in its chromite compared to equilibrated L chondrites (TiO₂ = 2.7 wt%) (Thorslund et al., 1984), it has been speculated that it could represent a remnant of the projectile body that impacted and broke up the L chondrite parent body. However, critical re-examination by Alwmark and Schmitz (2009b) shows that the low TiO₂ can be explained by the meteorite being somewhere in the L4 range between unequilibrated and equilibrated meteorites

(Grossman et al., 2009), where also L chondrite chromite may have low TiO₂ values (Alwmark and Schmitz, 2009b). Chondrule size studies, oxygen isotopes in the chromites, and chemistry of inclusions in the chromite show that Brunflo without doubt is an L4 chondrite. It thus attests to a high flux of L chondrites also ~4–5 Myr after the meteorite-rich sediments in the Thorsberg quarry formed (Alwmark and Schmitz, 2009a,b; Heck et al., 2010).

The quarry at Thorsberg provides a unique window into the flux of meteorites to Earth immediately after the largest documented parent body breakup in the asteroid belt during the past 2–3 Gyr. Thanks to the abundant and excellently preserved relict spinel grains in the fossil meteorites, it is possible to reconstruct even detailed aspects of the ancient meteorite flux.

2.3. Mid-Ordovician sediment-dispersed extraterrestrial spinel grains

The search for fossil meteorites in sediments is a tedious and slow process, and requires cooperation with an active quarry using the right industrial methods. In order to test the hypothesis that the high abundance of fossil L chondrites found in the Thorsberg quarry indeed represents an enhanced flux of L chondrite material to the entire Earth we developed an alternative approach to reconstruct the meteorite flux. It has been built on the assumption that most micrometeorites and meteorites that fell on the ancient Ordovician sea floor were not preserved as recognizable fossil meteorites, but instead decomposed and disintegrated. The resistant spinel grains were liberated and dispersed on the sea floor and later incorporated in limestone as the soft sea-floor sediments lithified. By dissolving large samples (from a few kilograms to tons) of slowly deposited limestone in hydrochloric (HCl) and hydrofluoric (HF) acid, these very resistant extraterrestrial (ordinary chondritic) chromite (EC) grains (~63–250 µm in diameter) can be recovered. With this approach we have located in mid-Ordovician condensed (=very slowly formed) sediment sections worldwide, a consistent stratigraphic level in the strata, at or close to the base of the *Lenodus variabilis* Conodont Zone, below which meteoritic EC grains are exceedingly rare, 1–2 grains per 100 kg of rock, whereas above this level concentrations lie typically at 1–10 L chondrite grains per kg (Figs. 11–13). The concentrations remain this high upward over a stratigraphic interval representing at least 2 Myr. This pattern has been reproduced in condensed limestone sections at sites in Sweden, western Russia and central China (Schmitz et al., 2003; Schmitz and Häggström, 2006; Cronholm and Schmitz, 2010; Lindskog et al., 2012). The average element and oxygen isotope compositions of the abundant chromite grains show that they are dominantly (or always) of L chondrite origin. High concentrations of solar wind implanted Ne in most of the grains indicate that a major fraction originate from decomposed micrometeorites (Heck et al., 2008, 2010; Meier et al., 2010).

In a composite section for the Thorsberg quarry and the abandoned, but vertically more extended Hällekis quarry 4 km to the northwest (Fig. 3), a clear two orders of magnitude change in the abundance of EC grains is recorded, beginning at the base of the *L. variabilis* Zone (Fig. 13). From this level and upwards ~3 m through the section we found 1–3 EC grains per kg of rock in each of 10 samples studied (Schmitz and Häggström, 2006). In the 9 m interval below the base of the *L. variabilis* Zone only 5 EC grains were found in a total of 379 kg of rock from many different beds in the interval. The dramatic change in EC concentration cannot be explained by a change in sedimentation rates. The section is made up of the same type of condensed Orthoceratite Limestone throughout, and conodont and trilobite zones show no evidence of being significantly more condensed or expanded in any part of the section studied. We interpret the level in the strata where the L chondrite chromite begins to be common as representing the time for the breakup of



Fig. 11. The more extended Hällekis section, four km northwest of the Thorsberg quarry. From this section a profile of the distribution of sediment-dispersed extraterrestrial chromite (EC) grains has been established, see Fig. 13. Below the yellow line EC concentrations are 1–2 grains per 100 kg, above the line, 1–3 grains per kg. The beds at the yellow line formed when the L chondrite parent body broke up in the asteroid belt. Asteroid breakup art work by Don Davis.

the L chondrite parent body in the asteroid belt (Fig. 11). This is also supported by cosmic ray exposure ages of the fossil meteorites as measured in their relict chromite (Heck et al., 2004). The section in the active Thorsberg quarry begins stratigraphically about one meter above the first EC-rich level in the Hällekis section. The young (0.05–0.2 Myr) cosmic ray exposure ages of the oldest meteorites and the gradual increase in ages upwards through the section support an origin of all the fossil meteorites from a breakup event when the first EC-rich beds formed (see further Section 2.7).

A similar distribution of EC grains through condensed mid-Ordovician Orthoceratite Limestone has been found in the combined Fågelsång-Komstad quarries section in the southernmost part of Sweden, 350 km south of the Thorsberg quarry

(Fig. 13). In this part of Sweden the paleobasin was deeper, and conditions were anoxic on the sea floor, resulting in the formation of black, organic-rich Orthoceratite Limestone. Here the concentrations of EC change from 2 grains per 125 kg of rock sampled over ~8 m of section to 2–6 grains per kg in the overlying 2 m (Häggström and Schmitz, 2007). Within the uncertainty of about 1–2 m in the biostratigraphic correlations, one can say that the change in EC abundance at Fågelsång-Komstad takes place at the same stratigraphic level as in the Hällekis-Thorsberg section. Also, single limestone samples from just above the base of the *L. variabilis* Zone in the Siljan area and Öland, ~300 km north and southeast of Thorsberg, respectively, yield about 2 EC grains per kg rock (Schmitz et al., 2003). Thus, the mid-Ordovician two-orders of magnitude

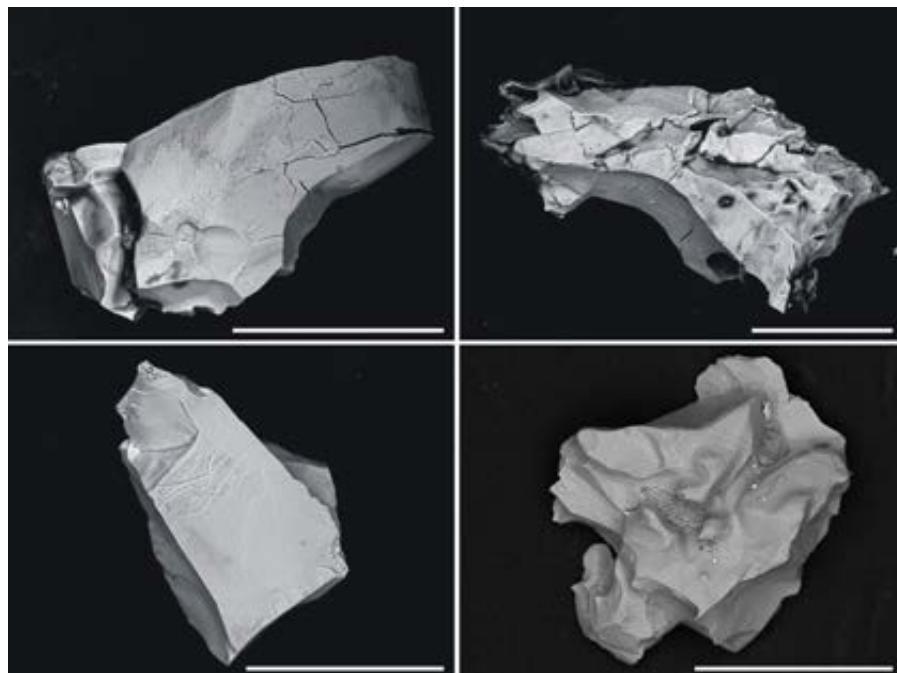


Fig. 12. Four typical extraterrestrial chromite (EC) grains (>63 µm) recovered from the Hällekis section in beds that formed shortly after the breakup of the L chondrite parent body. Scale bars = 100 µm.

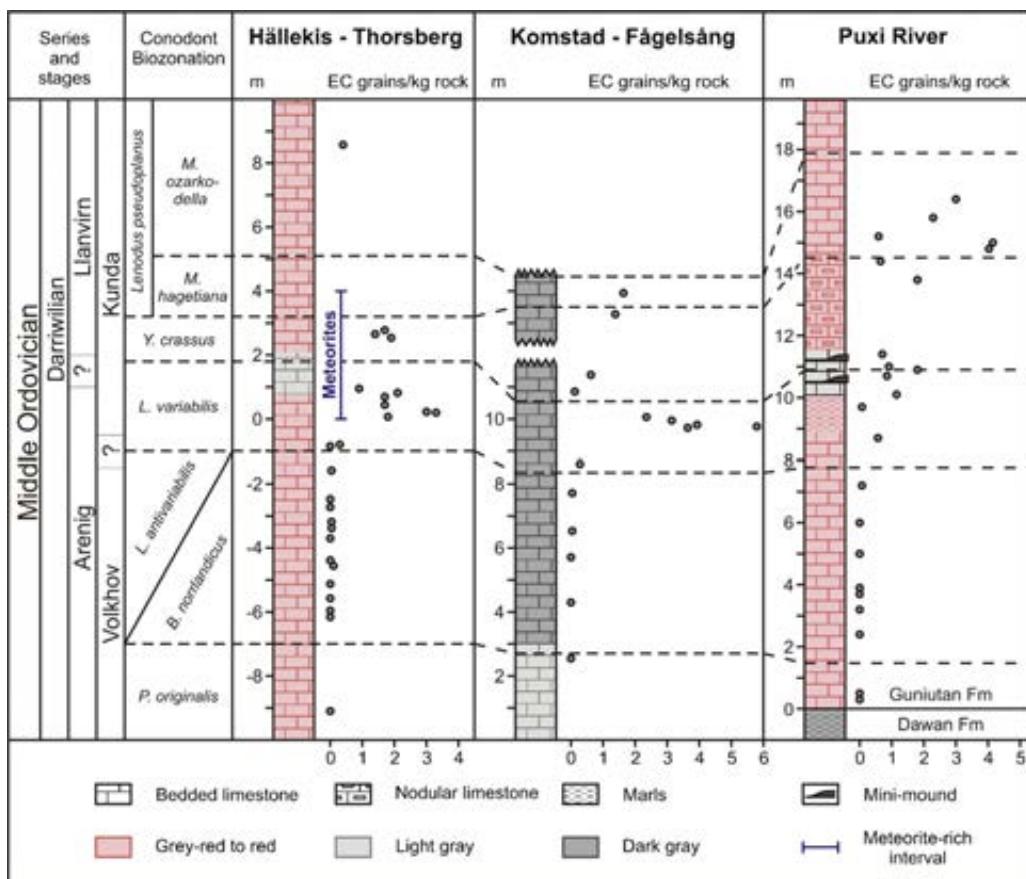


Fig. 13. Distribution of extraterrestrial chromite across two mid-Ordovician sections in Sweden and one in China (Puxi) (Schmitz et al., 2008). Note two orders of magnitude increase in chromite abundance in *L. variabilis* Conodont Zone. Interval with fossil meteorites in Thorsberg section is marked. See also Fig. 11 for the stratigraphic level of the L chondrite breakup event in the Hällekis section.

enrichment in EC occurs over an area of at least 250,000 km² of southern and central Sweden.

The same pattern has been reproduced to the east near St. Petersburg in Russia in a mid-Ordovician section with marine condensed limestone at Lynna River (Lindskog et al., 2012) (Fig. 2). This is a classical section through the mid-Ordovician where many paleontological and sedimentological studies have been performed since the early 20th century. In strata just below the level corresponding to the base of the *L. variabilis* Zone only 2 EC grains were found in a total of 38 kg of rock from three beds. In each of five beds of the section corresponding to the *L. variabilis* and succeeding *Yangtzeplacognathus crassus* zones, extremely high concentrations of EC were found, 5–10 grains per kg of rock (Lindskog et al., 2012). In the mid-Ordovician the Lynna River site was located 1100 km east of the Thorsberg quarry, on the same Baltoscandian plate as Sweden, in the easternmost extension of the large paleobasin where Orthoceratite Limestone formed (Fig. 14).

Very similar results as for the Baltoscandian sections have been obtained at Puxi River in the Hubei district of central China in a section with condensed mid-Ordovician limestone (Cronholm and Schmitz, 2010) (Fig. 13). In the mid-Ordovician the Puxi River site was located on the South China Plate, separated by a couple of thousand kilometers of open ocean to the east of the Baltica Plate (Cocks and Torsvik, 2002) (Fig. 14). In the Puxi River section 110 kg of limestone representing the 8 m interval of section below the base of the *L. variabilis* Zone yielded only 1 EC grain. Above the base, through the succeeding 9 m of section, 13 beds searched yielded in the range 0.6–4 EC grains per kg limestone. From the upper interval in total 290 L chondrite chromite grains were found in 178 kg of limestone.

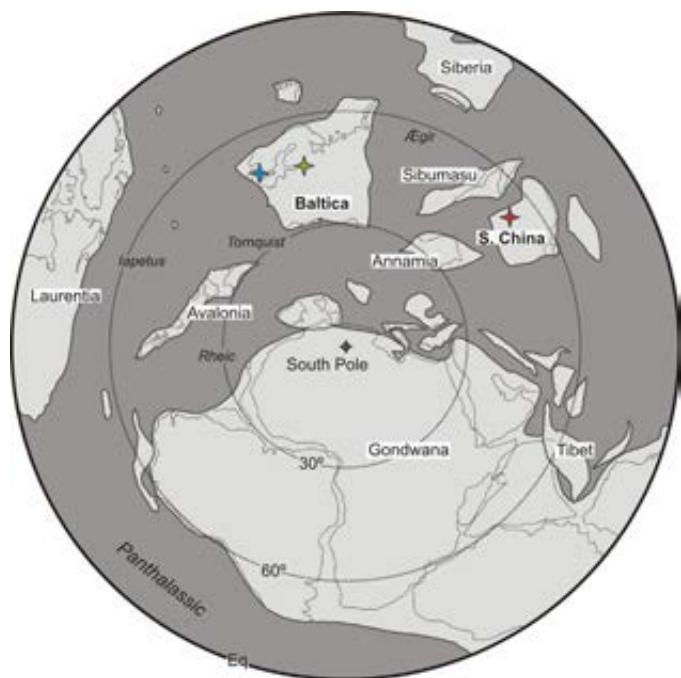


Fig. 14. Southern hemisphere paleogeography of the Middle Ordovician world ~470 Myr ago, modified from Cocks and Torsvik (2002, 2005) and Fortey and Cocks (2003). Studied localities marked by crosses.

The sediment-dispersed EC grains and fossil meteorites thus each give independent evidence for a two orders of magnitude increase in the flux of L chondrite material to Earth around 470 Myr ago. For the fossil meteorites, the enhanced flux is estimated from comparison with the recent flux of meteorites to Earth, and for the sediment-dispersed EC grains, by studying variations in the EC abundance through the mid-Ordovician sediment column. The change in EC concentrations appears to represent a world-wide, stratigraphically usable time marker.

2.4. Elemental composition of ordinary chondrite chromite

Relict ordinary chondrite chromite recovered from sediment samples or fossil meteorites can be readily identified by its unique elemental composition (Fig. 15). This can be done with a well-calibrated, standard EDX-analyser attached to a scanning-electron microscope. In the 1960s pioneering work on the geochemistry of the chromite phase in many meteorite groups laid the foundation for identifying extraterrestrial spinel grains from sediments (Bunch et al., 1967, 1970; Snetinger et al., 1967; Bunch and Keil, 1971). Further studies by Schmitz et al. (2001) and Wlotzka (2005) have extended the data base and confirmed the trends that were already established. Focus is on the “coarse” chromite of the ordinary chondrites (Ramdohr, 1963, 1967). This chromite from equilibrated ordinary chondrites has narrow ranges in elemental signatures. The most characteristic features are high Cr/(Cr+Al) mol% ratios of >0.85, and narrow ranges in V_2O_3 , ~0.6–0.8 wt% and TiO_2 , ~2.1–3.4 wt%. However, within the narrow compositional ranges there are also small differences that can be used to separate the three groups, L, H and LL. One of the most diagnostic oxides is TiO_2 , with an average in chromite of 2.2 wt% in H chondrites, 2.7 wt% in L chondrites, and 3.4 wt% in LL chondrites. There is overlap in the compositional ranges for H, L and LL chromite, so elemental signatures are not diagnostic for individual grains, only for populations of grains. Unequilibrated ordinary chondrites of petrologic type 3 contain much fewer and on average smaller chromite grains than equilibrated ordinary chondrites. Because these meteorites also are much rarer, the likelihood of finding such EC grains in the >63 μm fraction of a sediment is small. Chromite from type 3 chondrites can show large compositional variability, within a single meteorite and between meteorites, and single grains can be heterogeneous in their composition (Bunch et al., 1967). Some of this variability is also seen in chromites from L4 chondrites, which are not fully equilibrated (Grossman et al., 2009). In an evaluation of 13 recent L4 chondrites, Almmark and Schmitz (2009b) showed that chromites from these meteorites are chemically very similar to those of equilibrated L5–L6s, except that most of them have TiO_2 values between 1.4 and 2.1 wt%, instead of the typical L5/6 value around 2.7 wt%. When identifying extraterrestrial chromite from an ancient sediment, the effects of weathering on the grains must also be considered. Although the chromite grains generally are extremely resistant to weathering some diagenetic effects on element signatures have been observed. The Ti and V are generally unaffected by diagenesis, and also Cr, Al and Mg show considerable stability. Iron, on the other hand, has a tendency to be lost from the grains and replaced primarily by Zn, but possibly also by Mn. The ZnO contents can be high, 2–10%, but are often highest on exposed surfaces of the grains, like in cracks in the grains. The FeO contents of the grains can decrease by up to 10%, from 28–32 wt% to 18–22 wt%, but the $FeO + MnO + ZnO$ contents almost always are in the range 26–33 wt%.

Schmitz and Häggström (2006) defined a sedimentary EC grain, i.e. of ordinary chondrite origin, as being characterized by high Cr_2O_3 contents of ~55–60 wt%, FeO in the range of ~25–30 wt%, low Al_2O_3 at ~5–8 wt%, and MgO of ~1.5–4 wt%. The most discriminant feature, however, are the narrow ranges of V_2O_3 , ~0.6–0.9 wt%,

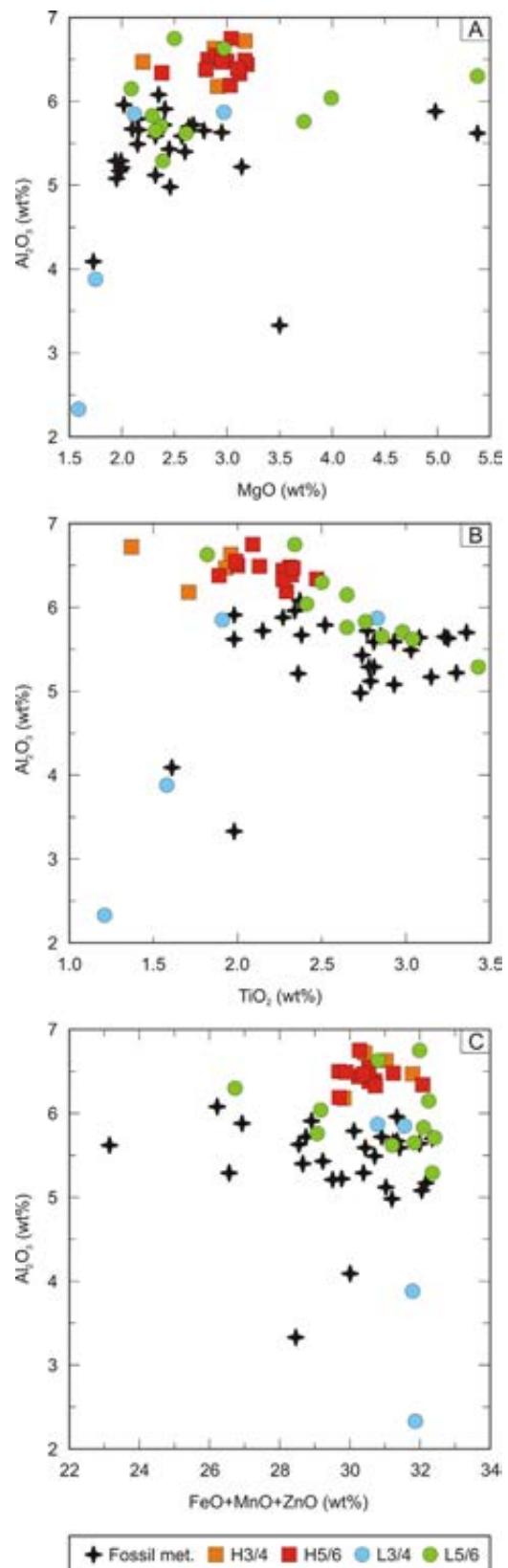


Fig. 15. Chemical composition (averages) of chromite grains from 28 fossil meteorites and 31 recent H and L chondrites (Schmitz et al., 2001, plus some new data). Note that the fossil meteorites plot with the L chondrites. The following recent chondrites were analyzed. H3: Brownfield 1937; H4: Bath, Dimmitt, Kesen; H5: Abajo, Agen, Forest City, Hesse, Jilin, Pultusk; H6: Archie, Benoni, Estacado, Kernouve, Ozona, Mills; L3: Julesburg; L4: Barratta, Saratov, Waltman; L5: Ausson, Elenovka, Ergheo, Etter, Tsarev; L6: Aztec, Baroti, Holbrook, Modoc 1905, New Concord, Wal ters.

and TiO_2 , ~2.0–3.5 wt%. It must be stressed that 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. For example, a grain with TiO_2 and V_2O_3 within the defined ranges, but with an Al_2O_3 concentration significantly outside this range is classified as “other chrome spinel”. This does not mean that the grain is terrestrial or not from an ordinary chondrite, it just means that its origin cannot be confidently determined based on composition alone. Based on the finding by Almärk and Schmitz (2009b) that many L4 meteorites have low TiO_2 concentrations, Cronholm and Schmitz (2010) revised the “accepted” EC range for TiO_2 to ~1.4–3.5 wt%. Because L4 grains make up only a minor fraction of all EC grains recovered, and not all L4 meteorites have chromite with lower TiO_2 , this generally has no implications for the distribution trends of EC grains in sedimentary sequences.

A worthwhile future study would be to determine the distribution of trace elements at parts per million levels in chromite of different recent ordinary chondrite falls and finds. It is likely that there are also trace elements that vary in a systematic way between the different ordinary chondrite groups. Trace elements to consider would particularly be those with an ionic radii similar to the elements that now are used to classify the grains. Trace element variations could be highly diagnostic and would diminish the need for difficult oxygen isotopic analyses in order to provide more robust group assignments for single chromite grains.

2.5. Oxygen isotopes in mid-Ordovician chromite

Oxygen three-isotope measurements have confirmed an L or LL chondrite origin of chromite grains from mid-Ordovician fossil meteorites and marine limestone. For recent ordinary chondrite falls and finds whole-rock oxygen isotope analyses have been used successfully to assign individual meteorites to their H, L or LL groups (Clayton et al., 1991; Clayton, 1993). For whole-rock samples both $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$ increase from H to L to LL groups (Fig. 16; see figure caption for definitions of $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$). In the oxygen three-isotope diagram the H chondrites are clearly separated from the L and LL meteorites, that lie close to each other and partly overlap. Heck et al. (2010) performed SIMS oxygen three-isotope analyses on single chromite grains from four of the Thorsberg fossil meteorites and the Brunflo meteorite. They also analyzed chromites from four recent meteorite falls (Guarena H6; Hessle H5; Ergheo L5; Saint-Séverin LL6). Just as for the whole-rock results for recent chondrites, the chromite oxygen isotope results for recent H chondrites are clearly separated from those of the recent L and LL meteorites, which give overlapping results (Fig. 17). All four Thorsberg fossil meteorites plot with the recent L or LL meteorites, distinctly separated from the H chondrites. For the Brunflo meteorite Heck et al. (2010) could provide the ultimate evidence that this is an L or LL chondrite, rather than an H chondrite (Fig. 17). The average major and minor elemental composition of chromite grains from each of the four Thorsberg fossil meteorites with an L or LL oxygen isotope composition is identical to that of chromite from the 26 fossil meteorites that had been analyzed by 2000 (Schmitz et al., 2001). This is a strong evidence that all or almost all of the fossil meteorites found in the Thorsberg quarry are L or LL chondrites. Heck et al. (2010) also analyzed sediment-dispersed EC grains from two beds in the Thorsberg quarry and two beds from the same biostratigraphically constrained interval in the Puxi River section in China. These grains gave the same average oxygen isotope composition as the recent L or LL chondrites and the five analyzed fossil meteorites (Fig. 17). The average elemental compositions of the sediment-dispersed grains both from Sweden and China are also very similar to that of chromite from the fossil meteorites, consistent with a common origin (Cronholm and Schmitz, 2010).

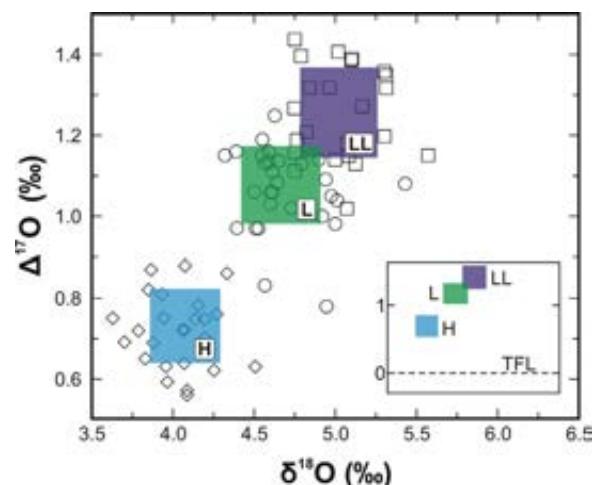


Fig. 16. Plot of whole-rock $\delta^{18}\text{O}$ versus $\Delta^{17}\text{O}$ for equilibrated ordinary chondrites. Shaded boxes represent the 1σ error on the whole-rock group mean values for analyses of 22, 26 and 20 recent H, L and LL chondrites, respectively, by Clayton et al. (1991); figure modified from Greenwood et al. (2007). Isotope ratios are defined as: $\delta^{18}\text{O} = ((^{18}\text{O}/^{16}\text{O}_{\text{sample}}/^{18}\text{O}/^{16}\text{O}_{\text{ref}} - 1) \times 1000$ and similarly for $\Delta^{17}\text{O}$ using the $^{17}\text{O}/^{16}\text{O}$ ratio. Values are expressed as per mil (‰) deviation from the international reference standard V-SMOW (Vienna-Standard Mean Ocean Water). When $\delta^{18}\text{O}$ is plotted against $\Delta^{17}\text{O}$, samples related to each other by mass dependent fractionation processes define linear arrays. Silicate rocks on Earth plot as such an array which is generally referred to as the Terrestrial Fractionation Line (TFL), having a slope of approximately 0.52. The TFL is a reference for determining the degree to which extraterrestrial material deviates from the isotopic composition of the Earth's oxygen reservoir. The term $\Delta^{17}\text{O}$ is used to quantify the extent to which a sample departs from the TFL and is defined as $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52\delta^{18}\text{O}$ (Clayton and Mayeda, 1996).

Chromite grains from one of the five fossil meteorites analyzed by Heck et al. (2010), Österplana 029, had earlier been analyzed for oxygen three-isotopes using laser-assisted fluorination by Greenwood et al. (2007). These authors determined the oxygen isotopic composition of a batch of about 130 chromite grains (total mass of ~0.9 mg) from this meteorite. Based on

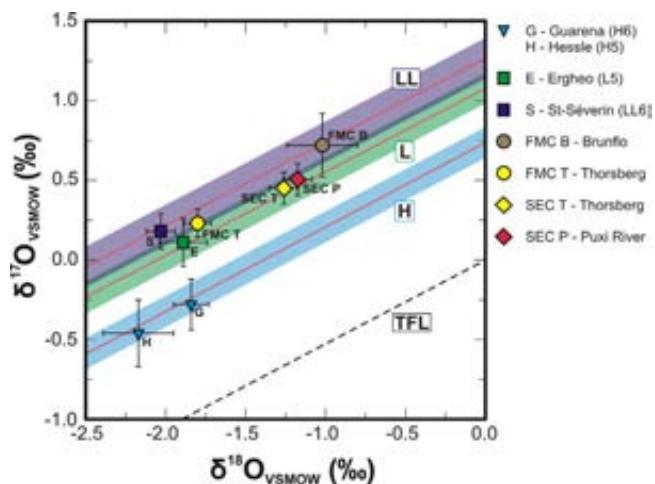


Fig. 17. Oxygen three-isotope results based on SIMS analyses of extraterrestrial chromite grains from fossil meteorites and from mid-Ordovician limestone from Sweden and China (Heck et al., 2010). Error bars are 2σ . Solid lines labeled H, L and LL are defined by average $\Delta^{17}\text{O}$ values of ordinary chondrite data from Clayton et al. (1991), see Fig. 16. The colored fields associated with each type of ordinary chondrite is defined by the standard deviation of the individual $\Delta^{17}\text{O}$ values within each ordinary chondrite group. For definitions of $\delta^{17}\text{O}$, $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$, see caption for Fig. 16. FMC B = fossil meteorite Brunflo; FMC T = average for four fossil meteorites from the Thorsberg quarry; SEC = sediment-dispersed extraterrestrial chromite grains; TFL = terrestrial mass-fractionation line, see Fig. 16.

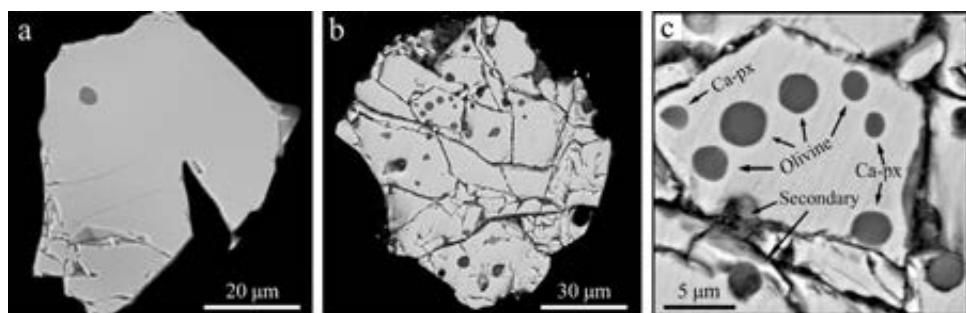


Fig. 18. Back-scattered electron images of silicate inclusions in polished, relict extraterrestrial chromite. (a) Solitary Ca-poor pyroxene inclusion in chromite grain from fossil meteorite Österplana 034; (b) olivine and Ca-rich pyroxene inclusions in chromite from fossil meteorite Brunflo; and (c) close-up of (b). From Alwmark and Schmitz (2009a).

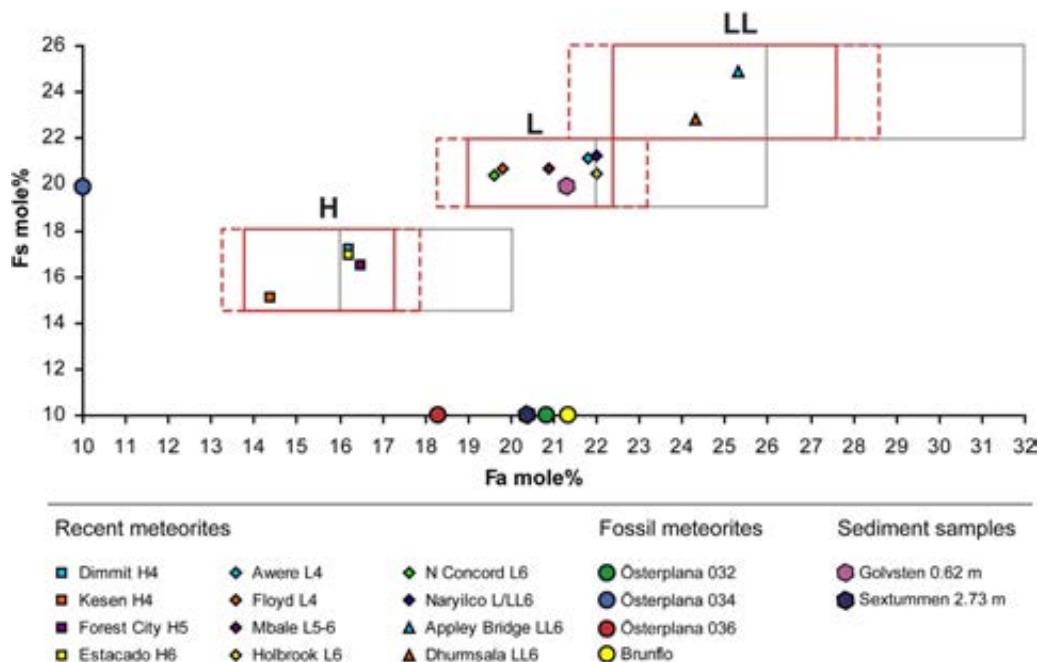


Fig. 19. Fayalite (Fa) content in olivine and ferrosilite (Fs) content in Ca-poor pyroxene inclusions in chromite (see Fig. 18) of recent and fossil meteorites and sediment-dispersed micrometeorites. All inclusions of fossil mid-Ordovician chromite show an L chondrite composition (Alwmark and Schmitz, 2009a).

abundance, size and chemistry of its relict chromite Bridges et al. (2007) considered this meteorite to be an L6 chondrite. The oxygen isotopic results by Greenwood et al. (2007) for Österplana 029 are very similar to the results for single-grain analyses by Heck et al. (2010), further supporting that this is an equilibrated L or LL chondrite.

In summary, the oxygen isotopic studies give strong support that all or most of the common fossil meteorites and sediment-dispersed EC grains in the mid-Ordovician strata have a single asteroidal source, very likely the breakup of the L chondrite parent body. We know from independent evidence, K-Ar ages of recent L chondrite finds and falls, that this body broke up at about 470 ± 6 Myr ago, which is very similar to the biostratigraphic age of 466 Myr for the sediments rich in fossil meteorites (Korochantseva et al., 2007; Gradstein et al., 2012).

2.6. Silicate inclusions in mid-Ordovician chromite

Encapsulated in the chondrite chromite grains from mid-Ordovician limestone and fossil meteorites are relict inclusions of many of the other types of minerals that made up the bulk of the decomposed original meteorite (Figs. 18–20) (Alwmark and

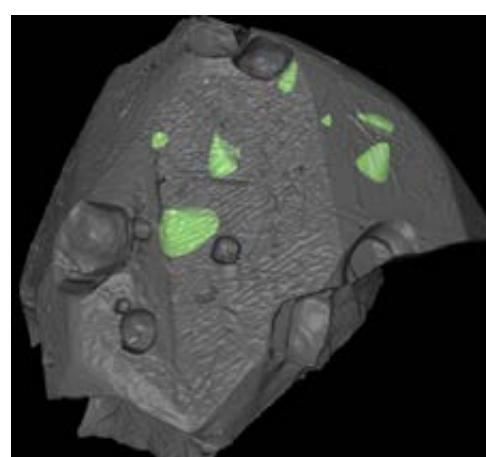


Fig. 20. Silicate inclusions in a chromite grain from the Farmington chondrite as revealed by 3-D synchrotron X-ray tomography at the Paul Sherrer Institute, Villigen, Switzerland. Field of view is 150 × 150 μm.

Schmitz, 2009a; Alwmark et al., 2011). The most common minerals, in the form of small anhedral inclusions (<1–10 µm), are olivine and pyroxene. In addition, sporadic merrillite and plagioclase are found. The resistant chromite has protected these minerals for almost half a Gyr from weathering processes. Olivine in particular is very susceptible in the terrestrial environment to decomposition through weathering. By element analyses of the inclusions, the fayalite (Fa) content of the olivine and the ferrosilite (Fs) content of Ca-poor pyroxene can be determined (Alwmark and Schmitz, 2009b). These two parameters are used in classifying recent meteorite finds and falls into H, L or LL groups (Keil and Fredriksson, 1964; Van Schmus and Wood, 1967). Thus by analogy to this classification system, a similar approach can be applied for fossil single chromite grains with inclusions. Inclusions in chromite from three of the Thorsberg and the Brunflo fossil meteorites, and from two sediment-dispersed EC grains from two beds in the Thorsberg quarry, have been analyzed for Fa and/or Fs content (Fig. 19) (Alwmark and Schmitz, 2009a). The results were compared with the ranges of Fa and Fs in chromite inclusions in 12 classified recently fallen, equilibrated (type 4–6) chondrites of the H, L and LL groups. The three groups give clearly defined and separate fields in a Fa versus Fs plot, with all the fossil meteoritic material falling within the L group field. Since for olivine-pyroxene compositions, there is no major overlap between the L and LL fields, as is the case for oxygen isotopes and the elemental composition of ECs, the inclusion study represents the best evidence so far that the abundant ECs in mid-Ordovician sediments are of L rather than LL chondrite origin.

Inclusions in chromite can be found through stepwise layer-by-layer polishing of epoxy-mounted grains, but this is a very tedious and imprecise method. The method includes repeated mild polishing followed by studies of the recovered surfaces in the scanning electron microscope. This process has to be repeated until an inclusion is found. The method is destructive, and also chromite grains without inclusions will be lost. Therefore a method was developed to search for inclusions in chromite grains from ordinary chondrites using 3-D synchrotron X-ray tomographic microscopy (Fig. 20) (Alwmark et al., 2011). The studies were performed at the TOM-CAT beamline at the Paul Scherrer Institute in Switzerland. The method is non-destructive and time efficient for locating inclusions, and allows quantitative and morphological studies of both host chromite grains and inclusions in three dimensions. The 3-D imaging of 385 chromite grains from eight recently fallen chondrites (H4–6, L4–6, LL4, LL6) showed that about two-thirds of all chromite grains contain inclusions, regardless of meteorite group and petrologic type. However, the study showed a clear relation between the size and number of the inclusions in the chromite and the petrologic type. With higher petrologic type, the inclusions in chromite grains become larger and fewer. This is what one would expect from the increase in equilibration with higher grade thermal metamorphism.

In principle, it is possible to also determine the petrologic type of a fossil meteorite by scanning its chromite grains for inclusions, and compare the results with those for recent meteorites. In addition to the number and size of the inclusions, the TiO₂ concentration and the size of the chromite grains will give independent, robust information about petrologic type (Bridges et al., 2007; Alwmark and Schmitz, 2009a). Determining the petrologic type of fossil meteorites could be important in testing, for example, if there was a “memory” in the structure of the L chondrite parent body 470 Myr ago of an original onion-like structure (Williams et al., 2000; p. 414 in Hutchison, 2004). It could be that the outer parts of the body were still richer in L3 and L4 material than deeper parts of the asteroid. Possibly, immediately after the breakup of the parent body, Earth was reached primarily by material from the outer parts of the asteroid. Today, when secondary collisions of large asteroids from the breakup event produce material that reaches Earth, the flux is

dominated by material from deeper parts of the parent body. Bridges et al. (2007) in their study of Ordovician fossil meteorites found indications of a higher proportion of meteorites of lower petrologic type than among recently fallen L chondrites, however, this was based on only six fossil meteorites.

2.7. Noble gases in mid-Ordovician spinels

The chondrite chromite grains from mid-Ordovician fossil meteorites and limestone also hold a record of noble gases induced by the solar wind and galactic cosmic rays (Heck et al., 2004, 2008; Meier et al., 2010; Alwmark et al., 2012). The Ne from cosmic rays is a spallation product formed when high-energy protons hit the target material, whereas solar wind Ne represents implanted atoms traveling with the solar wind. Grains from fossil meteorites are dominated by cosmic ray induced Ne, whereas in the sediment-dispersed grains this signal is over-printed by a strong solar wind Ne component. This is a reflection of the different penetration depths of the energetic galactic and the less energetic solar radiations. The former can reach depths of up to 2 m in a meteoroid, whereas the latter only penetrate the outermost 100 nm. This difference in how the Ne signal is acquired attests to a micrometeorite origin of most of the sediment-dispersed EC grains (Heck et al., 2008). The consistently different Ne signals for the two populations of grains from the same beds, sediment-dispersed or in situ in a meteorite, show that signals are original, acquired in space, rather than in the sedimentary rock. This is further corroborated by a complete absence of extraterrestrial Ne in terrestrial chrome spinel grains recovered from the same beds (Meier et al., 2010). We do not rule out that some extraterrestrial Ne has been lost from many EC grains during diagenesis, but believe that even partially preserved signals can give insights about major trends in anciently acquired extraterrestrial noble gases. The ³He record of the EC grains does not reproduce the Ne trends, instead it appears that this gas has experienced substantial diffusion loss out of the grains. In our studies of noble gases in single chromite grains, we have used a unique ultra-high-sensitivity mass spectrometer with a low-blank extraction line at the Department of Earth Sciences at ETH Zürich. The mass spectrometer concentrates gases into the ion source by a molecular drag pump as described by Baur (1999).

Chromite grains from nine fossil meteorites from six beds across ~3.8 m of vertical distance in the Thorsberg quarry show comparatively low cosmic ray exposure ages based on ²¹Ne, from ~0.1–0.2 to ~0.8–1.2 Myr, and the cosmic ray exposure ages generally increase upward in the sediment column (Fig. 21) (Heck et al., 2004). The difference in cosmic ray exposure age between the stratigraphically highest and lowest meteorite, ~1 Myr, is similar to the time difference in deposition on the sea floor based on biostratigraphic estimates of sedimentation rates (see Schmitz et al., 1996; Alwmark et al., 2012). The meteorites in the oldest bed quarried, Arkeologen, occur stratigraphically only ~80 cm above the first level (in the Hällekis section) with abundant sediment-dispersed EC grains, i.e., the level most likely representing the time for the breakup of the L chondrite parent body (Fig. 11). The young cosmic ray exposure ages, ~0.05–0.2 Myr, of the Arkeologen meteorites and the gradual increase upward of the exposure ages conforms with the idea that all meteorites originated from one breakup event, and reached Earth at successively later times. The meteorite found in the Gullhögen quarry, 35 km to the southeast, at the same biostratigraphically defined level as the meteorites with the highest cosmic ray exposure ages at the Thorsberg quarry, gives a similar high ²¹Ne age, ~0.9 Myr (Heck et al., 2008). All the measured cosmic ray exposure ages are substantially lower than exposure ages of recent ordinary chondrite finds and falls, that lie typically in the range of 4–60 Myr (Wieler and Graf, 2001). Orbital simulations of very large collisions in the asteroid belt leading to a

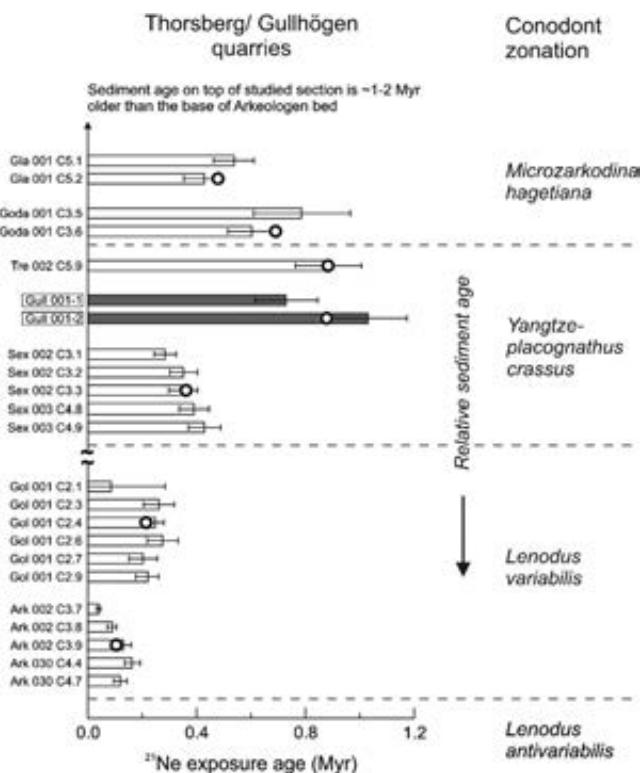


Fig. 21. ^{21}Ne cosmic ray exposure ages of chromite from eight fossil meteorites from the Thorsberg quarry, spanning samples from the Arkeologen to the Glaskarten 1 bed (see Fig. 10) and the fossil meteorite (dark bars) from the Gullhögen quarry (Heck et al., 2004, 2008). The ^{21}Ne ages increase upward in the section, in consistence with sedimentation rates, indicating that all meteorites originate from one breakup event having taken place shortly before the Arkeologen bed formed. This event in time most likely corresponds to the stratigraphic level where the first abundant EC grains appear, at about 0.8 m below the base of the Arkeologen bed (see Figs. 11 and 13). The total sedimentation time of the ~4 m thick sedimentary section from which the fossil meteorites derive is ~1–2 Myr based on conodont biochronology and the Geological Time Scale (Schmitz et al., 1996; 2001; Zhang, 1998; Gradstein et al., 2004, 2012; Mellgren and Eriksson, 2010). Error bars are 1σ , and include weighing uncertainties, ion statistics, and blanks. The results are grouped based on the beds from which the fossil meteorites originate. Open large circles are averaged ^{21}Ne exposure ages of all analyses from the same quarry bed. Different preatmospheric sizes of meteorites and moderate losses of cosmogenic Ne during diagenesis may also have affected the individual results, but not to the extent that the general trend for the data set was lost. Informal names of fossil meteorites are used in the figure. These names relate the meteorite to the bed in which it was found (see Fig. 10), or quarry, for Gull 001. Bed names: Ark = Arkeologen; Gol = Golvsten; Sex = Sextummen; Tre = Tredje Karten; Goda = Goda Lagret; Gla = Glaskarten 1. Quarry name: Gull = Gullhögen.

temporary increase in the meteorite flux to the inner solar system indicate that fragments from such events may reach Earth considerably faster than the typical transit times of meteorites falling today. A fast delivery is expected if the asteroid collision occurred close to a major orbital resonance in the inner asteroid belt (Gladman et al., 1997; Zappalà et al., 1998; Nesvorný et al., 2007). Two important resonances are the 3:1 orbital resonance with Jupiter at about 2.5 AU, and the Saturn and Jupiter ν_6 resonance at 2–2.5 AU, depending on orbital inclinations (Heck et al., 2008). These two resonances have probably played a crucial role in transport of asteroidal matter to the inner solar system through the ages. Based on the short cosmic ray exposure ages of the mid-Ordovician fossil meteorites, Nesvorný et al. (2009) suggested an origin from the breakup event that created the Gefion asteroid family in the outer part of the main asteroid belt. In their simulations, numerous fragments evolved into Earth-crossing orbits by the 5:2 resonance with Jupiter.

Only one of the ten fossil meteorites analyzed for Ne and He isotopes in their chromite contained a solar wind Ne component.

This meteorite, Österplana 002 (informal name: Ark 002), is interpreted as a regolith breccia, with the solar wind signal acquired while it resided in the regolith of the L chondrite body (Heck et al., 2004). The surface-seated solar wind signal, acquired during transport to Earth, is lost when the surface of a large meteoroid is vaporized and melted during its passage through Earth's atmosphere, but the internal regolith signal is retained. In collections of recent L chondrites regolith breccias make up about 3%, a value not too different from the 1 out of 10 among mid-Ordovician meteorites. In stark contrast almost all of the mid-Ordovician sediment-dispersed chromite grains contain substantial amounts of solar wind implanted Ne, attesting to the fact that they are micrometeorites or fragments thereof. At least a part of the chromite grain surface must have been shared with the surface of the enclosing micrometeorite. Heck et al. (2008) analyzed batches of 4–6 sediment-dispersed EC grains from eight samples representing three different beds in the meteorite-yielding part of the composite Hällekis-Thorsberg section. In each of the samples they found high levels of solar wind derived Ne, indicating that at least one of the grains in each batch had to represent a micrometeorite. Meier et al. (2010) took the issue one step further by analysing 37 individual sediment-dispersed L chondrite chromite grains, and showed that at least 35 of them (~95%) contain surface implanted He and Ne of fractionated solar wind composition. The presence of surface implanted solar wind gases indicate that the precursor micrometeoroids were not heated to high temperatures. This conforms with what is known about the solar wind implanted noble gases in recent interplanetary dust particles collected in the stratosphere and micrometeorites from cryogenic lakes on Greenland. The contents of extraterrestrial noble gases are comparable with the lower range of concentrations in stratospheric interplanetary dust particles, and mostly higher than concentrations found in recent micrometeorites. The least gas-rich chromites in the study of Heck et al. (2008) have similar Ne concentrations as the most gas-rich micrometeorites from Greenland ice (Olinger et al., 1990). The fact that the mid-Ordovician SEC grains have retained all or most of their noble gases when landing on the sea floor, would be consistent with a shallow, slow (12 km/s) entry into the Earth's atmosphere (Meier et al., 2010). This is typical for particles on circular heliocentric orbits, such as the ones reaching Earth by Pointing-Robertson drag.

One initial goal of the analyses of sediment-dispersed EC grains was to determine the cosmic ray exposure ages of the grains, but in many cases the copious amounts of solar gases give $^{21}\text{Ne}/^{22}\text{Ne}$ ratios too close to solar values to reliably determine any excess of cosmogenic ^{21}Ne (Heck et al., 2008; Meier et al., 2010). A prospective future approach would be to somehow etch off the outermost solar gas bearing layers of the chromite grains so that only the galactic ray induced He and Ne component would remain. Meier et al. (2010), in addition to the fractionated solar wind component, also found significant amounts of cosmogenic ^{21}Ne in several sediment-dispersed EC grains. This yielded cosmic ray exposure ages up to ~50 Myr, which exceeds both dynamical lifetimes for asteroidal micrometeorites of this size as well as cosmic ray exposure ages measured in chromites from larger fossil meteorites in the same beds (<1.2 Myr) (Fig. 22). Such high cosmic ray exposure ages have also been reported for micrometeorites from recent Greenland ice and were interpreted in terms of a cometary, or Kuiper belt, origin of the micrometeorites, with on the order 10 Myr transport times to Earth (Osawa and Nagao, 2002; Kehm et al., 2006). The mid-Ordovician chromite grains, however, have a definitive ordinary chondrite, i.e. asteroidal origin, showing that such material can also have high cosmic ray exposure ages. Meier et al. (2010) suggested that the grains collected their cosmogenic ^{21}Ne while residing in the regolith layer of their parent body. About a third of the grains in the study showed exposure ages >3 Myr and up to

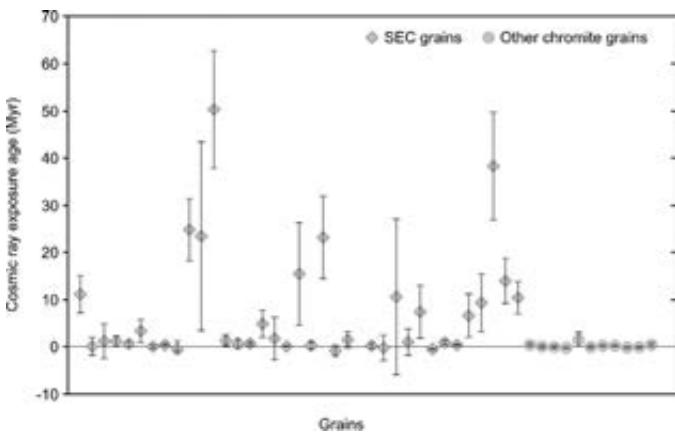


Fig. 22. Cosmic ray exposure ages calculated from cosmogenic ^{21}Ne excesses observed in individual sediment-dispersed EC grains (SEC, diamonds) and other chromite grains (circles), presumed to be of terrestrial origin based on their elemental composition. The grains originate from the Arkeologen or Sextummen beds, see Fig. 10. All error bars are 1σ . The cosmogenic ^{21}Ne may have been collected by the SEC grains while residing in the regolith layer of their parent body. The high proportion of regolithic grains in 470 Myr old sediments may reflect that the outermost parts of the exploded parent body was the material that first reached Earth. From Meier et al. (2010).

~50 Myr, the remaining grains had much shorter cosmic ray exposure ages (Fig. 22). The two populations of micrometeorite grains in the same beds must have had very different exposure histories. The grains with high cosmic ray exposure ages probably resided near the surface in the regolith of the L chondrite parent body, where they picked up the bulk of their Ne from solar wind and galactic cosmic rays. The gas poor EC grains with low exposure ages were shielded from cosmic rays over the history of the solar system, and acquired their smaller amounts of solar wind gas only during the “fast” transport to Earth. The high amount of regolith material among Ordovician micrometeorites is explained by the fact that the bulk mass of the L chondrite parent body was not pulverized in the breakup event, but instead formed a collisional family, like the Gefion asteroid family (Nesvorný et al., 2009). Immediately after the breakup event material from the outer parts of the parent body and near-surface material may have represented a larger fraction of the material reaching Earth, than at later times.

2.8. Spinel from a mid-Ordovician 600 m sized asteroid

Only for a few of Earth's ~180 known impact structures has the type of impacting projectile been determined (Grieve, 2001; Grieve and Stöffler, 2012). Large projectiles creating craters larger than 1–2 km tend to completely vaporize upon impact, leaving behind only a chemical fingerprint that may be fractionated by various processes, both in the ejecta plume and during diagenesis of the ejecta in sediments (Schmitz et al., 2011; Koeberl et al., 2012). In rare cases whole pieces of the impactor are preserved as for the Eltanin and Morokweng impact events (Kyte, 2002; Maier et al., 2006). A third case is the Ordovician Lockne crater, 10 km in diameter, in central Sweden. In iridium-rich (~4.5 ppb Ir) ejecta samples from the rim of the crater, Alwmark and Schmitz (2007) found extremely high concentrations of chromite grains (>63 μm) with L chondrite composition. More than 75 grains were found per kilogram of ejecta, representing an enrichment by many orders of magnitude compared to background. This is the most EC-enriched sediment sample known today. The grains were interpreted as relict remains of an L chondrite impactor, representing the high-mass fraction in the enhanced flux of extraterrestrial matter after the breakup of the L chondrite parent body. Tagle et al. (2008) challenged this view based on Ir/Cr and platinum group element ratios in the Lockne

ejecta as well as a reinterpretation of the elemental signatures of the chromite grains. They instead favored that the impactor was a non-magmatic iron meteorite. Schmitz et al. (2011) provided oxygen isotopic data for the Lockne chromite grains, confirming an L chondrite origin. They also showed that the chromite grains are completely devoid of extraterrestrial He and Ne, in line with an origin from a large body at depths to which the galactic cosmic rays could not penetrate. The Lockne impactor has been estimated to have had a size of 600 m in diameter (Ormö and Lindström, 2000). The penetration depth for galactic cosmic rays is 1–2 m, so less than 2% of the chromite grains of the impactor would have been exposed to galactic cosmic rays. Schmitz et al. (2011) also showed that when calculating the average platinum group element (PGE) concentration of the many Lockne ejecta samples analyzed by Tagle et al. (2008), rather than using the regression slope approach of these authors, the ejecta has ordinary-chondrite PGE ratios. The regression slope approach of Tagle and Claeys (2005) and Tagle et al. (2008) has also been criticized by Farley (2009) who showed that different results can be obtained just by switching x and y axes in the regression plots, and that single outlier data may affect the conclusions significantly.

The extraterrestrial chromite grains in the Lockne ejecta most likely represent the relict residues of weathered small pieces of the impactor (Alwmark and Schmitz, 2007). The Lockne crater formed in ~500–1000 m deep water, and when an asteroid hits deep water pieces of the impactor may escape vaporization as shown by the abundant unmelted meteorite fragments found in sediments from the late Pliocene Eltanin impact site in the Southern Ocean (Kyte, 2002). Detailed and robust biostratigraphy has established that the Lockne crater formed at ~458 Myr ago, ~8 Myr after the first micrometeorites from the breakup of the L chondrite parent body showered the Earth (Grahn et al., 1996; Schmitz and Häggström, 2006). This time lag is in agreement with modeling simulations of large breakup events showing that the larger km-sized bodies typically tend to reach Earth on the order of 1–30 Myr later than the dust particles (e.g., Zappalà et al., 1998; Dermott et al., 2002). Poynting-Robertson light drag is important in transferring particles <500 μm directly from the asteroid belt to the inner solar system, whereas km-sized bodies usually slowly drift via the Yarkovsky effect into orbital resonances (e.g., Bottke et al., 2002), which can lead to rapid change of orbital parameters and injection into the inner solar system (Gladman et al., 1997).

The terrestrial crater record is very incomplete, representing only a fraction of a percent of all craters that have formed on Earth, and assigned ages of many craters have large uncertainties (Grieve, 2001). Nevertheless, Schmitz et al. (2001) noted a factor of three to four enhancement in well-dated craters with ages of ~450–480 Myr. Five out of the known 17 impact craters in Baltoscandia are confidently dated by biostratigraphy to the mid-Ordovician (Tvären, Lockne, Hummeln, Granby and Kärdla). An increase in mid-Ordovician craters was also suggested by Korochantseva et al. (2007). A study of the ages of lunar melt droplets formed by impacts similarly found a peak in the mid-Ordovician interpreted as a reflection of an increased bombardment of the lunar surface following the L chondrite breakup event (Culler et al., 2000). Modeling suggests that such an increase in lunar impacts would be an expected effect of the breakup of the L chondrite parent body (Artemieva and Shuvalov, 2008).

The discovery in the Lockne crater ejecta of abundant relict mineral grains from a large asteroid opens up some new, interesting research venues. The Lockne crater is well exposed and access can be gained to Ir-rich ejecta at many places along the crater rim (Sturkell, 1998). Most likely the samples also contain asteroidal chromite, with different parts of the asteroid represented in the ejecta at different places in the crater. By quantifying the number and sizes of silicate inclusions in the chromite grains, and

comparing with data for recent meteorite finds and falls of different petrologic types (Alwmark et al., 2011), clues may be obtained as to the mix of petrologic types in a 600 m large asteroidal body such as the Lockne impactor. The PGE-rich ejecta of the Lockne crater, with a known L chondrite impactor, may also give a better understanding of how extraterrestrial PGE interelemental ratios can become fractionated during impact and in the sediment (Schmitz et al., 2011).

2.9. Mid-Ordovician spinels as proxies of sedimentation rates

A classical problem in studies of Earth's geological record has been to determine the sedimentation rate of a specific bed or a series of beds in the rock strata. The dramatically enhanced flux of extraterrestrial matter in the mid-Ordovician, and the robust proxy representing that flux (extraterrestrial chromite), opens up for new ways of reconstructing the formation of sediments in various paleoenvironments at this time. Sedimentation rates for a section are normally determined by interpolation, using the ages of, for example, bio- or magneto-zone boundaries as given in the most recent Geological Time Scale (Gradstein et al., 2012). The ages of the zone boundaries in the time scale are typically established by interpolation between radiometrically dated ash layers. The estimates of sedimentation rates by this approach can only give the average sedimentation rate for an extended part of a section. Intrazonal hiatuses cannot be accounted for and sedimentation rates for a specific bed in the section cannot be determined. This was the challenge that originally led Alvarez et al. (1980) to measure the element iridium in the K-T boundary clay at Gubbio in Italy. The intent was to determine whether the boundary clay was, in a geological context, a slowly or rapidly formed bed. Its iridium content would give an estimate of the amount of extraterrestrial dust in the clay. By comparison with the known recent flux of extraterrestrial dust to Earth, the rate of formation of the boundary clay was to be estimated. The project, however, "failed" as the boundary clay turned out to be the product of the largest known asteroid impact on Earth during the Phanerozoic. Other attempts have been made to relate iridium content to sedimentation rate, but additional sources of iridium, mainly from seawater, complicate the interpretations. For example, the relatively high iridium concentrations in condensed deep-sea sediments were once believed to be mainly of extraterrestrial origin, but later studies using osmium isotopes have indicated instead that the iridium dominantly (70–85%) originates from sea water (Peucker-Ehrenbrink, 1996; Dalai and Ravizza, 2006).

The distribution of EC grains in China, Russia and Sweden in mid-Ordovician sediments formed after the L chondrite breakup event, provides new information about relative variations in sedimentation rates, including on a bed-by-bed basis. The EC grains at all three sites are dominantly of a micrometeoritic origin, based on noble gas measurements of the grains (Meier et al., 2010, 2013; Alwmark et al., 2012). After the breakup event, the micrometeorites were transported to Earth in a large cloud of dust by Poynting-Robertson drag. The dust cloud was most likely quite homogeneous over large astronomical distances. In the time perspective of tens to a few hundred thousand years, the dust flux to Earth would not have differed significantly. Any major difference in EC grain content in, for example, two limestone beds adjacent to each other, would in most cases mainly reflect differences in sedimentation rate. By comparing trends in EC distribution through time from several sections, it would also be possible to disentangle if any changes in flux have occurred with time or whether differences between sections are entirely related to site-specific variations in sedimentation rate. From the EC distribution across the section at Puxi River in China one can see that sedimentation rates gradually decrease by a factor of two to four upward through the section. Beds in the upper part of the section generally contain 2–4 EC grains per kg rock,

compared to 0.7–1.8 EC grains per kg in the middle part of the EC-rich interval (Cronholm and Schmitz, 2010). There is no similar gradual increase in EC grains upward through the section at Hällekis-Thorsberg in Sweden. Instead a relatively stable trend is observed with EC grain concentrations typically close to 2 grains per kg rock through most of the middle and upper EC-rich part (Schmitz and Häggström, 2006). The lower EC concentrations, ~0.3 grains per kg rock in the first meter of strata showing EC grains at Hällekis-Thorsberg, however, could very well be related to a weaker dust flux before the main dust pulse reached Earth after the L chondrite breakup (Häggström and Schmitz, 2007). The most EC-rich limestone beds are found at the Lynna River site in Russia, with concentrations up to 10 grains per kg rock (Lindskog et al., 2012). In the >63 µm fraction in these beds, EC is typically the most common opaque mineral, outnumbering even the very common terrestrial ilmenite.

In the EC-rich part of the mid-Ordovician sections in Sweden, China and Russia there are some lithologically anomalous beds at about the same biostratigraphic level, i.e., close to the *L. variabilis*–*Y. crassus* conodont zones boundary (Schmitz et al., 2008; Lindskog et al., 2012). Over large parts of Sweden, at this level in the column of reddish condensed limestone, there is a prominent one-meter thick grey, glauconitic interval rich in clay intercalations, the so called Täljsten interval (Schmitz and Häggström, 2006; Eriksson et al., 2012). In Russia, the same level is represented by a distinct and anomalous ~10 cm thick red clay bed in a context of otherwise condensed red limestone (Lindskog et al., 2012). At the same stratigraphic level in the Puxi River section in China, there are highly unusual so called micromounds (e.g., Lindström et al., 1991). These are clayey, biologically mediated, 0.2–0.5 m high, mound-like structures that formed on the sea floor. The sedimentological evidence thus indicates that worldwide there were unusual perturbations in the oceans coinciding with the peak in the enhanced flux of meteoritic debris to Earth. Based on high clay content and some faunal parameters, the grey Täljsten interval in Sweden has generally been interpreted as the expression of a sea-level fall (Tinn and Meidla, 2001; Mellgren and Eriksson, 2010; Eriksson et al., 2012). This would have led to a more near-shore facies, with a higher detrital input and higher sedimentation rates. The amount of EC grains in the beds that make up the Täljsten is similar to the condensed red limestone beds above and below. The EC data for the Täljsten interval thus do not support faster sedimentation and a sea-level fall. Rather the emerging data suggest a large-scale oceanic event, with enhanced plankton productivity in the water mass, perhaps with similarities to the global Kellwasser events in connection with the major mass extinction event at the Frasnian-Famennian boundary (Tribovillard et al., 2004).

3. Spinels and the meteorite flux through the ages

3.1. Dissolving tons of rock per year

The insights gained from studies of Ordovician extraterrestrial spinels allow for a "first-order" reconstruction of variations in the meteorite flux to Earth through the ages. From many periods of Earth's history we know of preserved sediments that formed very slowly, far from land and sediment sources. In today's world, the brown pelagic clays of the central Pacific represent such condensed sediments, forming at rates of a few millimetres per thousand years. Some well known examples of ancient condensed sediments are the Orthoceratite Limestone in Baltoscandia and China, the late Devonian limestones of the Montaigne Noire region in France, the Jurassic Ammonitico Rosso limestones of southern Europe, and the early Paleogene limestones of the Italian Apennines. By analysing large (100–1000 kg) samples of condensed sediments for

spinels, “windows” into the meteorite flux at particularly interesting periods in Earth's history can be created. The size of the samples required to obtain a representative set of relict extraterrestrial minerals relates both to sedimentation rate and to the flux rates of meteorites at the particular time of interest. The more rare a specific type of meteorite is, and the lower the content of weathering-resistant spinels in that meteorite type, the larger the samples must be. By searching for meteoritic minerals in smaller size fractions than $63\text{ }\mu\text{m}$, one would find more grains, but they would also be more diluted with terrestrial grains, increasing the “needle-in-the-haystack” problem. The optimal approach appears to adjust the size of the studied sediment samples so that representative collections of meteoritic grains larger than $\sim 30\text{ }\mu\text{m}$ are found. Such grains can also be analyzed for their elemental and oxygen isotopic compositions with relatively straightforward methods, such as EMPA and SIMS. So far we have studied a few large pilot samples of condensed limestone from periods other than the mid-Ordovician for relict extraterrestrial spinels. For example, from the Scaglia Rossa section at Gubbio in Italy, 210 kg of limestone from different levels of the early Paleocene and uppermost Cretaceous interval were searched for spinels. These sediments are pelagic and formed at $\sim 2\text{--}5\text{ mm per Kyr}$. In this sample 6 EC grains and one pallasitic chromite grain were found (Cronholm and Schmitz, 2007). A flux rate of 0.26 chromite grains from unmelted micrometeorites per m^2 per Kyr was estimated for the early Paleocene. The number of grains recovered are too few, however, for any speculations about the distribution of different meteorite types in the flux at $\sim 66\text{ Myr}$ ago. Similar results were obtained in a study of similarly condensed Ordovician limestone deposited before the L chondrite disruption event. In 379 kg of limestone, 5 EC grains were found, giving a flux rate for chromite about half as that for the Paleocene (Schmitz and Häggström, 2006). These estimates, however, are only valid at the order-of-magnitude scale. In an ongoing study of condensed limestone from the Frasnian-Famennian boundary of the Montaigne Noire region on the order of 10–15 EC grains per 400 kg rock have been found, well in line with the other results (Schmitz et al., in prep.). Test samples of 30–100 kg from several other sites and periods have also been studied, e.g. Ammonitico Rosso limestone of Jurassic age from Adnet in Austria and early Paleocene limestones from the Zumaia section in Spain. Typically only single extraterrestrial grains were found in these samples. Clearly, in order to recover on the order of one hundred chondritic grains from the most condensed parts of a section such as at Gubbio, with a sedimentation rate of 2–5 mm per Kyr, one would need to dissolve about three tons of rock.

At the present a specially designed laboratory at Lund University is being built that can handle 5–10 tons of sedimentary limestone per year for recovery of extraterrestrial minerals. The laboratory will be based on simple mechanical principles. Rock samples are placed in large plastic barrels and the acid is transported by acid-resistant pumps between different containers. After the rocks are dissolved, surplus acid is removed and neutralized. The acid leaching will consume a factor of two as much HCl as rock is dissolved, but this is still an insignificant amount of acid compared to that used routinely in various industrial applications. After HCl leaching of 5 tons of rock, about half a ton of clay will remain. Further steps in the recovery of EC grains will involve sieving at $\sim 30\text{ }\mu\text{m}$, tenside treatment to deflocculate clays, HF leaching, and in some cases heavy liquid separation. From 5 tons of carefully selected condensed limestone we estimate to recover on the order of 150–500 extraterrestrial spinel grains $> 63\text{ }\mu\text{m}$, and one order of magnitude more grains in the $32\text{--}63\text{ }\mu\text{m}$ fraction. Focus will be on the $> 63\text{ }\mu\text{m}$ grains, because they allow much more detailed chemical and petrological studies. The smaller size fraction can be used for reconstructions of the flux of rare meteorite types and meteorite types with smaller spinel grains. The large samples from

different time periods should ideally give an as complete a spectrum as possible of the types of meteorites and dust reaching Earth. The material representing each time “window” would need to represent a composite sample collected at different levels in a section or in different sections in order to bypass bias from unusual meteorite showers or asteroid breakups in the water column or atmosphere. It is important to make major efforts in selecting the optimal type of condensed limestone. A sediment formed at 1 mm rather than 3 mm per Kyr would require two thirds less labor and acid. Only the most condensed sediments are useful for this approach, as shown by the results from 167 kg of hemipelagic limestone from the late Eocene Massignano section in Italy (Schmitz et al., 2009). This sample yielded 373 terrestrial chrome spinel grains and only 1 EC grain. The EC result is congruent with estimated sedimentation rates for the Massignano section a factor of three higher than, for example, the condensed Paleocene sediments at Gubbio (see above).

3.2. Future studies: some possible windows into the ancient skies

For future studies some particularly interesting time windows have been identified from which information about the meteorite flux would be of value:

- *Late Quaternary to recent deep-sea sediments or polar ice, 0–1 Myr ago.* Analyses of the extraterrestrial spinels from “recent” condensed pelagic clay or carbonate ooze would be a test of whether our approach gives results in accord with recent meteorite fall statistics. The spinel assemblages from micrometeorites recovered from melted Greenland or Antarctic ice (Engrand and Maurette, 1998; Suavet et al., 2010) could also represent a standard against which results for older “windows” can be compared and discussed.
- *Late Eocene interval with high extraterrestrial ${}^3\text{He}$, 35 Myr ago.* For the late Eocene a comet or an asteroid shower has been proposed based on high amounts of extraterrestrial ${}^3\text{He}$ over a stratigraphic interval representing 2.2 Myr, together with several microtektite or krystite beds and iridium anomalies (Montanari et al., 1993; Farley et al., 1998; Glass et al., 2004; Farley, 2009). Two of the largest Cenozoic impact craters, Popigai (90–100 km diameter) and Chesapeake Bay (40–90 km diameter), together with several smaller craters, formed at the same time as the ${}^3\text{He}$ anomaly developed (Montanari et al., 1998). This is also the time for climate change from the early Paleogene greenhouse climate to the present ice-house climate (Zachos et al., 2001). The first major Antarctic ice sheets formed at this time, and from then continental ices at the poles have more or less continuously grown in size. Could externally forced perturbations of the solar system have triggered a comet or asteroid shower and also affected Earth's orbit with climatic effects? According to Farley et al. (1998), enhanced ${}^3\text{He}$ and excess craters reflect a comet shower; Fritz et al. (2007) argue that ${}^3\text{He}$ originates from lunar regolith ejected by an L chondrite asteroid shower; Tagle and Claeys (2004) argue for an asteroid shower based on an L chondritic PGE signature of Popigai crater rock; and Kyte et al. (2011), based on chromium isotope measurements of the Popigai distal ejecta bed, give support for a likely asteroid belt source of the impactor. Detailed quantifications of the extraterrestrial spinels in condensed sediments from this time could give important information about the events in space.
- *Earliest Paleocene after the K-T boundary, 66–64 Myr ago.* Was the K-T boundary impactor part of a generally enhanced flux of some type of extraterrestrial material to Earth or does it represent a lone impactor? It is known from Cr isotopic studies of the K-T boundary clay that the impactor either was a comet or a carbonaceous chondrite (Kyte, 1998; Trinquier et al., 2006). Carbonaceous



Fig. 23. The upper Maastrichtian–lower Danian section at Zumaia, northern Spain. The K-T boundary is at the level where the person dressed in red stands. The lower Danian limestones are relatively condensed hemipelagic deposits. The limestones record a detailed Milankovitch cyclicity, opening up for detailed temporally constrained estimates of the flux of micrometeorites in the time after the K-T boundary.

meteorites and micrometeorites contain characteristic spinels that readily can be extracted from sediments and would represent fragments of the K-T boundary impactor (Bjärnborg and Schmitz, 2013). There are excellent condensed earliest Paleocene sections at Gubbio in Italy and at Zumaia in northern Spain (Fig. 23).

- *Middle Cretaceous, ~120–80 Myr ago.* The formation of the Tycho crater on the Moon 109 Myr ago may have ejected substantial amounts of lunar debris into Earth crossing orbits. Artemieva and Shuvalov (2008) envision that Earth was covered by meteorites from the Tycho crater with a mean density of 0.1–0.3 kg per m². This extraneous component would represent a recognizable fraction in a deep-sea sediment having formed at a typical rate of ~5 kg per m² per Kyr. This sample would perhaps also lie in the tail of the increased flux from the breakup of the large Baptistina asteroid in the asteroid belt, an event that occurred somewhere between 160 and 80 Myr ago (Bottke et al., 2007; Farley et al., 2012). A ³He anomaly possibly related to the breakup event was found in 80 Myr old strata in the Italian Apennines (Farley et al., 2012). These sediments are relatively condensed and could be a worthwhile target for searches of extraterrestrial spinels.

- *Jurassic to early Cretaceous, Ammonitico Rosso, 180–140 Myr ago.* In the Mediterranean region there are abundant outcrops of the highly condensed Ammonitico Rosso sediment. In fact, this lithology is remarkably similar in appearance to the mid-Ordovician meteorite-rich limestones from Sweden. Close in time to the Jurassic-Cretaceous boundary, 145 Myr ago, the >70 km in diameter Morokweng impact crater formed in South Africa. A fossil LL chondrite fragment of the impacting asteroid has been found in the impact suevite (Maier et al., 2006). Were LL chondrites a more common component in the flux at this time? From Ammonitico Rosso deposits in Italy, condensed hard-ground levels have been described (e.g., Castellarin et al., 1974) from which abundant cosmic melted spherules were recovered. These deposits would probably also yield abundant extraterrestrial spinel grains. There are excellent sections of the Ammonitico Rosso on the Trento Plateau, Italy; in the Adnet region, Austria, and the Betic Cordillera in southern Spain (Fig. 24).

- *Late Devonian-Griotte facies, ~375 Myr ago.* From the late Devonian to early Carboniferous, three large impact craters are known (Fig. 1; including the 52 km large Siljan crater in Sweden) and faunal turnovers are conspicuous in the late Devonian (Reimold et al., 2005). Argon isotope dating of recent H chondrites may indicate a breakup event affecting the H chondrite parent body at about this time (Swindle et al., 2009) (see Section 5). In southern France and other places in Europe a very condensed type of limestone formed in the Late Devonian, the “Griotte” pelagic, cephalopod-rich limestone, also very similar to the Ordovician Orthoceratite Limestone. There are many potential sections of condensed limestone from this period, e.g., Montaigne Noire, southern France, the Fiumara Assi section in Calabria, and Griotte Limestones of the Pyrenees and Cantabria. Spinel searches in these sediments could unravel whether the impact craters formed at this time can be tied to a collision event involving the H chondrite parent body in the asteroid belt.

- *Early Ordovician condensed sediments, ~485 Myr ago.* This sample would primarily address the question of which types of meteorites dominated the flux to Earth before the disruption of the L chondrite parent body. Today debris from the L and H chondrite parent bodies dominate the flux, but what was the situation before their disruptions? This is a major and fundamental blank spot on our map of knowledge about the solar system, and this sample is of particular interest. Excellent condensed sections of early and middle Ordovician sediments exist at many places in Baltoscandia and in central and southern China.



Fig. 24. The condensed, middle to late Jurassic, Fortuna section in the Betic Cordillera of southeastern Spain. A very condensed stratigraphic interval in this section records the Callovian-Oxfordian transition, a major event in Earth's history. Photo by V. Pujalte.

- **Snowball Earth-meteorites from the ices, ~850 to 580 Myr ago.** Probably major parts of the oceans were repeatedly covered by ices during many million years during the Snowball Earth episode. Large numbers of meteorites must have accumulated on the ices, and when ices rapidly melted, the meteorites fell to the sea floor. Bodiselitsch et al. (2005) reported an Ir anomaly at the base of the cap carbonates covering Snowball Earth tills in South Africa, but the extraterrestrial origin of the Ir still awaits confirmation. Nevertheless, searching for extraterrestrial minerals in adequate cap carbonates formed directly after the Snowball Earth event may be rewarding. Perhaps the type of extraterrestrial flux may also give clues on the reasons for the global glaciation. For example, the climate anomaly may represent the effect of a near-by passing star perturbing orbits of bodies in the solar system. This could have both affected Earth's orbit, and led to changes in the delivery of meteorites to Earth. The large Copernicus crater on the Moon formed at 800 Myr ago, and Ar–Ar evidence for several other coeval impacts on the Moon indicate it may be related to an asteroid or comet shower (Zellner et al., 2009). Possibly there was a major asteroid breakup that may have also left signals in the geological record on Earth.
- **Proterozoic and Archean sediments 3500–1000 Myr ago.** We have no knowledge at all about what types of meteorites impacted Earth during the Proterozoic and Archean. The spinel separation approach may provide the first feasible method to establish this knowledge. The main problem will be to find condensed sediments. There is no biostratigraphy that can be used for assessing sedimentation rates, and it is difficult to distinguish pelagic settings in the sedimentary record (Bose et al., 2012). The question is whether any pelagic sediments are preserved at all. A trial-and-error approach has to be adopted on different lithologies, including pilot tests on smaller samples, until the first few extraterrestrial grains are found.

4. Large spinels from different meteorite types

In order to determine the origin of extraterrestrial spinels from sediments and fossil meteorites, a thorough understanding is required of the distribution of >30 µm spinels in different meteorite types. Spinels are generally rather neglected in studies of recent meteorite finds and falls. Chromite dominates the oxide fraction of ordinary chondrites, in enstatite chondrites oxides are rare, and in R chondrites chromian spinel dominates. In carbonaceous chondrites a wide range of oxide minerals occur, e.g., magnetite, chromite and different spinel group varieties (Rubin, 1997). In the silicate inclusions of iron meteorites, chromite is found, but its composition is more variable and complex than in ordinary chondrites (Bunch et al., 1970). Achondrites and mesosiderites contain chromite with large grain-to-grain compositional variability, whereas pallasites contain chemically homogeneous chromite grains that tend to be larger (up to 1–3 mm large) than in other meteorite types (Bunch and Keil, 1971).

In order to use sedimentary extraterrestrial spinels to reconstruct the relative proportions of different types of meteorites and micrometeorites over time in the sedimentary record, it is necessary to establish a detailed database quantifying the abundances and detailed proportions of various types of spinel minerals in the meteorites that are falling now. Dissolving a meteorite in hydrofluoric acid will give a relatively good representation of the spinel mineral assemblage that will survive extensive weathering on the sea floor as well as subsequent recovery through acid dissolution of sediments (Bjärnborg and Schmitz, 2013). This has been shown by comparing the HF-dissolved residues of recent L chondrites with the spinel assemblages of fossil L chondrites in mid-Ordovician limestone. The CM carbonaceous chondrites make up an important

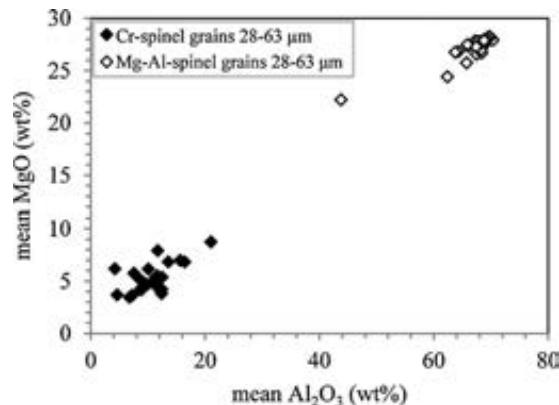


Fig. 25. In CM2 meteorite Acfer 331 there are two major populations of spinel grains that may survive when a CM2 meteorite weathers on a sea floor. The opaque, black Cr-rich spinels are distinctly separated from the transparent, colorless, pink to red or blue Mg-Al spinels. Each of the major populations, can be divided into smaller groups based on heterogeneity and Cr-content.

From Bjärnborg and Schmitz (2013).

fraction of the micrometeorite flux to Earth today (Gounelle, 2011; Taylor et al., 2012), and the same probably applies to the past, hence it is crucial to quantify whether it is likely to recover spinels from this fraction of the flux in the ancient sediments. Bjärnborg and Schmitz (2013) dissolved eight gram of the Acfer 331 CM2 carbonaceous chondrite in hydrofluoric acid in order to quantify the abundance of >28 µm durable spinels. The results showed that large spinels are surprisingly common in CM2 meteorites. Both common transparent Mg-Al and opaque chrome spinels were found (Fig. 25) (see also, Simon et al., 1994). As regards Mg-Al spinels, Acfer 331 contains on average 4.6 grains per gram in the 63–250 µm fraction and 130 grains in the 28–63 µm fraction. Black, opaque chrome spinel grains are absent from the >63 µm fraction, but in the 28–63 µm fraction, 65 grains per gram were found (Fig. 26). The Mg-Al spinel grains can be further divided into two groups, the Cr-rich, commonly pink or red, and the Cr-poor, colorless or blue, grains (Fig. 27). The Mg-Al spinels are also characterized by significant intragrain heterogeneity and abundant inclusions of e.g. diopside and Fe-Ni metal (Fig. 28). The content of spinels in the smaller size fraction, 28–63 µm, of CM meteorites is comparable at the order of magnitude level to the content of >63 µm sized chromite grains in ordinary chondrites. Thus it should be possible to recover CM spinels from ancient sediments, but the smaller size fraction of the acid residue should be searched.

A data base with spinel information similar to that for Acfer 331 should be established for all the important meteorite groups. In order to rule out terrestrial origins of recovered spinels, data bases for the distribution and composition of spinels in terrestrial rocks are also required; a very useful such data base has been produced by Barnes and Roeder (2001). In a sedimentary assemblage of mixed spinel grains from many different meteorite types, oxygen isotope analyses may contribute complementary information about the detailed origin of individual grains. Thus the proportions of the different types of meteorites in the flux at a particular time and their variations with time could be determined in great detail.

5. Parent body breakup events and meteorite flux

Changes in the micrometeorite flux to Earth may primarily relate to perturbations of the orbits of solar-system bodies and/or to asteroid or comet breakup events. Other factors may be changes in the Near Earth Asteroid population or nearby passages of major comets. For a start, the spinel approach may give a refined understanding of the sequence of major breakups of meteorite parent

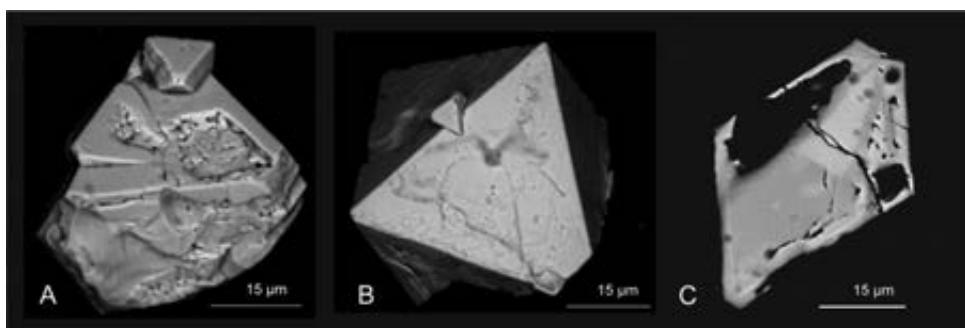


Fig. 26. The opaque Cr-spinel grains (28–63 μm) from the CM2 meteorite Acfer 331 are mostly (A) angular, but (B) euhedral, octahedral grains are also common. (C) Most Cr-rich spinel grains are heterogeneous in composition and some show distinct zoning in BSE image. The opaque Cr-spinels can be identified by narrow ranges of maximum TiO_2 (0.6–1.6 wt%) and V_2O_3 (0.5–1.0 wt%) contents.

From Bjärnborg and Schmitz (2013).

bodies through the ages. The disruptions may either be random events or reflect larger disturbances of the solar system, such as perturbations of the Oort comet cloud, triggering a comet shower to the inner solar system (Perryman, 2009). The sequence of major

asteroid breakup events has been reconstructed so far mainly based on the K-Ar (or Ar-Ar) ages of recent meteorite finds and falls (for reviews, see Keil et al., 1995; Bogard, 1995, 2011). The K-Ar ages of recently fallen meteorites span the entire age of the solar system, and measure the gas retention age i.e., the time when the K-Ar radiometric clock is reset to zero by diffusion of argon from internal parent body metamorphism or impact heating. Most meteorite types have K-Ar ages dating back to the early solar system, with ages generally older than 3.4 Gyr, and in many cases older than 4 Gyr. The ordinary chondrites, on the other hand, show an interesting bimodal distribution with impact-reset ages of either <1.5 Gyr or >3.4 Gyr (Figs. 29 and 30) (Bogard, 1995, 2011; Swindle et al., 2009, 2013). The common gas retention ages of >3.4 Gyr for ordinary chondrites and all other meteorite types most likely reflect the more intense bombardment in the early solar system, as shown also for the surface of the Moon, where most large craters date from this period. The common gas retention ages of <1.5 Gyr (and mostly <1 Gyr) for all three groups of ordinary chondrites represent a major mystery, as discussed in detail by Bogard (1995, 2011). Why do only the three ordinary chondrite groups, and no other meteorite type, show these young gas retention ages? A possibility would be that there has been an enhanced bombardment the last 1 Gyr in the region where the ordinary chondrite parent bodies reside.

The K-Ar dating of the parent body disruptions give often rather imprecise ages. For a long time, the breakup of the L chondrite parent body was thought to have happened at about 500 Myr ago (Keil et al., 1995; Haack et al., 1996), but this age estimate was based on data with a spread from ~420 to 580 Myr ago, reflecting the complexity of the K-Ar approach. The main problem is that meteoritic material may not be completely degassed upon impact, yielding too large ages for the breakup events. With the new spinel approach the signatures of large breakup events can be located at very precise levels in Earth's sedimentary record. Using conventional bio- and magnetostratigraphy and the Geological Time Scale (e.g., Gradstein et al., 2012), this level can be given an absolute age with a resolution on the order of one to a few Myr. The precision of the relative age, i.e., compared to other events in the geological record, is even higher because the time of the breakup event can be defined even to a single bed in the strata. This implies a relative time resolution of the order of 0.1–10 Kyr, at least for dating the arrival of the first material to Earth from a breakup event. It now appears from studies of EC grains in Ordovician limestone that the L chondrite breakup can be defined to a level close to the base of the *L. variabilis* Conodont Zone in the Darriwilian Stage (see Section 2.3). According to the latest Geological Time Scale this corresponds to ~466 ± 1 Myr ago (Gradstein et al., 2012). It is compelling that if one reads the pre-1996 literature on the L chondrite breakup event, before the first reports on abundant fossil L chondrites appeared (Schmitz et al.,

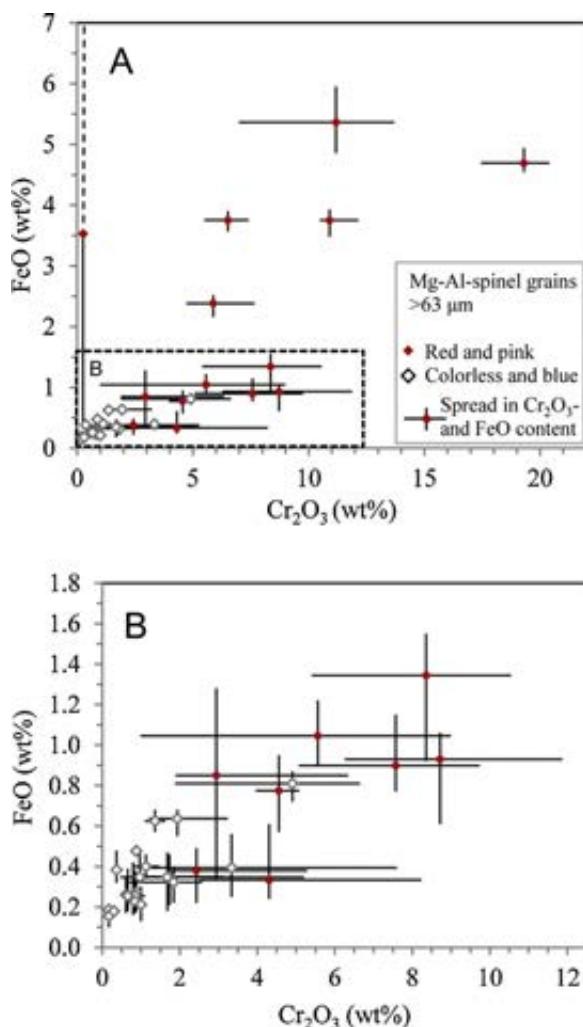


Fig. 27. (A) In the CM2 meteorite Acfer 331 the pink and red Mg-Al spinel grains have generally a higher content of Cr and Fe than the colorless grains. The plot shows wt% FeO versus wt% Cr_2O_3 for separated grains $>63 \mu\text{m}$. The spot indicates the mean content whereas the vertical and horizontal lines show the spread in content measured within the grain. (B) Enlargement of area B showing the grains with lower Cr_2O_3 and FeO contents.

From Bjärnborg and Schmitz (2013).

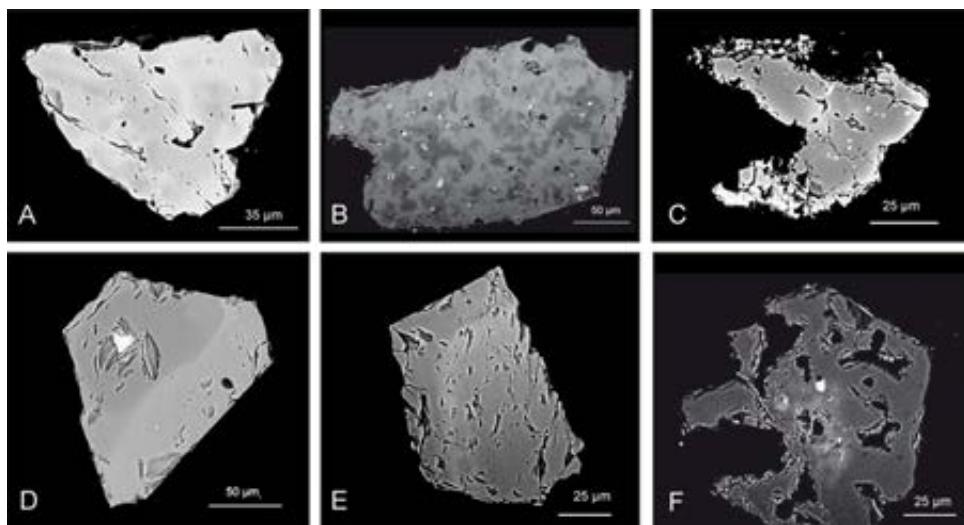


Fig. 28. Most Mg-Al spinels $> 63 \mu\text{m}$ from the CM2 chondrite Acfer 331 are heterogeneous in composition and have abundant inclusions. Here SEM-backscattered electron images show the variations in polished sections, where areas with lower concentrations of heavy elements, mainly Fe and Cr, are darker. (A) Patchy heterogeneity with smooth transitions. (B) Patchy heterogeneity with more distinct transitions. Grain has abundant inclusions, probably diopside. (C) This grain is Cr-homogeneous, but the Fe-content increases toward grain boundaries and cracks. Small, rounded bright inclusions of presumably perovskite. (D) Grain with two distinct zones of different Cr content. The grain also has a triangular bright inclusion, probably of diopside. (E) Chevron-zoned grain with alternating bands of higher/lower Cr content. (F) Cr-poor grain with a patchy center of higher Cr-content, and small rounded inclusions of varying composition, including most likely diopside and Fe-Ni metal.

From Bjärnborg and Schmitz (2013).

1996), some of the best isotopic analyses already then indicated a breakup age of around 470 Myr ago, rather than the then generally accepted age of ~ 0.5 Gyr. Bogard (1995) highlights two particularly carefully executed studies, one of the Chico L6 impact melt breccia (Fujiwara and Nakamura, 1992; Bogard et al., 1995) and another of the shocked Peace River L6 chondrite (McConville et al., 1988). A sample from the interior of the Chico impact melt dike gives a Rb-Sr isochron age of 467 ± 15 Myr ago, whereas the Ar-Ar ages of several samples of melt and unmelted parts of Chico indicate an excess ^{40}Ar component, yielding Ar-Ar ages higher than the Rb-Sr ages by 10–15%. McConville et al. (1988) determined an Ar-Ar resetting age of 450 ± 30 Myr ago of the Peace River chondrite, based on both whole rock samples and spots degassed with laser from shock-glass veins. Recently, some groups have readdressed the Ar-Ar approach to date the L chondrite parent body breakup. By using refined techniques to identify and remove trapped Ar components in several L chondrites, an age of 470 ± 6 Myr ago was established for the breakup (Korochantseva et al., 2007). In another careful Ar-Ar study of the NWA 091 shocked L6 meteorite, including Cd-shielding during neutron irradiation, the breakup age was determined to 475 ± 6 Myr ago (Weirich et al., 2012). Apparently, the discovery of the L chondrite grains in the mid-Ordovician sediments has provided a powerful means to calibrate the K-Ar approach to date ancient breakup events.

The K-Ar ages of H chondrites, which are as abundant as the L chondrites in the present day flux to Earth, have recently been dealt with in greater detail (Swindle et al., 2009; Bogard, 2011). Included in the data set are now also many shock darkened H chondrites. The H chondrites show a clear bimodal pattern with ages of either > 3.4 Gyr or < 1.5 Gyr, and all but two of the latter plot at < 1 Gyr. It is clear that many heavily shocked H chondrites were affected by one or several major events in the past 1 Gyr, but the pattern is much muddier than for the L chondrites dominated by the event at ~ 470 Myr ago. The three highest peaks in the H chondrite Ar-Ar plot are at 280, 350 and 460 Myr ago (Fig. 30). The peaks may be reflections of one large impact event with only partial resetting recorded in some of the samples analyzed. Swindle et al. (2009) argue that the presently best estimate may be two thermal events affecting the parent body (or bodies) of recent H chondrites at ~ 300

and 450–500 Myr ago. The reality of an event 450–500 Myr ago, however, is not supported by the data for fossil micrometeorites and meteorites from this time. These assemblages, based on oxygen isotopes and elemental composition (see Section 2), appear to be completely dominated by L chondrites, without a single confirmed H chondritic contribution so far. The crucial message here, however, is that any hypothesis about breakup events based on K-Ar measurements of meteorites can now be tested by searches for spinel grains in condensed sediments. Our ongoing studies of condensed sections of Frasnian-Fammenian age (372 Myr ago) in southern France, although very preliminarily, show a 1:1 distribution in H versus L chondrite micrometeorites (based so far on only 13 recovered EC grains), similar to the distribution in today's flux of meteorites. This may indicate either that a 1:1 relation has been the norm throughout much of the late history of the solar system, interrupted only by short term aberrations like after the breakup of

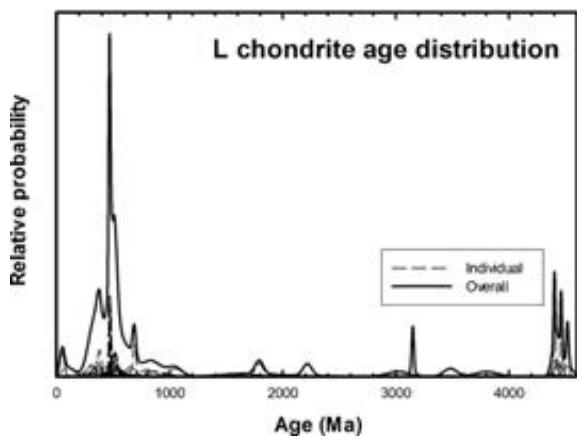


Fig. 29. K-Ar ages of recent L chondrites, from Swindle et al. (2013). The plot shows individual probability distributions for ages of individual meteorites (dashed line) and a combined probability distribution (solid line) for all of the data. The diagram takes into account the uncertainties in the individual data points in a graphical way by giving each data point equal area. There is a clear bimodal distribution in the impact resetting ages of the L chondrites. See further Swindle et al. (2013).

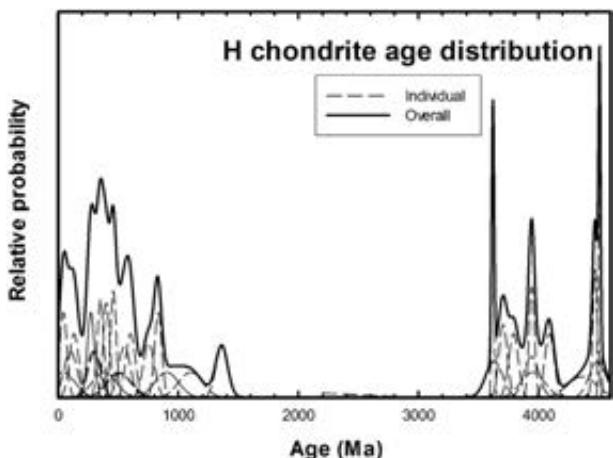


Fig. 30. K-Ar ages of recent H chondrites, from Swindle et al. (2013). The plot shows individual probability distributions for ages of individual meteorites (dashed line) and a combined probability distribution (solid line) for all of the data. The diagram takes into account the uncertainties in the individual data points in a graphical way by giving each data point equal area. There are essentially no impact resetting events recorded for H chondrites between 1.3 and 3.5 Gyr. See further Swindle et al. (2013).

the L chondrite parent body. Alternatively, the results can be reconciled with an H chondrite breakup event somewhere between 450 and 375 Myr ago. More detailed studies, however, are required before any firm conclusions can be reached on this matter.

The LL chondrites show the same bimodal K-Ar ages, with ages either older than 3.4 Gyr, or younger than 1.3 Gyr (Dixon et al., 2004; Swindle et al., 2013). Partly because of their greater rarity in the flux today, fewer analyses have been performed on LL chondrites. An age of 625 ± 163 Myr was measured on a single multi-grain aliquot from the (well preserved) fossil LL meteorite fragment recovered from a drill core into the Morokweng impact crater (Jourdan et al., 2010). Swindle et al. (2013) compiled data for two Antarctic LL chondrites, one with a Rb-Sr isochron age of 1.197 ± 0.054 Gyr and a slightly higher Ar-Ar age, ~1.26 Gyr, and the other having a Rb-Sr age of 1.270 ± 0.190 Gyr. Weirich et al. (2009) Ar-Ar dated two shocked LL chondrites and obtained scattered values in the range 900–1250 Myr ago (see also, Swindle et al., 2011). Again it cannot be said with any certainty whether the spread over the total range for LL chondrites from ~600 to 1300 Myr ago reflects partial retention of Ar and one single event in this time interval, or several major impacts on the LL chondrite parent body (or bodies) over this interval.

For many recent ordinary chondritic meteorite finds and falls cosmic ray exposure ages have also been determined giving information on the time an object has traveled in space as a small ($<\sim 1$ m) object. The cosmic ray exposure ages rarely coincide with K-Ar ages, for example, many L chondrites have K-Ar ages of ~470 Myr and cosmic ray exposure ages of ~40 Myr. The typical cosmic ray exposure ages for H chondrites lie around 7–8 Myr, and 15 Myr for LL chondrites (Wieler and Graf, 2001). The discrepancy between K-Ar and cosmic ray exposure ages reflects that the latter only represent smaller breakup events. For example, today's L chondrite finds and falls may originate from a kilometer-sized asteroid that was blasted off a 200 km-sized L chondrite parent 470 Myr ago, but then broke up into smaller pieces and dust because of a minor collision 40 Myr ago. Judging from the characteristic groupings of cosmic ray exposure ages of the ordinary chondrites, these smaller breakup events are crucial in determining which types of meteorites that reach Earth at a particular time. There must have been many such smaller breakup events through Earth's history, and although they would not lead to the same dramatic change in the meteorite flux as one of the major K-Ar resetting events, it

is likely that a pulse of micrometeorites of a specific type would follow in the wake of such an event. Probably many such smaller breakup events are represented in Earth's sedimentary record by changes in the extraterrestrial spinel component.

There is a conundrum underlying our present understanding of changes in the extraterrestrial dust flux to Earth through the past 66 Myr. Detailed searches for extraterrestrial ^3He in the Cenozoic sediment record have located two episodes with enhanced fluxes of interplanetary dust particles, one in the late Miocene at around 8 Myr ago, and the other in the late Eocene at ~35 Myr ago (Farley, 2009). The 8 Myr dust event has been tied to the breakup of the asteroid that formed the Veritas family (Farley et al., 2006). This is an event estimated to have happened 8.3 ± 0.5 Myr ago, based on tracking of the orbits of Veritas family members backwards in time to their source (Nesvorný et al., 2003). With the prominent 7–8 Myr peak in the cosmic ray exposure ages of recent H chondrite falls and finds, one would expect the original Veritas asteroid to be the parent body of the H chondrites, but spectral data do not support this. The Veritas family is of low-albedo, primitive, C or D type taxonomy (Di Martino et al., 1997), not S type, which is thought to represent the ordinary chondrites. Neither do the recent H chondrites show any K-Ar resetting that would be a likely effect of a major collision like the one that formed the Veritas family. Spinel searches in 6–8 Myr old sediments could possibly unravel what type of extraterrestrial matter reached Earth from the Veritas breakup as well as the exact timing of a smaller H chondrite breakup relative to the Veritas event. The situation is similar for the Late Eocene event, where a ^3He anomaly extends over 2.2 Myr, and were two major impact signatures, from the Popigai and Chesapeake Bay impactors, are recorded (see Section 3.2). Kyte et al. (2011), based on Cr-isotopes, relate the 35 Myr dust and impactors to the formation of the Brangäne asteroid, with a 50 ± 40 Myr age based on orbits. This is an S type asteroid family likely made up of ordinary chondrites. Kyte et al. (2011) also note a peak in cosmic ray exposure ages for H chondrites in the interval 36–33 Myr ago and suggest that the 35 Myr event relates to an enhanced flux of H chondritic material to Earth. Trying to understand the 35 Myr dust event, Schmitz et al. (2009) searched for extraterrestrial spinels in the ^3He rich interval of the Late Eocene at Massignano, but found only one ordinary chondritic grain in 167 kg sediment (see Section 3.1). The 35 Myr event must have been orders of magnitudes smaller than the 470 Myr event that resulted in common chromite grains from micrometeorites in the sediments. The mid-Ordovician event has been associated with either the ~6000 member Flora asteroid family, or the ~1000 member Gefion family, which resulted from two of the largest documented breakup events during the past billion years (Nesvorný et al., 2007, 2009; Kyte et al., 2011). It appears that the effects of major breakup events on the flux of material to Earth is also strongly dependent of where in relation to particular orbital resonances the breakup occurs (Zappalà et al., 1998).

6. Perturbations in the asteroid belt and effects on Earth

That an extraterrestrial event at the K-T boundary had a crucial effect on the evolution of life can be concluded from more than thirty years of intense research on this event (e.g., Alvarez et al., 1980; Schulte et al., 2010). The main evidence is the precise coincidence in the stratigraphic record between the worldwide occurrence of an iridium-rich ejecta layer and the last appearance of the typical Cretaceous fossil fauna. In the following, some coincidences between events in the asteroid belt and on Earth will be discussed. These coincidences, admittedly, may just be coincidences, but as Miss Marple, the detective hero of Agatha Christie's novels once said: "Any coincidence is always worth noting. You can always throw it away later if it is only a coincidence". The

main point here is to show how the spinel approach can potentially tie the histories of Earth and the astronomical realm to each other.

The long period from ~3.4 to ~1 Gyr ago when no K-Ar gas resetting impacts appear to have occurred in the asteroid belt reflects tranquil conditions in at least the parts of the asteroid belt where the parent bodies of the ordinary chondrites reside. At least the latter part of this period is known also as a very tranquil period in Earth's history. The one billion years from ~1.85 to 0.85 Gyr, are named the Intermediate Ocean, but also, more sardonically, the "Boring Billion" (Hazen, 2012), because so little appears to have happened in terms of evolution of life, and no obvious major environmental perturbations such as ice ages or major impacts are known on Earth from this long time interval. During the entire period single-celled eukaryotes represented the highest form of life on Earth. Then around ~0.85 Gyr ago, the Earth enters, what could be called the "Dynamic 0.85 Gyr", starting with the ~200 Myr of repeated Snowball Earth glaciations, the Cryogenian Period from ~0.85 to 0.64 Gyr ago, and the concurrent emergence of multicellular animal life (Smith, 2009). The most recent 0.85 Gyr of Earth's history are characterized by a spectacular evolution of animals from single-celled eukaryotes to today's humans, and at least four major ice age episodes, Snowball Earth, late Ordovician, Carboniferous-Permian, and the late Cenozoic, the one we live in (Fig. 1). During the period also many large impact craters formed both on Earth and the Moon. The ice age before the Snowball Earth glaciations dates back as far as to 2.4 Gyr ago, the Huronian glaciation, which has left a record primarily in North America. Only one large crater, the 30 km diameter and 1.63 Gyr old Shoemaker crater in Australia, is representing the Boring Billion years on Earth (Grieve, 2001). The large, 250 km diameter Sudbury and 300 km diameter Vredefort craters are dated to 1.85 and 2.0 Gyr ago, respectively. On the other hand, the onset of the Cryogenian Period appears to coincide with an asteroid shower on the Earth-Moon system as evidenced from common and widely distributed lunar impact glasses with K-Ar ages around 0.8 Gyr ago (Zellner et al., 2009). One of the Moon's most prominent "young" craters the 93 km diameter Copernicus crater also formed at 0.8 Gyr ago (Eberhardt et al., 1973; Bogard et al., 1994). The common Moon impacts and the weakly defined K-Ar ages of the LL chondrites in the range 1.2–0.6 Gyr ago may be related to an LL chondrite parent body breakup and accompanying asteroid shower at around the time of the Snowball Earth glaciations. Another coincidence that may indicate an extraterrestrial forcing is the ca. 250–300 Myr recurrence seen for the three past major ice ages (Fig. 1), which is not too different from the estimated period of ca. 225–250 Myr for the Sun's orbit around the galaxy (Perryman, 2009).

The next crucial event in the history of life is the emergence of shelled animals at the base of the Cambrian ~540 Myr ago, but biodiversity remained low, with comparatively few species, until the Great Ordovician Biodiversification Event (GOBE) in the mid-Ordovician at ~470–450 Myr ago. This is one of the most important events in the history of life. At the genera level, it represents an even more prominent change in biodiversity than some of the later major extinction events, but the GOBE is primarily a "mass origination" event. It represents the most intense phase of species radiation during the Paleozoic and led to irreversible changes in the faunas of Earth's sea floors. For the GOBE there is now data indicating a close temporal coincidence with the 466 Myr ago breakup event of the L chondrite parent body (Schmitz et al., 2008). Bed-by-bed studies of the fossil fauna in the mid-Ordovician sedimentary strata indicate that the GOBE takes place over the ~20 Myr in the immediate aftermath of the breakup of the L chondrite parent body. The period is characterized by an at least two orders of magnitude increase in the flux of meteorites to Earth (Schmitz et al., 2008). Many small craters (~3–30 km in diameter) are known on Earth from this

time interval (Grieve, 2001), but one or a few larger craters may have been destroyed or are covered by sediments. In Baltoscandia the first EC grains from the breakup of the L chondrite parent body occur at the same level in the strata where the main phase of the diversification of brachiopod species starts (Schmitz et al., 2008). The origination event is preceded by an extinction event, but because of the relatively low number of brachiopod species before the GOBE, this extinction event does not stand out in the record. All the succeeding major mass extinction events during the Phanerozoic are actually coupled extinction-origination events, but most focus has been directed to the extinction phase. In order for a new fauna to invade, the old fauna must first be removed. Perhaps the best evidence for the eradication of the dinosaurs by the Chicxulub impactor comes from the rapid diversification of mammal species after the K-T boundary (Fastovsky and Sheehan, 2005). The 20 Myr after the breakup of the L chondrite parent body is also characterized by prominent volcanism and dramatic tectonic reorganizations (Thompson et al., 2012). Whether the relations described here simply reflect random coincidence, or if there is a common cause for extraterrestrial and terrestrial perturbations, requires further investigations.

In the late Ordovician, after the GOBE has stabilized, there is a minor glaciation event, and towards the end of the Ordovician one of Earth's five major mass extinction events occur, but no strong evidence for any coeval perturbations in the asteroid or comet flux are known. The next major event on Earth is in the late Devonian, at ~372 Myr ago with another of the big mass extinctions, at the Frasnian-Famennian stages boundary. The late Devonian-early Carboniferous is also a time when three large impact craters formed, the Charlevoix (54 km), Siljan (52 km), and Woodleigh (40 km) craters (Fig. 1). Not long after the late Devonian mass extinctions the Earth enters into the long (~80 Myr) and severe Carboniferous-Permian glaciation. The next major event, at the Permian-Triassic boundary, 252 Myr ago, is the largest mass extinction event in Earth history, but no evidence of extraterrestrial forcing exists. The mass extinction event at the K-T boundary 66 Myr ago, is clearly related to an extraterrestrial event, and it remains to be established whether the K-T impactor possibly relates to a breakup of a major carbonaceous chondritic parent body. It is notable that the onset of the cooling leading to today's ice age, coincides rather precisely (on a geological scale) in the Late Eocene 35 Myr ago with the period of enhanced ${}^3\text{He}$ in sediments, and formation of two unusually large craters on Earth, Chesapeake Bay and Popigai.

There are other data and facts that may concur with a period of enhanced impacts and perturbations of the orbits of the bodies of the inner solar system during the past ~0.85 Gyr. The combined age data from lunar spherules, lunar impact melts and lunar craters indicates that after ~3.2 Gyr ago the impact rate on the Moon may have been stable within a factor of two, except for a peak during the last 0.5 Gyr or so (Culler et al., 2000; Levine et al., 2005; Hartmann et al., 2007). This peak is mainly seen in the data for the smaller craters, and the largest peak in K-Ar ages for lunar spherules is at 470 Myr ago, reflecting most likely that the spherule collections are weighted to representing smaller craters.

At least three major hits on the three ordinary chondritic parent bodies from ~1 to ~0.3 Gyr would probably require significant perturbations of orbital parameters in the parts of the inner solar system where the increase of cratering occurs. The increase in frequency of K-Ar resetting impacts on asteroid bodies are likely to be accompanied by comparable variations in the flux of impactors that hit the inner planets and our Moon or a general increase in flux of bodies of a size that can reset portions of meteorite parent bodies without destroying the bodies (Swindle et al., 2009). The latter fact can explain the protracted range of K-Ar ages for ordinary chondrites in case they do not just reflect three major impact events as favored here.

It is remarkable that in the past 110 Myr there are many very large craters on the Earth-Moon system, Tycho on the Moon, and e.g., Chicxulub, Popigai, Chesapeake Bay, Kara-Kul and Kara on Earth (Fig. 1). One can note that the eminent astrogeologist, the late Eugene Shoemaker in his last and unfinished paper, in his last paragraph writes: "For a number of years I have suspected that the long-term average cratering rate may have increased late in geological time, perhaps as much as a factor of two. Hints of this increase comes from comparison of the terrestrial cratering rate estimated from the present flux of asteroids and comets and from the Phanerozoic crater record of North America and Europe with the 3.2 Ga crater record of North America and Europe with the 3.2 Ga crater record of the Moon. The number of small craters on the rim deposits of the large young lunar craters Copernicus and Tycho also suggest a late increase in the cratering rate" (Shoemaker, 1998; see also McEwen et al., 1997).

It is possible that parent body breakups as well as Earth glaciations relate to astronomical disturbances of the inner solar system during the past 0.85 Gyr. The main point made here, however, is that the spinel approach can give new and crucial quantitative proxy data that can resolve issues about the frequency of impacts in the inner solar system and the precise timing and sequence of asteroid breakup events, both smaller ones that reset cosmic ray exposure ages, and larger ones that reset K-Ar ages. Earth's cratering record is very incomplete, only a percent of all craters formed are known and for only a fraction of these do we know the precise impact age and/or the type of impactor involved (Jourdan, 2012). Any inferences based on such a data set will be very uncertain. In Earth's sedimentary record, at least for the last ~0.6 Gyr, there are traces of most or all major asteroid and comet impacts on Earth, as well as the different micrometeorites that reached Earth at different times. The problem is that the "haystack" in which the extraterrestrial signatures, "the needles", are hidden is so large.

7. Potential perturbations of the solar system

Much has been written on possible perturbations of the solar system in the longer term (>10 Myr) geological perspective. The literature, however, tends to be speculative and many of the proposed scenarios lack adequate empirical verification by proxy approaches. It is beyond the scope of this paper to give more than a brief summary of the astronomical processes at stake. The most commonly discussed processes include near-by encounters with stars, passages through interstellar molecular or dust clouds, or unusual planetary alignments within the solar system (e.g., Varadi et al., 1999, 2003; Perryman, 2009). There are also processes on a larger astronomical scale, such as galactic gravity waves and galactic cannibalism, for which the potential effects on the solar system are not well understood. There are major factors, such as dark matter and dark energy, or the expansion of the universe, that we do not understand well, so it appears likely that there are also unknown processes that may have affected the solar system.

The Oort Cloud of comets is inferred to surround the solar system to distances on the order of 10^4 – 10^5 AU. It consists of some 10^{12} – 10^{13} comets with a total mass about ten times that of Earth (Oort, 1950; Weissman, 1996; Perryman, 2009). Perturbations of the Oort cloud by nearby encounters with random passing stars has been suggested to trigger comet showers to the inner solar system. This would lead to an increase also in the cometary micrometeorite flux to Earth, but the insufficient knowledge about cometary spinels would make it difficult to locate a cometary spinel spike in the sedimentary strata. Cometary showers could also have indirect effects, such as increasing the flux of micrometeorites from inner solar system bodies having been hit by comets. A comet shower could also suddenly fill up the space up to ~30 AU from the sun, within the

orbital radius of Neptune, with new comets replacing those that have been expelled from the solar system by the outer planets. This would increase the new-comet rate by a factor of ~40 (Frogel and Gould, 1998). Near encounters with stars that can perturb the Oort cloud is a feasible process in the geological time perspective as shown by the Hipparcos satellite data. García-Sánchez et al. (1997, 1999) identified 1194 stars that could pass close enough to significantly perturb the Oort cloud, i.e., within 2–3 pc. In recent space, within a time framework of ± 10 Myr, the object Gliese 710 has a predicted closest approach of less than 10^5 AU, ~0.5 pc, which would lead to a minor comet shower, but increasing the cratering rate by only 5% (García-Sánchez, 2000; García-Sánchez et al., 1997, 2001). In the 100–1000 Myr perspective, however, more dramatic comet showers most likely have occurred.

Other factors that potentially can perturb the Oort comet cloud are giant molecular clouds, the galactic gravitational field, and supernova shock waves (Perryman, 2009). In the 1980s and 90s there was an extensive discussion in the literature on whether the undulating movement of the solar system perpendicularly to the galactic equatorial plane could cause periodic comet showers at ~30 Myr intervals to the inner solar system (e.g., Hills, 1981; Rampino and Stothers, 1984; Hut et al., 1987; Rampino, 1998; Stothers, 1998). At the time it was generally held that many, if not most, large craters on Earth were related to comet impacts (e.g., Shoemaker, 1998; Stothers, 1998). The periodic comet hypothesis was attractive in its simplicity, tying the astro- and the geobiosphere together, but it has not withstood the test of time. Large uncertainties in the crater ages and the very fragmental crater record preclude robust conclusions about cratering periodicities. For some of the largest known craters (e.g., Morokweng, Popigai, Chicxulub), the data now tend to support an asteroidal rather than a cometary impactor (but then remains the issue that some or many comets may be very similar to asteroids in their composition). Recent evaluation of data from the Hipparchos satellite indicate that the passage through the equatorial plane may rather take place every ~41 Myr (Perryman, 2009). The galactic tide has been proposed to be the dominant cause of orbital evolution in the Oort cloud, inducing a loss rate of comets by about a factor 1.5–2 larger than due to stellar perturbations (Heisler and Tremaine, 1986). Adiabatically varying galactic tide should deliver comets individually, whereas a close molecular cloud encounter should trigger a comet shower. Passages of the solar system through the galactic spiral arms have been thought to affect climate and life on Earth by changes in the cosmic ray flux (Gies and Helsel, 2005; Svensmark, 2012). Upon entering the spiral arms shock fronts may perturb the solar system. Astronomical simulations further indicate that the dynamics of the solar system is chaotic, and unusual planetary alignments may perturb orbits of bodies over regions of the solar system at times (Varadi et al., 1999, 2003). If it turns out that most craters on Earth have formed by bodies from the main asteroid belt, this may indicate that the planetary orbital evolution is a dominating factor. So far much of this research is based on theoretical speculations and modeling work. Careful studies of the extraterrestrial fraction of Earth's sedimentary record may provide the empirical basis on which a more robust understanding of how the history of life on Earth and cosmos are intertwined.

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