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Meteorite flux to Earth in the Early Cretaceous as reconstructed from sediment-dispersed extraterrestrial spinels

Birger Schmitz[1,2]*, Philipp R. Heck[2], Walter Alvarez[3,4], Noriko T. Kita[5], Surya S. Rout[2], Anders Cronholm[1], Céline Defouilloy[5], Ellinor Martin[1], Jan Smit[4,6], and Fredrik Terfelt[1]
[1]Astrogeobiology Laboratory, Department of Physics, Lund University, SE-22100 Lund, Sweden
[2]Robert A. Pritzker Center for Meteoritics and Polar Studies, The Field Museum of Natural History, Chicago, Illinois 60605, USA
[3]Department of Earth and Planetary Science, University of California, Berkeley, California 94720, USA
[4]Osservatorio Geologico di Coldigioco, Contrada Coldigioco 4, 62021 Apiro, Italy
[5]WiscSIMS, Department of Geoscience, University of Wisconsin–Madison, Madison, Wisconsin 53706, USA
[6]Department of Sedimentary Geology, Vrije Universiteit, 1081 HV Amsterdam, Netherlands

ABSTRACT

We show that Earth's sedimentary strata can provide a record of the collisional evolution of the asteroid belt. From 1652 kg of pelagic Maiolica limestone of Berriasian–Hauterivian age from Italy, we recovered 108 extraterrestrial spinel grains (32–250 µm) representing relict minerals from coarse micrometeorites. Elemental and three oxygen isotope analyses were used to characterize the grains, providing a first-order estimate of the major types of asteroids delivering material at the time. Comparisons were made with meteorite-flux time "windows" in the Ordovician before and after the L-chondrite parent-body breakup. In the Early Cretaceous, ~80% of the extraterrestrial spinels originated from ordinary chondrites. The ratios between the three groups of ordinary chondrites, H, L, LL, appear similar to the present, ~1:1:0.2, but differ significantly from Ordovician ratios. We found no signs of a hypothesized Baptistina LL-chondrite breakup event. About 10% of the grains in the Maiolica originate from achondritic meteorite types that are very rare (<1%) on Earth today, but that were even more common in the Ordovician. Because most meteorite groups have lower spinel content than the ordinary chondrites, our data indicate that the latter did not dominate the flux during the Early Cretaceous to the same extent as today. Based on studies of three windows in deep time, we argue that there may have been a gradual long-term (a few hundred million years) turnover in the meteorite flux from dominance of achondrites in the early Phanerozoic to ordinary chondrites in the late Phanerozoic, interrupted by short-term (a few million years) meteorite cascades from single asteroid breakup events.

INTRODUCTION

Much knowledge about the history of life, climate, tectonics, magnetic polarity, and chemistry of seawater has accumulated during the past two centuries from studies of Earth's sedimentary strata. With the discovery of the asteroid impact at the Cretaceous-Paleogene (K-Pg) boundary and its effects on life (Alvarez et al., 1980), an interest has grown in integrating astronomical and geological perspectives. A new approach that can relate ancient events in space to coeval events on Earth is the search for relict spinel minerals from micrometeorites and meteorites in condensed sediments (Schmitz, 2013). The method has so far been primarily applied in reconstructions of the Ordovician L chondrite parent-body breakup (LCPB), the largest documented collisional event in the asteroid belt in the past 3 b.y. (Schmitz et al., 2003). This breakup probably led to the formation of one of the major asteroid families (Nesvorný et al., 2009). The event has been dated by ^{40}Ar-^{39}Ar analyses of recently fallen L chondrites to 470 ± 6 Ma (Korochantseva et al., 2007), but the most precise relative date is given by an abrupt two-orders-of magnitude increase worldwide in sand-sized L-chondritic chromite grains in mid-Ordovician sediments (Schmitz et al., 2003; Heck et al., 2016).

By dissolving 100-kg-sized samples of condensed sediments from different time periods in various acids, the highly refractory extraterrestrial spinel minerals can be concentrated. The recovered grains typically contain high concentrations of solar-wind noble gases, indicating that they dominantly represent fragments of coarse micrometeorites (Heck et al., 2008). Pelagic carbonates are the best material for sampling the population of extraterrestrial chromite grains, because of their low content of detrital minerals that obscure the extraterrestrial fraction and the ease with which they can be dissolved in acid. As a part of a larger effort to create "windows" into the meteorite flux to Earth at different times during the Phanerozoic (Schmitz, 2013), we focus here on a part of the Lower Cretaceous (145–133 Ma) Maiolica limestone in central Italy. This pelagic limestone is exceptionally "clean," i.e., having very low contents of terrestrial, detrital mineral grains, making it useful for reconstructions of the micrometeorite flux even in the small spinel size ranges, such as the 32–63 µm range primarily used here. From a total of 1652 kg of limestone from the earliest Berriasian to early Hauterivian Maiolica Formation, we recovered 108 extraterrestrial spinel grains (Fig. 1). Using three oxygen isotope and elemental analyses of the grains, we obtained the very first insights into what types of meteorites fell on Earth at times other than today and in the mid-Ordovician. These data can be used to test and develop models on the dynamics of meteorite transport from the asteroid belt to Earth and how the asteroid belt has evolved over time. For example, here we add perspectives on Bottke et al.'s (2007) hypothesis, based on astronomical data, that an ~170-km-diameter asteroid broke up between 190 and 140 Ma, leading to the formation of the Baptistina asteroid family, one of the youngest major asteroid families in the inner main asteroid belt.

MATERIALS AND METHODS

Samples were collected from 12 beds in two stratigraphically separated groups along the 240-m-thick Monte Acuto section of the Maiolica limestone in central Italy (43°27.83′N, 12°40.27′E). We collected a total of 513 kg from four beds in the Berriasian part of the section, and 1015 kg from eight beds in the late Valanginian to early Hauterivian part (Fig. 1). The size of the individual samples varied between 103 and 433 kg (plus one sample of 27 kg). One additional sample from the Bosso section, 12 km northwest of Monte Acuto, weighed 124 kg. The rocks were dissolved in HCl (6 *M*) and HF (11 *M*) at room temperature in the Astrogeobiology Laboratory specially built for separation of extraterrestrial minerals from sediments. After sieving at mesh sizes 32 and 63 µm, opaque Cr-spinel grains were identified by handpicking under the binocular microscope and

*E-mail: birger.schmitz@nuclear.lu.se

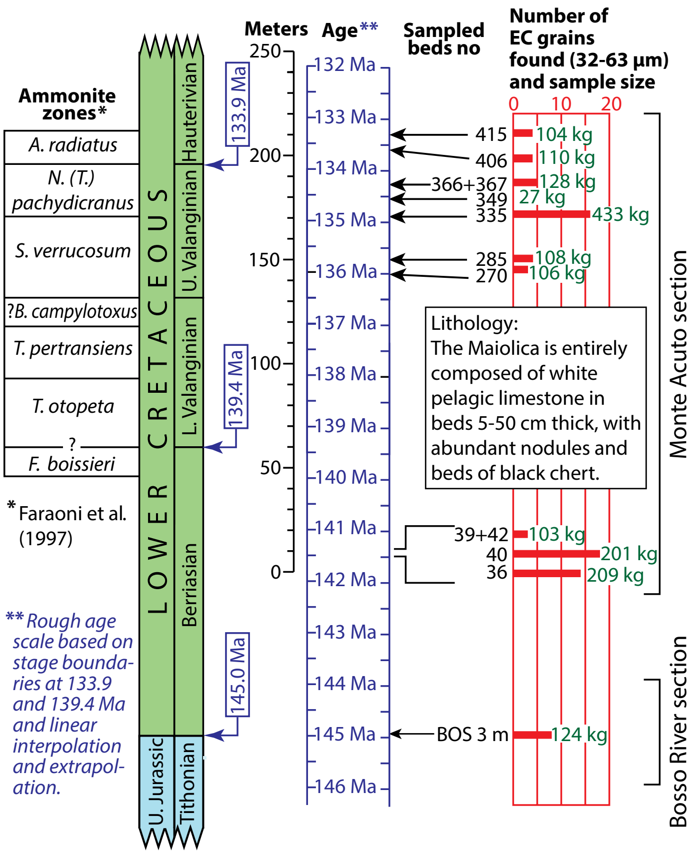

Figure 1. Stratigraphic context of Maiolica Formation, Italy, with position and size of samples analyzed and number of extraterrestrial chromite grains recovered. EC—equilibrated ordinary chondritic grains.

subsequent qualitative scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS) element analysis. From all samples, all transparent grains were also collected and analyzed in order to determine if any Mg-Al-spinels were present.

Polished epoxy mounts were prepared for all Cr-spinels together with an analytical standard UWCr-3 (Heck et al., 2017). Element concentrations were analyzed quantitatively with a calibrated Oxford-Link EDS mounted on a Hitachi SEM. Isotopes of $^{16}O^-$, $^{17}O^-$, and $^{18}O^-$ were analyzed with a Cameca IMS 1280 secondary ion mass spectrometer (SIMS) in three separate sessions with the procedures similar to Heck et al. (2017). We determined parts per thousand deviations from Vienna standard mean ocean water (VSMOW) as $\delta^{18}O$ and $\delta^{17}O$, and as $\Delta^{17}O$ from the terrestrial mass-fraction line ($= \delta^{17}O - 0.52 \times \delta^{18}O$), the latter being the main indicator for an extraterrestrial origin.

For further details on materials and methods, including a summary on the sedimentology of the Maiolica Formation, see the GSA Data Repository[1].

Division of Grains and Definitions

Equilibrated, ordinary chondritic chromite (EC) has a very distinct and narrow elemental composition and can readily be identified based on this criterion alone (Schmitz, 2013). The EC grains can then be further divided into the three groups H, L, and LL based on their oxygen isotope and TiO_2 content (see following). The remaining Cr-spinel grains were divided into two groups, "other chrome spinel with ≥0.45 wt% V_2O_3" (OC-V) and "all other chrome spinel" (OC). The basis for this is that most Cr-spinels from meteorites are rich in V, whereas V-rich (>0.5 wt%) terrestrial Cr-spinels are generally rare. The OC-V and OC grains were further subdivided based mainly on their oxygen isotopic composition.

The three groups of ordinary chondrites, H, L, and LL, have different average values of $\Delta^{17}O$ (0.73‰, 1.07‰, and 1.26‰, respectively; Clayton et al., 1991) and TiO_2 (2.2, 2.7, and 3.4 wt%, respectively; Schmitz, 2013). Around these averages, the $\Delta^{17}O$ and TiO_2 values are spread following a Gaussian distribution, but the distributional tails overlap (Heck et al., 2016). The exact ranges for dividing grains based on TiO_2 and/or $\Delta^{17}O$ can in principle be arbitrarily set, but they must be used consistently when comparing different time periods.

RESULTS

Among the total of 108 extraterrestrial Cr-spinel grains (32–250 µm large) recovered, 81 are clearly equilibrated ordinary chondritic (EC), and 27 are vanadium-rich grains (OC-V), probably originating from other types of meteorites (Table 1; Tables DR1 and DR2). In the >63 µm fraction, only two extraterrestrial grains were found; both were EC grains. In the 32–63 µm fraction, based on element analyses alone, we found 79 grains with clear EC elemental composition. Oxygen-isotopic analysis was performed on 46 of these grains, confirming that they are all ordinary chondritic grains (Fig. 2).

The division of the 81 recovered EC grains into their three subgroups, based entirely on their TiO_2 content, is given in Table 2. The reason for using TiO_2 is the substantially larger data set for grains having been analyzed for TiO_2 compared to $\Delta^{17}O$. These data were compared with estimates based on the same approach for EC grains from the mid-Ordovician before and after the LCPB, as well as the proportions among recent meteorite falls following the Meteoritical Bulletin Database. From our comparisons of divisions based on either TiO_2 or $\Delta^{17}O$, as well as both parameters combined, we know that the main trends based on TiO_2 alone, can be seen in all three approaches (Table DR3).

Of the 27 OC-V grains in the 32–63 µm fraction, 17 grains could be analyzed for three oxygen isotopes. Of these, 13 gave $\Delta^{17}O$ values separated (at 2 standard deviations [SD]) from the terrestrial fractionation line (TFL), i.e., indicating a meteoritic origin (Fig. 2). Four of the grains show $\Delta^{17}O$ indistinguishable from the TFL, but we argue that these grains nevertheless originate from achondritic meteorite types that have $\Delta^{17}O$ values at or very close to the TFL. There is nothing in our data or the general paleogeographic setting that indicates that the types of rare terrestrial rocks that potentially could yield V-rich Cr-spinels existed in the study region in the Early Cretaceous. Two of the 17 OC-V grains analyzed lie clearly below (at 2 SD) the TFL, with $\Delta^{17}O$ values of –0.4‰. Altogether, the six OC-V grains with values at or just below the TFL may originate from the howardite, eucrite, diogenite (HED) types of meteorites, or from primitive or ungrouped achondrites. Five of the 17 OC-V grains analyzed

TABLE 1. COMPARISON OF FLUX OF EQUILIBRATED ORDINARY CHONDRITES TO OTHER CHROME-SPINEL–BEARING METEORITE TYPES

Type of chrome spinel*	Before LCPB Ordovician no. (%) of grains	Early Cretaceous no. (%) of grains	Today % of flux†
EC	23 (56)	81 (75)	90
OC-V	15 (37)	27 (25)	9
AC-low-V	3 (7)	?	<1
	Ratios§		
EC/OC-V	1.5	3	10

*EC—chromite from equilibrated ordinary chondrite; OC-V—other chrome spinel with $V_2O_3 \geq 0.45$ wt%; AC-low-V—chrome spinel from achondrites judging from $\Delta^{17}O$ value below terrestrial fractionation line, but with $V_2O_3 \leq 0.44$ wt%.

†Fraction of the flux excluding the recent major meteorite groups poor in large Cr-spinel (see Heck et al., 2017).

§The ratios for the ancient fluxes are not directly comparable to the ratio for today's flux. Most achondrites have lower contents of chrome spinel (>32 µm) than the equilibrated ordinary chondrites (Heck et al., 2017). If this could be accounted for, the Ordovician and Cretaceous ratios would be even lower than given here.

[1]GSA Data Repository item 2017273, Item DR1 and Tables DR1–DR6 (additional data and descriptions of methods), is available online at http://www.geosociety.org/datarepository/2017/, or on request from editing@geosociety.org.

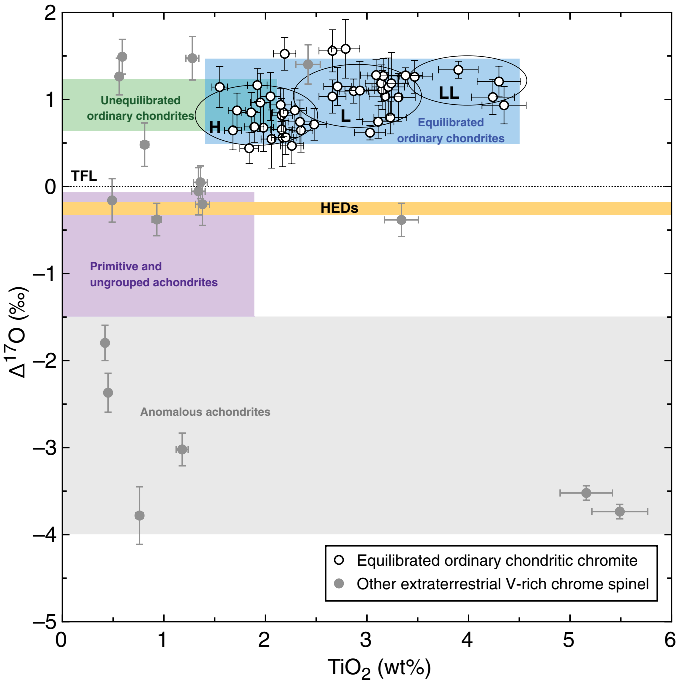

Figure 2. Oxygen isotope and TiO_2 composition for 63 out of 108 total extraterrestrial Cr-spinel grains recovered from Maiolica Formation, Italy. TFL—terrestrial fractionation line; HED—howardite, eucrite, and diogenite meteorites.

TABLE 2. DIVISION OF EQUILIBRATED ORDINARY CHONDRITIC (EC) GRAINS USING TiO_2 (wt%)

	H	**L**	**LL**
TiO_2 (wt%)	**≤2.50%**	**2.51%–3.39%**	**≥3.40%**
Recent falls* (%)	42	47	11
Early Cretaceous† (*n* = 81)	36 grains	34 grains	11 grains
Percent	44%	42%	14%
Mid-Ordovician, post-LCPB§ (*n* = 119)	10 grains	102 grains	7 grains
Percent	8%	86%	6%
Mid-Ordovician, pre-LCPB# (*n* = 215)	80 grains	71 grains	64 grains
Percent	37%	33%	30%

Note: LCPB—L-chondrite parent body breakup event.
*From Meteoritical Bulletin Database.
†This study.
§Heck et al. (2016).
#This study and Heck et al. (2017).

have $\Delta^{17}O$ values similar to ordinary chondrites, despite having an elemental composition significantly outside the defined EC range. The grains have $\Delta^{17}O$ values spanning the range from H- to LL-chondritic meteorites (0.48‰ to 1.49‰) and may originate from unequilibrated ordinary chondrites, i.e., petrographic types 3–4. The last six of the OC-V grains analyzed for $\Delta^{17}O$ have anomalously low $\Delta^{17}O$ values, in the range −1.80‰ to −3.78‰, indicating that they originate from primitive (e.g., winonaites, lodranites, acapulcoites) or ungrouped or anomalous achondrites that are very rare on Earth today (see following). In summary, 12 of the 17 OC-V grains analyzed for oxygen isotopes are from achondrites, and five are from (unequilibrated?) ordinary chondrites.

We also recovered a total of 33 and 65 grains in the >63 and 32–63 µm fractions, respectively, classified as “other Cr-spinel” (OC) grains, meaning that they have a different composition than the EC grains, but with V_2O_3 concentrations <0.45 wt%. These grains are almost certainly terrestrial, except for possibly two of them. We analyzed 23 of them for oxygen isotopes, and 21 have $\Delta^{17}O$ values right on the TFL within ~2 SD (Table DR2). Two of the grains lie slightly off the TFL (−0.31‰ ± 0.18‰, and 0.34‰ ± 0.21‰), which could indicate an extraterrestrial origin, but the element compositions of the grains are similar to that of typical terrestrial OC grains in the section.

The Berriasian part of the Monte Acuto section has higher concentrations of EC grains, 7.0 per 100 kg rock, compared to 3.5 per 100 kg in the younger Valanginian–Hauterivian part of the section. The ratio OC-V versus EC grains is about the same, ~0.3, throughout the entire section, which supports the view that all of the OC-V grains are extraterrestrial. In the Monte Acuto section, all except three of the 88 OC grains were found in the younger part of the section.

Because acid residues of the Maiolica limestone are so clean, it has been possible to also quantify the amount of transparent Mg-Al-spinels. In some of the more common carbonaceous chondritic meteorite types that fall on Earth today (e.g., CM and CV types), Mg-Al-spinels are common in the 32–63 µm fraction and dominate over opaque Cr-spinels (Schmitz, 2013). Only one Mg-Al-spinel grain was found in our samples, which is significant for the interpretation of the origin of the other Cr-spinel grains (see following). The single grain was found in Bed 406 together with many terrestrial zircons. This fact and the low V_2O_3 content (<0.3 wt%) of the grain indicate that it is most likely of terrestrial origin.

DISCUSSION

Flux of Meteorites through the Phanerozoic

Before the present study, the only periods in deep time for which information existed about the types of meteorites that commonly fell on Earth were for the mid-Ordovician before and after the LCPB (Schmitz et al., 2003; Schmitz, 2013; Heck et al., 2016, 2017). The addition here of a third “window” adds an important new perspective, but interpretations and generalizations must be preliminary awaiting the results for additional time windows. Perhaps the most important result so far is that it indeed is possible to obtain quantitative insights into the history of the asteroid belt from Earth’s sedimentary record.

The ratio of ordinary chondritic/achondritic meteorites from Early Cretaceous strata (~3) is somewhere between the ratios for the mid-Ordovician before the LCPB (~1.5) and recent time (~10; Table 1). It appears that the background meteorite flux may have evolved gradually over the Phanerozoic, from being dominated by achondrites to the present situation, where ordinary chondrites dominate (>80% of all meteorite falls) and Cr-spinel-rich achondrites (other than the ~8% HED meteorites) are very rare, representing less than 1%. Even if the ordinary chondrites may have had a more subordinate role at times, our data show that they likely have always represented an important fraction of the flux.

There may also be a trend in the flux of ordinary chondrites through the Phanerozoic (Table 2). Within the resolution of our approach, the ratios between the different groups of ordinary chondrites in the Early Cretaceous (H:L:LL ~1:1:0.2) are identical to the ratios today (~1:1:0.2). In the mid-Ordovician after the LCPB, the EC grains are 100% (or close to) L-chondritic. The 8% and 6% H- and LL-chondritic grains given in Table 2 most likely reflect L grains in the tails that overlap in TiO_2 content with the “neighbor groups.” Heck et al. (2016) analyzed 120 post-LCPB EC grains for oxygen isotopes and found that ≥99% of the grains were L-chondritic. Before the LCPB, the LL chondrites (based on 215 EC grains analyzed for TiO_2) represent ~30% of the ordinary chondritic flux, compared to ~10% in the Early Cretaceous and today (Table 2; Table DR4; Heck et al., 2017). The L and H chondrites share the remaining 70% of the pre-LCPB flux in about equal proportions. The high relative abundance of LL chondrites in the pre-LCPB meteorite assemblage probably reflects the tail of the meteorite flux related to the breakup of the LL-chondritic Flora family ca. 1 Ga (see, Heck et al., 2017). With its ~14,000 members, this is one of the largest asteroid families (Nesvorný et al., 2015).

The many OC-V grains that cannot be assigned to unequilibrated ordinary chondrites likely originate from achondritic meteorites. The carbonaceous chondrites (with rare exceptions) contain only low concentrations of opaque spinels in the grain-size ranges we analyzed as compared to ordinary chondrites and most of the achondrites (Heck et al., 2017). Because of the clear dominance of transparent Mg-Al-spinels over opaque spinels in carbonaceous chondrites, the scarcity of Mg-Al-spinels in our samples gives support for that none of the recovered opaque grains is from carbonaceous chondrites. Iron meteorites, mesosiderites, and pallasites contain spinel concentrations too low to be of any significance in this type of study. Rumuruti chondrites are rich in Cr-spinels but have $\Delta^{17}O$ of ~2‰, values not observed here. Enstatite chondrites are too reduced to contain significant amounts of any Cr-rich oxides.

In our assemblage of Cr-spinel grains, there is a significant amount (~10%) of achondritic grains with $\Delta^{17}O$ values at or below −2‰ (Fig. 2; Table DR5). Such achondrites are very rare on Earth today, representing less than a tenth of a percent of the known recent meteorites. In Bed 36, there are three such grains: two grains with $\Delta^{17}O$ close to −2‰ and one grain with a value of −3.8‰. In Bed 40, there are two grains with values of −3.5‰ and almost identical elemental composition, indicating that they may have come from the same micrometeorite. In Bed 406, one grain has a $\Delta^{17}O$ value of −3.0‰. These six grains may originate from "extinct" anomalous achondrites similar to the mid-Ordovician fossil meteorite Österplana 065 (Schmitz et al., 2016). Based on Cr and O isotopic analyses, this meteorite is believed to originate from a type of meteorite that no longer falls on Earth because its parent body in the asteroid belt has been consumed by collisions. Altogether, the six OC-V grains with $\Delta^{17}O$ values just below or at the TFL may originate from primitive achondrites or HED achondrites, the latter having $\Delta^{17}O$ of ~−0.2‰ and representing ~8% of the recent flux. The HED achondrites originate from the 4 Vesta asteroid, the second largest asteroid in the asteroid belt

Baptistina Family Breakup

We found no apparent signature of the Baptistina (LL-chondritic) asteroid family-forming event. By back-tracking the orbits of asteroids among the members of the Baptistina asteroid family in the inner main belt, Bottke et al. (2007) estimated that the ~170 km parent body of the Baptistina asteroid family broke up at 160 Ma (with an uncertainty range 190–140 Ma). Masiero et al. (2012) suggested a revised event age of 190 ± 30 Ma based on astronomical data. This is one of the youngest major asteroid family-forming events, resulting in a family of ~2500 members (Nesvorný et al., 2015). Bottke et al. (2007) postulated that the impactors creating the K-Pg boundary Chicxulub crater on the Yucatan Peninsula and the Tycho crater on the Moon around 109 Ma originate from this collisional event. However, refined spectral studies have shown that the Baptistina family members are LL chondrites (Reddy et al., 2014), whereas Cr-isotopic analyses of K-Pg boundary ejecta rule out such an impactor, favoring instead a cometary or carbonaceous (CM type) impactor (Trinquier et al., 2006). Our samples span the range from 145 to 133 Ma, and nowhere in the section do we see any enrichment in LL-chondritic grains, neither do we see any tailing-off trend in LL-grain abundances between the oldest and youngest samples. Our results together with the data of Masiero et al. (2012) constrain the breakup event to have occurred between 220 and 145 Ma.

CONCLUSIONS

A history of the asteroid belt can be reconstructed from Earth's sedimentary record by recovering extraterrestrial spinels in condensed sediments. The three first "windows" into the meteorite flux in deep time have been reconstructed, providing insights into the flux in the Early Cretaceous as well as before and after the breakup of the L-chondrite parent body in the mid-Ordovician. The background meteorite flux in the early Paleozoic appears to have been significantly different from the flux in the Cretaceous, which is more similar to today's flux. Achondrites dominated in the early Paleozoic, and ordinary chondrites dominate today. This general trend was at times overprinted (for 1–10 m.y.) by floods of single types of meteorites from occasional major breakup events in the asteroid belt, such as after the LCPB.

ACKNOWLEDGMENTS

This work was supported by a European Research Council Advanced Grant to Schmitz. We thank A. Montanari, S. Boschi, K. Terfelt, M. Alvarez, and E. Schmitz, L. Schmitz, and N. Schmitz for field support, and F. Iqbal (Lund University, Sweden), J. Kern (University of Wisconsin, USA), and B. Strack (Field Museum of Natural History, Chicago, USA) for laboratory assistance. Heck was funded by the Tawani Foundation. WiscSIMS is partly supported by U.S. National Science Foundation (NSF) grants EAR-03-19230 and EAR-13-55590. Three-dimensional microscopy performed at the Keck-II facility NUANCE Center was supported by the Nanoscale Science and Engineering Center (NSF EEC-0647560), Materials Research Science and Engineering Centers (NSF DMR-1121262), the Keck Foundation, the State of Illinois, and Northwestern University. We thank P. Claeys, A. Wittmann, and an anonymous reviewer for helpful comments.

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Manuscript received 3 March 2017
Revised manuscript received 7 May 2017
Manuscript accepted 8 May 2017

Printed in USA