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Chromium isotopic studies of terrestrial impact craters: Identification of meteoritic components at Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart

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Abstract

Chromium isotopic compositions and elemental abundances in impact melt rock and impact glass samples from four terrestrial impact craters were measured to verify the presence of an extraterrestrial component and to identify the meteorite type of the impactor. All meteorite classes have Cr isotopic signatures that are different from those of terrestrial rocks; thus, precise measurements of Cr isotopic abundances can unequivocally distinguish terrestrial from extraterrestrial materials. For all four studied craters — Bosumtwi (Ghana), Clearwater East (Canada), Lappajärvi (Finland), and Rochechouart (France) we found positive ^{53}Cr excesses that eliminate carbonaceous chondrite projectiles (because those would show apparent negative excesses) and enstatite chondrites (because of the magnitude of the excess). In all four cases, ordinary chondrites have been identified as the best fit for the data; in the case of Lappajärvi interelement correlations together with the Cr isotope data make an H-chondrite the most likely projectile, whereas in the case of Clearwater East both L or H chondrites are possible. For Bosumtwi and Rochechouart the high indigenous contents of the siderophile elements, and disturbances of the elemental abundances by weathering and hydrothermal alteration, respectively, do not allow further constraints to be placed on the type of ordinary chondrite involved in the impact.

Keywords: impact cratering; chromium isotopes; Bosumtwi crater, Ghana; Lappajärvi crater, Finland; Clearwater East crater, Canada; Rochechouart crater, France; meteoritic components

1. Introduction: identification of meteoritic components at terrestrial impact craters

On Earth today about 170 craters and structures are known that have formed by the impact of extraterrestrial bodies — mostly asteroids. Most, if not all, of the impacting body is destroyed (melted and/or vaporized) during the crater-forming event, and, because the volume of the projectile is several orders of magnitude smaller than the volume of terrestrial material that is vaporized, melted, or brecciated, the minor amount of extraterrestrial melt or vapor is severely diluted by terrestrial material. In a few cases, meteoritic fragments can spall off the impacting body, preserving some meteoritic material (for example, at Meteor Crater, Arizona — [1]), but during erosion of the impact

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structure, such material is usually rapidly destroyed. Even though "normal" meteorites provide abundant material from meteorite parent bodies, i.e., asteroids, for study, it is nevertheless of importance to understand the sources of large, crater-forming impactors. In addition, the detection and verification of an extraterrestrial component in impact-derived melt rocks or breccias can be of diagnostic value to provide confirming evidence for an impact origin of a geological structure (see the review by [2]).

A limiting factor is that, as mentioned above, only a very small amount of the finely dispersed meteoritic melt or vapor is mixed with a much larger quantity of target rock vapor and melt, and this mixture later forms impact melt rocks, melt breccias, or impact glass. In most cases, the contribution of extraterrestrial matter to these impactite lithologies is very small — mostly much less than 1% by weight, leading to only slight chemical changes in the resulting impactites. The detection of such small amounts of meteoritic matter within the normal upper crustal compositional signature of the target rocks is extremely difficult. Only elements that have high abundances in meteorites, but low abundances in terrestrial crustal rocks (in particular, the siderophile elements) can be used reliably. Another complication is the existence of a variety of meteorite groups and types (the three main groups are stony meteorites, iron meteorites, and stony-iron meteorites), which have a wide range of siderophile element compositions. Distinctly higher siderophile element contents in impact melts, compared to target rock abundances, can be indicative of the presence of either a chondritic or an iron meteoritic component. The signatures of achondritic projectiles (stony meteorites that underwent magmatic differentiation) are much more difficult to detect, because they have significantly lower abundances of the key siderophile elements — at levels that are similar to terrestrial mantle rocks. Furthermore, in order to reliably constrain the target rock contribution to the composition of melt rocks or breccias, the so-called indigenous component, absolute certainty must be attained that all contributing terrestrial target rocks have been identified and their relative contributions to the melt mixture are reasonably well known. In cases where this is difficult, the regression approach discussed by McDonald et al. [3] offers an alternative way to estimate the average input from the target rocks. This approach depends on a good quality regression line, but has the advantage that not all target rocks need to be identified and analyzed.

Meteoritic components can be identified not only in crater-fill rocks, but can also be of diagnostic value in the identification and confirmation of distal ejecta layers. Maybe the best example for such a study is the identification of an extraterrestrial component at the Cretaceous–Tertiary (K/T) boundary [57], which was of crucial importance to a reassessment of the importance of impact events in the geological (and biological) history of the Earth.

Geochemical methods have been used to determine the presence of the traces of such an extraterrestrial component (including classical papers by, e.g., [4–10, 58]). In the absence of actual meteorite fragments, it is necessary to chemically search for traces of meteoritic material that is mixed in with the target rocks in breccias and melt rocks. Meteoritic components have been identified for just about 45 impact structures (for a current list, see [11]), out of the more than 170 impact structures that have so far been identified on Earth. This list includes mostly well-studied structures, and many of the identifications are not very precise, or preliminary, or the type of impactor has been subject to debate in cases where several independent investigations had taken place. Only a few impact structures were first identified by finding a meteoritic component, with the overwhelming majority having been confirmed by the identification of shock metamorphic effects.

The presence of a meteoritic component can be verified by measuring the abundances and interelement ratios of siderophile elements, especially the platinum group elements (PGEs), which are several orders of magnitude more abundant in meteorites than in terrestrial upper crustal rocks. Often the content of the element iridium is measured as a proxy for all PGEs, because it can be measured with the best detection limit of all PGEs by neutron activation analysis, but taken out of context, small Ir anomalies alone have little diagnostic power. More reliable results can be achieved by measuring the contents and ratios of a whole suite of elements, for example, the PGEs, which also avoids some of the ambiguities that result if only moderately siderophile elements (e.g., Cr, Co, Ni) are used. In the past, PGE data were used to estimate the type or class of meteorite for the impactor (e.g., [12,4,8,5,6,13]), but these attempts were not always successful. It is difficult to distinguish among different chondrite types based on siderophile element (or even PGE) abundances, which has led to conflicting conclusions regarding the nature of the impactor at a number of structures (see [2], for details). In addition, further complications arise because meteorites have a range of compositions within each class, if the target rocks have high abundances of siderophile elements, or if the siderophile element concentrations in the impactites are very low. Also, PGE data have been measured for only a few samples within each class, limiting the database for comparison (see, e.g., [14], for a discussion).

In such cases, the Os and Cr isotopic systems can be used to establish the presence of a meteoritic component in a number of impact melt rocks and breccias (e.g., [10,15,16]). Both of these methods are based on the observation that the isotopic compositions of the elements Os and Cr, respectively, are different in most meteorites compared to terrestrial rocks (see review in [11]); the Cr isotopic method allows, in addition, the identification of the type of meteoritic material involved. Here we describe the successful determination of the type of impactors for four terrestrial impact craters: Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart.

2. Geological background for studied impact craters

2.1. Bosumtwi (Ghana)

The Bosumtwi impact crater is located in the Ashanti Province of southern Ghana in West Africa. It is situated near the regional capital town of Kumasi, which is also the second largest city in Ghana, as well as the capital of the Ashanti Kingdom. The Bosumtwi structure is centered at 06° 32′ N and 01° 25′ W. It is one of only 19 confirmed impact structures known in Africa, and is the youngest well-preserved complex impact crater known on Earth. The structure, which has an age of 1.07 million years, is almost completely filled by Lake Bosumtwi of roughly 8.5 km diameter, and has a rim-to-rim diameter of about 10.5 km. The well-preserved crater rim is elevated by 210–350 m above the lake level; the lake level is about 80 to 100 m below the terrane outside of the rim. Seismic data indicate the presence of a small (ca. 2 km wide) central uplift preserved underneath the lake sediments. Because of its importance for cratering studies, and because of the potential of the lake sediments to preserve an uninterrupted 1-Ma paleoclimate record, Bosumtwi was the target, in 2004, of a large international and multidisciplinary drilling program. A recent review of Bosumtwi is provided by Koeberl and Reimold [17], from which this summary is assembled, and where further references are listed.

The region around Lake Bosumtwi is covered by very dense, tropical rainforest; thus outcrop is generally scarce and mostly limited to exposure or suboutcrop along roads, some outcrop – often badly weathered – in stream beds, and occasional small exposures on steep slopes. The Bosumtwi impact crater was excavated in lower greenschist facies metasediments of the 2.1–2.2 Ga Birimian Supergroup. These supracrustals comprise interbedded phyllites/mica and quartz-feldspar schists and meta-tuffs, together with meta-graywackes, quartzitic graywackes, shales, and slates. Birimian metavolcanic rocks (altered basic intrusives with some intercalated metasediments) occur to the southeast of the crater. Clastic sedimentary rocks of the Tarkwaian Group, which are regarded as the detritus of Birimian rocks, occur to the east and southeast of the crater. The late Proterozoic supracrustal strata are locally intruded by a series of granitic–dioritic bodies. A range of mafic intrusions, mostly in the form of local dike and sill developments, is known from the entire region. Rock formations associated with the formation of the crater include various breccias, and, especially, massive suevite (a polymict impact breccia of fine-grained clastic groundmass with mineral, lithic, and glass/melt clasts) deposits, which occur just outside the northern and southwestern crater rim, as well as within the crater fill.

Most importantly, Bosumtwi has been identified as the source crater of the Ivory Coast tektites — one of only four known tektite strewn fields on Earth. Ivory Coast tektites were first reported in 1934 from a geographically rather restricted area in the Ivory Coast (Cote d'Ivoire), West Africa. Microtektites were reported from deep-sea sediments of corresponding age from the eastern equatorial Atlantic Ocean west of Africa. Ivory Coast tektites and the Bosumtwi crater have the same age (e.g., [18,19]), and there are close similarities between the isotopic and chemical compositions of the tektites and crater rocks (e.g., [19,20]), confirming a connection between the crater and the tektites.

2.2. Clearwater East (Canada)

The Clearwater impact structures consist of two lake-filled features, the 22 km diameter Clearwater East impact structure (centered at 56° 05′ N and 74° 07′ W), and the nearby 32 km diameter Clearwater West structure (centered at 56° 13′ N and 74° 30′ W). The impact occurred into Precambrian rocks, composed mainly of granodiorites, quartz monzonites, and granite gneisses of Archean age. In the western lake, a ring of islands comprises the eroded remnant of the central uplift, whereas in Clearwater East the central uplift is submerged. The islands in the western structure expose impact melt rocks, whereas similar material was found in the eastern structure only by drilling. In the 1960s, two drill cores were obtained from Clearwater East, with one of them reaching a depth of over 1.1 km in gneissic bedrock, whereas the other was terminated at 340 m still within the impact melt layer. The melt rocks at both structures have similar compositions, with those in West Clearwater having a slightly more mafic composition, possibly indicating a slight difference in the target composition [6]. Dating of melt rock from Clearwater East, in the form of a Rb–Sr isochron, gave an age of 287±26 Ma [21], whereas a good Ar–Ar plateau could only be obtained for a melt rock sample from Clearwater West, and gave an age of 280±2 Ma [22]. The two structures are considered to have formed simultaneously (e.g., [5,21]).

2.3. Lappajärvi (Finland)

The Lappajärvi impact structure in western Finland is centered at 63° 12′ N and 23° 42′ E. It is eroded, is partly filled by Lake Lappajärvi, and has a current diameter of 23 km. The age, based on an Ar–Ar study of impact melt, is 77.3±0.4 Ma [23]; this was recently confirmed from U–Th–Pb dating of zircons in the melt rock [24]. The crater formed in Paleoproterozoic Svecofennian crystalline and metasedimentary rocks (mainly mica-schist, granite, pegmatite, amphibolite, and granodiorite) of 1900–1950 Ma age; peneplanation and weathering of these rocks was followed by the deposition of thin layers of Mesoproterozoic (ca. 1200 Ma) siltstones and sandstones, and, possibly, some later deposition of Cambrian age muddy to silty sandstones and limestones (e.g., [25]).

Outcrops on islands within Lake Lappajärvi show the presence of a coherent sheet of melt rocks (called kärnäite), and some suevitic breccia has been found as well. Drilling in the crater revealed that in its central part about 145 m of kärnäite are underlain by a 5-m thick layer of suevite, and fragmental breccia to a depth of at least 217 m (the depth of the drilling) [25]. In a detailed petrographic and geochemical study of the Lappajärvi melt rock, Reimold [26] calculated that the kärnäite, which is fairly homogeneous in terms of composition, can be reproduced by a mixture of 76 vol.% mica schist, 11 vol.% granite pegmatite, and 13 vol.% amphibolite.

2.4. Rochechouart (France)

The Rochechouart impact structure is centered at 45° 50′ N and 00° 56′ E in the northwestern French Massif Central, about 40 km west of the city of Limoges. The crater is deeply eroded and no impact-related topography remains; the present-day surface is at about the level of the crater floor. The reconstructed crater diameter, based on morphological, geophysical, and structural considerations, is about 20–25 km, whereas presently shock metamorphic effects in a variety of breccias and target rocks are confined to a zone about 13 km in diameter (e.g., [27,28]). All fallout suevite and other breccia were removed by erosion, and outcrop is poor. The target rocks are predominantly Hercynian granites and gneisses, with some amphibolites, diorites, and porphyries, often intruded by granitoids. In situ fracturing and brecciation has been noted up to 10 km from the crater center [27]. Various impact breccia dikes and pseudotachylite occur in the subcrater basement. The exposed allochthonous breccias form a horizontal unit up to 60 m thick (average 12 m) in the central part of the structure [28]. Three impactites are recognized — impact melt rock, suevite, and polymict lithic breccia. A complete range of shock effects, ranging from macroscopic effects such as shatter cones, to microscopic effects (e.g., planar deformation features in quartz within breccias) permitted a study of shock zoning [27]. The determination of the age of the structure was somewhat difficult, with K–Ar ages (around 160–170 Ma) probably influenced by alteration, so that a Rb–Sr isochron age on melt rocks of 186±8 Ma [29] was long taken as the correct age. More recently, Kelley and Spray [30] used laser spot fusion Ar–Ar dating on a pseudotachylites from the Champagnac quarry, ca. 6 km NE of the center of the structure, which yielded a Late Triassic age of 214±8 Ma (within error the same age as that of the ca. 100 km diameter Manicouagan impact structure, Canada).

3. Method

3.1. Background

As mentioned above, the verification of the presence of an extraterrestrial component in impactites requires meticulous geochemical analyses, and elemental content information may, in cases, yield ambiguous information. The Os and Cr isotopic methods potentially remove much of this ambiguity (cf. [31]), and the Cr isotopic method in particular can help to identify the type of meteorite involved in an impact event. The method is based on the determination of the relative abundances of ^{53}Cr, which is the daughter product of the extinct radionuclide ^{53}Mn (half life = 3.7 Ma). The ^{53}Cr relative abundances are measured as the deviations of the ^{53}Cr/^{52}Cr ratio in a sample relative to the standard terrestrial ^{53}Cr/^{52}Cr ratio by high-precision thermal ionization mass spectrometry [32]. These deviations are usually expressed in ε units (1 ε is 1 part in 10^4, or

0.01%). Terrestrial rocks are not expected to show (and, indeed, do not reveal) any variation in the $^{53}Cr/^{52}Cr$ ratio, because the homogenization of the Earth was completed long after all ^{53}Mn (which was derived from the solar nebula) had decayed. In contrast, data for most meteorite groups, such as carbonaceous, ordinary, and enstatite chondrites, primitive achondrites, and other differentiated meteorites (including the SNC meteorites) show a variable excess of ^{53}Cr relative to terrestrial samples [16,33–35]). For the various meteorite types, the range is about +0.1 to +1.3 ε. Only the carbonaceous chondrites show an apparent deficit in ^{53}Cr of about −0.4 ε. Some of these differences may reflect early Mn/Cr fractionation in the solar nebula [36] and in meteorite parent bodies [32]. The negative ε(53) values for carbonaceous chondrites are an artifact of using the $^{54}Cr/^{52}Cr$ ratio for a second order fractionation correction to increase precision [32,37]. Because these meteorites carry a pre-solar ^{54}Cr component [38] the $^{54}Cr/^{52}Cr$ ratios are elevated in these meteorites. An assumed normal $^{54}Cr/^{52}Cr$ ratio is used for the second-order correction. Note that the true, unnormalized $\varepsilon(^{53}Cr)$ values are positive and similar to those of other undifferentiated meteorites, and the apparent ^{53}Cr deficit in the carbonaceous chondrites is actually due to the excess of ^{54}Cr [38,37,36]. However, the presence of ^{54}Cr excesses in bulk carbonaceous chondrites allows distinguishing these meteorites clearly from the other meteorite classes.

The Cr isotopic method was used by Shukolyukov and Lugmair [16] on samples from the K/T boundary in Denmark and Spain, who found that about 80% of the Cr in these samples originated from an impactor with a carbonaceous chondritic composition, rather than an ordinary chondritic composition. More recently, Trinquier et al. [37] refined this identification to a CM2 carbonaceous chondrite. The only other case for a carbonaceous chondritic signal identified so far from Cr isotope data in impact ejecta is for some samples from Archean spherule layers in the Barberton Mountain Land, South Africa [38,39].

Despite the selectivity of the Cr isotopic method, it has two disadvantages. First, the analytical method is complicated and time-consuming, and second, a significant proportion of the Cr in an impactite, compared to the abundance in the target, has to be of extraterrestrial origin. This point was discussed in Koeberl et al. [31], who noted that the detection limit is a function of the Cr content in the (terrestrial) target rocks involved in the formation of the impact breccias or melt rocks. For example, if the average Cr concentration in the target is ~185 ppm (the average Cr concentration in the bulk continental crust, [40]), only an extraterrestrial component of more than 1.2 wt.% can be detected. In some cases combining the Cr isotope data together with inter-element correlations between siderophile elements allowed to constrain the specific type of an ordinary chondrite. For example, such an approach allowed to conclude that melt rocks from the ca. 70–80-km diameter, 145 Ma Morokweng impact structure (South Africa) showed contamination from an L-type ordinary chondrite [31], in agreement with independent approaches by Koeberl et al. [41], McDonald et al. [3], Koeberl and Reimold [42], and Maier et al. [43]. The latter authors reported on a meteorite fragment of possibly LL-chondritic composition within impact melt rock.

3.2. Experimental technique and samples

To avoid nugget effects, fairly large samples (at least 1 g) have been used. The isotopic composition, as well as the concentrations of Cr (by ICP-OES) were measured. The isotopic analyses of Cr were made with a VG-54E thermal ionization single-collector mass spectrometer. Chromium was loaded in dilute HCl on single prebaked W filaments and was analyzed as Cr^+. A mixture of silica gel and boric acid was used as an emitter. The Cr amount per load was usually 600–700 ng. The running temperature was typically 1150–1190 °C and the ion-current for ^{52}Cr of 5.5×10^{-11} A was held constant to within ±10%. For each sample, isobaric interferences were monitored via background scans before and sometimes during the experiment with the use of a Daly detector. Interferences from ^{50}Ti, ^{50}V, and ^{54}Fe were monitored at ^{48}Ti, ^{51}V, and ^{56}Fe and were always negligible. A chromium shelf standard solution and several chemically processed geological samples were used as a terrestrial reference material and test samples. More details of the chemical separation procedure for Cr and the mass spectrometric procedure are described in Lugmair and Shukolyukov [32] and Shukolyukov and Lugmair [35]. The isotopic variations of ^{53}Cr are measured as the deviation of the $^{53}Cr/^{52}Cr$ ratio from the normal terrestrial $^{53}Cr/^{52}Cr$ ratio. All Cr isotopes were normalized to ^{52}Cr and corrected for mass fractionation.

Impact melt rock of glass samples from four impact structures were analyzed for the present study; some preliminary results were presented in abstract form by Shukolyukov and Lugmair [33] and Koeberl et al. [44]. From the Bosumtwi crater, this was an Ivory Coast tektite, sample IVC 3395 (see [19], for details), in which previously an extraterrestrial component was found by

Table 1
Compositional information for siderophile and chalcophile elements in the samples from the Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart impact structures analyzed for Cr isotopic composition

Crater	Bosumtwi	Clearwater East	Lappajärvi	Rochechouart
Sample No.	IVC-3395	#3	L2-2	VR 1
Reference	A	B	C	C
Cr (ppm)	223	304	93	301
Fe (wt.%)	4.82	3.87	3.87	4.80
Co (ppm)	26.7	55.8	22.6	25.5
Ni (ppm)	112	853	240	252
Cu (ppm)	n.d.	1.9	22.0	97**
As (ppm)	0.26	20*	0.50	10.2
Sb (ppm)	0.14	n.d.	0.02	0.35
W (ppm)	1.45	n.d.	0.55	17.2
Ir (ppb)	0.4	36.3	7.6	15.2
Au (ppb)	55	7.9	2.8	0.8**

References: A: Koeberl et al. [19], B: Palme et al. [6], C: Palme et al. [7]. n.d.=no data. *Sample #2, same set of samples [6]. **Sample VR 2, same batch of samples [7].

Os isotopic analyses [10]. For Clearwater East, we analyzed impact melt rock sample #3 from the study of Palme et al. [6], which was retrieved from drillhole DCW-2-63 at a true depth of 277.7 m. The Lappajärvi and Rochechouart melt rock samples are samples no. L2-2 and VR 1, respectively, both from Palme et al. [7]. The contents of selected siderophile and chalcophile elements in these samples are given in Table 1. All samples have elevated contents of siderophile elements (e.g., Ir) compared to the respective target rocks.

4. Meteoritic components in four impact craters: results and discussion

The results of the Cr concentration and isotopic composition analyses of the materials from the four impact structures Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart are given in Table 2, together with values for the various meteorite groups, and some other related materials. In the following sections we discuss previous attempts at identifying meteoritic components at these craters, as well as our new results and their implications.

4.1. Bosumtwi

After the link between the Ivory Coast tektites and the Bosumtwi crater was found, the tektites were studied to see if they contain an extraterrestrial component. Palme et al. [5] noted that Ivory Coast tektites have high contents of siderophile elements, in particular Ir and Os (both on the order of 0.3–0.4 ppb), which, because of the non-chondritic interelement ratios, they interpreted as evidence of an iron meteorite contamination. However, Jones [45] noted that the high siderophile element contents in the tektites could be the result of high indigenous contents in the Bosumtwi target rocks due to local mineralization. The question regarding the presence of a meteoritic component in Ivory Coast tektites was only resolved in a Re–Os isotopic study by Koeberl and Shirey [10], who found a distinct meteoritic component (≤ 0.6%, chondritic composition) in Ivory Coast tektites, as well as a smaller one in impact glasses from the Bosumtwi crater. They also found that some samples of the target rocks had elevated contents of the siderophile elements (e.g., Os), but that the Os isotopic signature in these samples was terrestrial, in contrast to the clear extraterrestrial Os component in Ivory Coast tektites and glassy fractions from suevites.

Table 2
Chromium content and isotopic composition of samples from the Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart impact structures, in comparison with literature data for meteorites and other relevant material

Sample	Cr (ppm)	ε(53)
Terrestrial crust[1]	~185	≡ 0
Meteorites		
Ordinary chondrites[2]	~3740	0.48±0.04
E-chondrites[2]	~3100	0.17±0.03
C-chondrites[2]	2650–3600	−0.30 to −0.43
Eucrites	1600–3200	0.7–1.3
Diogenites	6000–13,000	0.4–0.6
Angrites	800–1800	0.4–0.7
Martian meteorites	1600–4000	0.22±0.05
Impact ejecta		
K/T boundary	130–990	−0.33 to −0.40
Archean bed S3	980–3700	−0.37 to −0.41
Late Archean (Hamersley)[3]	280–570	0.12–0.16
Impact craters		
Morokweng[4]	360–410	0.24–0.27
Morokweng[5]	4000	0.38±0.05
Bosumtwi	240	0.30±0.05
Clearwater East	260	0.21±0.05
Lappajärvi	120	0.17±0.05
Rochechouart	300	0.32±0.05

Cr abundances: [1]after [40]; [2]after [47]; uncertainties are 2σ mean. Cr isotopic data for terrestrial crust, meteorites, and impact ejecta from Lugmair and Shukolyukov [32], Shukolyukov and Lugmair [16,34,35]), Shukolyukov et al. [38,54,55]. [3]Data from Shokulyukov et al. [54] for two samples from the ca. 2.6 Ga late Archean spherule layers, Hamersley Basin, Western Australia; [4]data from Koeberl et al. [31]. Note that the second order correction was applied to the Cr isotope data (see text). [5]Data from Maier et al. [43] for a meteorite fragment found within the melt rock.

Subsequently, attempts were made to compare the meteoritic component found in the tektites with the siderophile element anomalies in the target rocks and impact breccias at the Bosumtwi crater. In 1997 and 1999 representative samples of impact breccias and target rocks were collected from the Bosumtwi impact crater to conduct the petrographic, geochemical and paleomagnetic studies (see summary and references in [17]). Major and trace elemental composition, as well as the platinum group element (PGE) abundances, were analyzed in selected target rocks (including shale, graywacke, and two different types of granites) and suevite-derived impact glass samples [46]. These data show elevated Ru, Pd, Os, Ir, and Ag contents in impact glasses, which, on first glance, suggest the presence of a meteoritic contribution. However, the target rocks also have rather high PGE abundances, so that a significant difference between the abundances of these two groups of samples is not obvious. The CI-chondrite-normalized abundances of PGEs, Au, Ag, and Re, for Bosumtwi target rocks and impactites show high and variable Au and Pt abundances in both target rocks and impactites. Thus, the relatively high values of regionally occurring lithologies could very well result from the fact that the Bosumtwi crater is located in an area known for its widespread gold mineralization, as already recognized by, e.g., Jones [45]. Thus, comparison of the indigenous abundances of PGE in the target rocks with PGE abundances in impactite samples does not allow the unambiguous identification of a meteoritic component in the impact breccias.

Even though the PGE abundances in the tektites and in the Bosumtwi impact breccias did not yield any information regarding the exact nature of the impactor at Bosumtwi, the Os isotopic data have clearly shown the presence of some meteoritic component in the tektites and impact glasses. However, Os isotope data do not allow the distinction between a chondritic and an iron meteorite component. Nevertheless, enrichments in Cr (besides Co and Ni) in the tektites compared to the Bosumtwi target rocks indicated that the impactor was more likely a chondritic meteorite than an iron meteorite, as had been suggested by Palme et al. [5].

Thus, this is an ideal case for the application of the Cr isotope method. In an aliquot of Ivory Coast tektite IVC-3395 (the same samples had yielded a meteoritic Os isotope signal, [10]) we found a distinct ^{53}Cr excess of 0.30±0.05 ε (uncertainty is $2\sigma_{mean}$). This value was obtained by repeat measurements of the Cr isotopic composition: 22 runs (300 ratios each). The result unequivocally confirms the presence of an extraterrestrial component, as also shown in Fig. 1. The data also

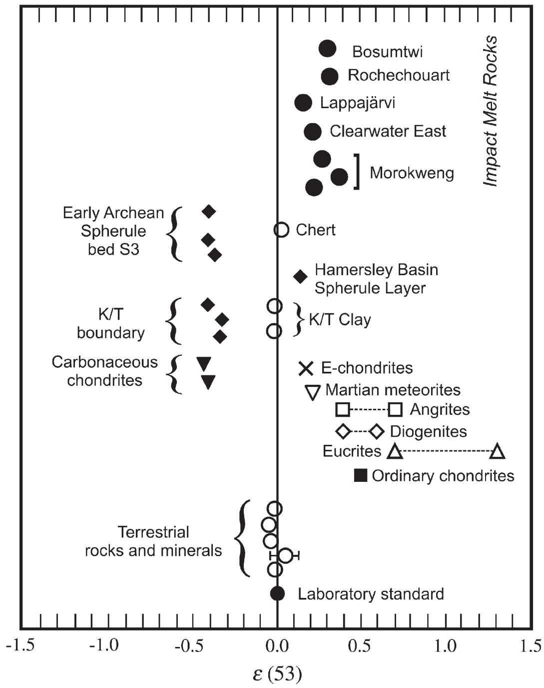

Fig. 1. Chromium content and isotopic composition (in ε units) of samples from the Bosumtwi, Clearwater East, Lappajärvi, and Rochechouart impact structures, in comparison with literature data for meteorites, terrestrial rocks, and distal impact ejecta (from [32,16,34,35,38,54,55,31]). The Cr data are second-order normalized (see text). Data for the early Archean spherules and the K–T boundary indicate a carbonaceous chondritic projectile, whereas the various impact craters discussed here (as well as Morokweng) have ordinary chondrite precursors. The late Archean spherule layer at Hamersley Basin (Western Australia) also has most likely an ordinary chondrite signature.

confirm that a carbonaceous chondrite type projectile can be excluded: as noted above, carbonaceous chondrites have an apparent deficit of ^{53}Cr in this representation. An enstatite chondrite type projectile can also be excluded because their characteristic $^{53}Cr/^{52}Cr$ ratio is significantly smaller, ~ 0.17 ε [35]. Thus, the most likely projectile is an ordinary chondrite type body. Ordinary chondrites have an average ^{53}Cr excess of ~ 0.48 ε [32]. The somewhat lower ^{53}Cr excess in IVC-3395 probably results from the presence of a normal terrestrial chromium component. Thus, the cosmic Cr in IVC-3395 would be ~ 63% or 150 ppm of the measured Cr concentration in IVC-3395 of 240 ppm. The Cr concentration in all classes of the ordinary chondrites is essentially the same: ~ 3740 ppm on average [47]. Therefore, based on the cosmic Cr concentration in IVC-3395 (and taking into account the uncertainty in the $^{53}Cr/^{52}Cr$ ratio) we calculate that this tektite incorporated 3–5% of an ordinary chondrite component. Due to the high indigenous PGE components it is not possible to combine these data with abundance data to derive the type of ordinary chondrite.

4.2. Clearwater East

Clearwater East is unusual among impact craters because of the high concentrations of PGE found in samples of melt rock from the crater. Interestingly, samples from Clearwater West do not show any significant PGE enrichment. Palme et al. [5,6,13] analyzed 13 samples from a 41-m thick section of melt rock from a drill core into Clearwater East and found that the siderophile elements that are characteristic of a meteoritic contamination are heterogeneously distributed in the melt sheet. The meteoritic elements were found to be concentrated in small Ni,Fe-sulfide grains, some of which have contents of the meteoritic elements of up to 300 times the values in the melt rock. Palme et al. [6] measured up to 67 ppb Ir, 0.13 wt.% Ni, 64 ppm Co, and 318 ppm Cr in bulk melt rock samples. They interpreted these high values, together with the elemental ratios of, e.g., Os/Ir, Pd/Ir, Ni/Ir and Ni/Cr, to be consistent with those found in chondrites. In particular, the simultaneous enrichment of Cr along with the PGE strongly suggested that the impacting projectile was a chondrite rather than an iron meteorite and that, relative to CI chondrite, projectile material was present at concentrations of up to 8 wt.% in the impact melt. Grieve et al. [48] noted that the carrier phases of the PGEs might be millerite (NiS) and other Ni,Fe-sulfides and suggested that those might be highly altered fragments of Fe–Ni metal derived from the impactor. Subsequent work on Clearwater East impact rocks in terms of PGE analyses were done by Evans et al. [9] and Schmidt [49]. Evans et al. [9] found that of the various interelement element ratios only Ru/Ir was chondritic within of less than a factor of two, whereas Schmidt [49] found PGE ratios no more than 30% different from chondrite ratios. After correcting for indigenous PGE abundances in the target rocks, Schmidt [49] concluded that the Clearwater East projectile was most likely a carbonaceous chondrite.

However, the interpretation that carbonaceous chondrite material is present at Clearwater East has recently been challenged in a very detailed study of newly measured PGE abundances and ratios by McDonald [14]. One of the problems with the interpretation by Schmidt [49] is the use of only one sample to represent the contribution of the target rocks. Given the irregular distribution of the PGEs in the melt rocks, both the variability of the ratios and the contribution of the indigenous component may be more important than was assumed by Schmidt [49].

The analyses by McDonald [14] show that, in terms of PGE ratios, the Clearwater East signature actually matches most closely with an ordinary chondrite rather than any carbonaceous chondrite. The reason for the discrepancy between PGE ratios and ratios involving other siderophile elements (e.g., Ni/Ir or Cr/Ir) is likely to be that the PGE are retained in metal or sulfide phases and preserve the meteoritic ratios on the scale of a hand sample. On the other hand, this may not be true of Ni, Cr, and Co, as these elements may fractionate into other phases that do not accommodate PGEs. McDonald [14] derived, from PGE ratio plots, a closer similarity to L-chondrites than to the other chondrite types.

We analyzed sample #3 from the collection of Palme et al. [6]. The results (Table 2; Fig. 1) show a distinct ^{53}Cr excess of 0.21±0.05 ε (uncertainty is $2\sigma_{mean}$). As above this value was obtained by repeat measurements of the Cr isotopic composition. The data clearly show that a carbonaceous chondrite type projectile can be excluded, because carbonaceous chondrites show an apparent deficit of ^{53}Cr. The Cr isotopic composition also excludes an enstatite chondrite type projectile because their characteristic $^{53}Cr/^{52}Cr$ ratio is smaller at about 0.17 ε [35]. Fig. 2 shows correlation graphs between the contents of various siderophile elements based solely on the data by Palme et al. [6]. The graphs demonstrate correlations between siderophile elements and seem to agree better with an H-chondrite type projectile. However, based on the PGE abundance ratios, McDonald [14] suggested that L-chondrite ratios provide a closer match.

If the Clearwater East projectile was indeed an ordinary chondrite, a problem based on the initial interpretation of Grieve et al. [48] may also be solved. These authors concluded that the presence of siderophile-rich particles required that the projectile contained Fe–Ni grains. Such grains do not occur in CI chondrites and are very rare in other carbonaceous chondrites, but Fe–Ni alloys are common in ordinary chondrites. An ordinary chondrite projectile, therefore, best explains the Cr isotopic signature, the PGE ratios, as well as the presence of the siderophile-rich particles at Clearwater East.

4.3. Lappajärvi

In contrast to Clearwater East, the impact melt rocks at Lappajärvi (kärnäite) are relatively homogeneous in terms of chemical composition, including the siderophile elements [7,26]. The contents of Ir, for example, are

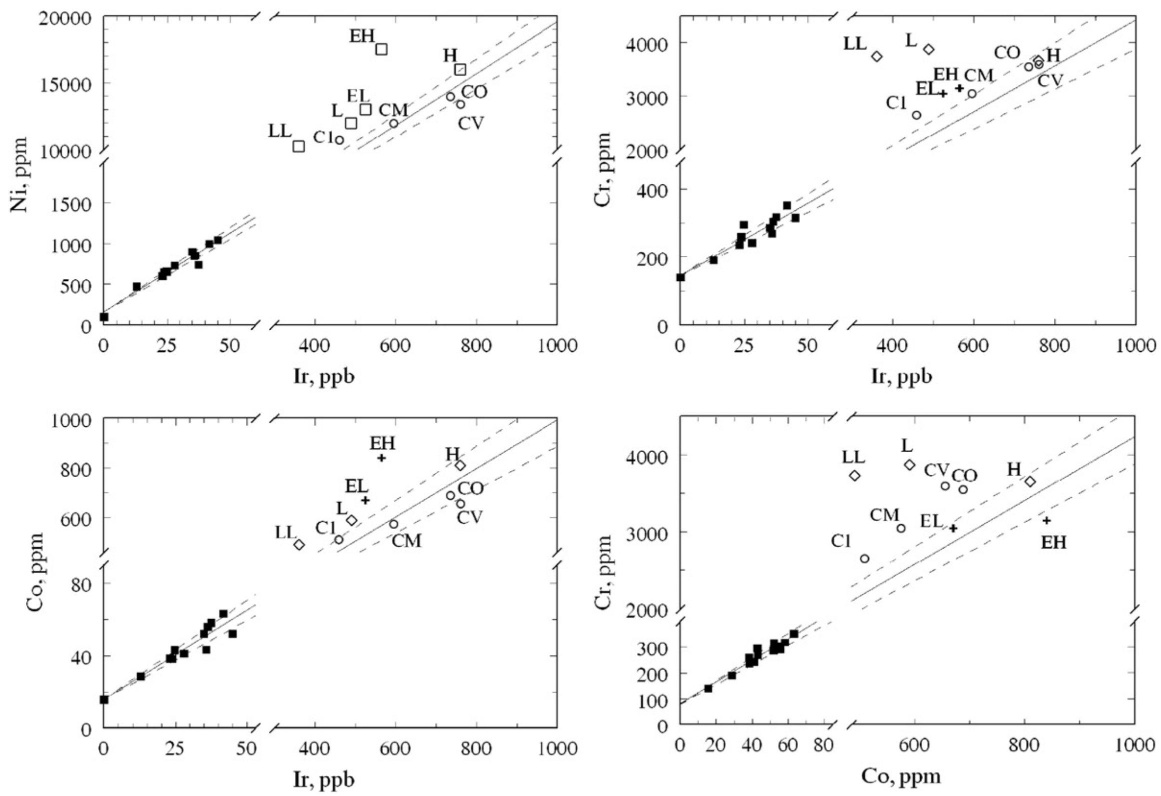

Fig. 2. Correlation diagrams for the abundances of Ir vs. Cr, Co, and Ni, and Cr vs. Co in Clearwater East impact melt rocks. Plotted are abundance data of Palme et al. [6]; the one sample with the highest Ir content of almost 67 ppb was omitted because of possible nugget effects. The dashed lines represent uncertainties on the projection, which were calculated from the uncertainties in the slopes of the regression lines through the experimental data points. They are one sigma standard errors in the coefficients. The concentrations for the different chondrite classes are from Wasson and Kallemeyn [47].

within a factor of 2 within all samples. From interelement ratios such as Ni/Ir and Co/Cr and high concentrations of volatile elements, Palme et al. [7] deduced evidence for a carbonaceous chondritic projectile, but did not exclude an H chondrite either. Analyses by Göbel et al. [50] agreed with the chondrite interpretation. Studies of siderophile element abundances in melt rock samples from a drill core near the center of the structure show relatively consistent high values (e.g., Ir at about 4 ppb) that were taken to show a meteoritic signature [25]. More recently, Tagle et al. [51] reported on PGE analyses of 14 melt rock and 2 target rock samples and their plots of various PGE interelement ratios show that their data agree best with either H chondrite or C1 carbonaceous chondrite data, within error bars.

We analyzed kärnäite (impact melt rock) sample L2-2 from the collection of Palme et al. [7]. The results (Table 2, Fig. 1) show, once again, a distinct ^{53}Cr excess of 0.17 ± 0.05 ε ($2\sigma_{\mathrm{mean}}$ uncertainty). As for the other craters this value was obtained by repeat measurements of the Cr isotopic composition. As for Bosumtwi and Clearwater East, the data are inconsistent with a carbonaceous chondrite type projectile, because, after second order correction, ε(53) is positive and not negative, as would be expected from excess ^{54}Cr in carbonaceous chondrites. The Cr isotopic composition also makes an enstatite chondrite type projectile highly unlikely, because in this case all of the Cr present would have to be of extraterrestrial origin, which is highly unlikely given the basement Cr abundances [26]. The data thus clearly suggest an ordinary chondritic projectile, with about 30% of extraterrestrial chromium in the melt rock. Further indications can be gained from a correlation diagram of Cr vs. Co abundances in samples of Lappajärvi impact melt rock (from [7]), compared with data for average meteorite groups (from [47]), as shown in Fig. 3. The extrapolated correlation of the Lappajärvi melt rock data agree best with H and EL type chondrites, as well as various carbonaceous chondrite groups. However, as the Cr isotope data eliminate carbonaceous chondrites and enstatite chondrites, an H chondritic projectile is the most likely match.

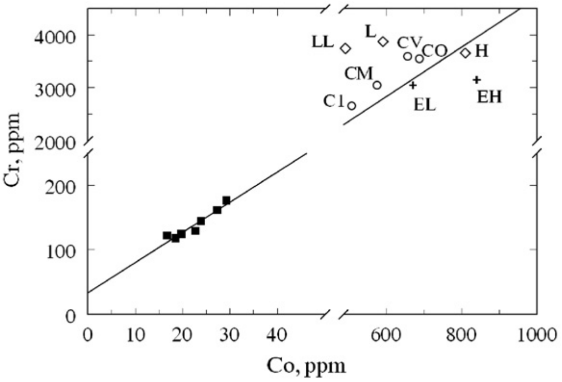

Fig. 3. Correlation diagram of Cr vs. Co abundances in samples of Lappajärvi impact melt rock (from [7]), compared with data for average meteorite groups (from [47]), showing a similarity of the Lappajärvi data with H and EL type chondrites, as well as various carbonaceous chondrite groups. The Cr isotope data eliminate carbonaceous and enstatite chondrites, leaving H chondrites as the most likely match.

4.4. Rochechouart

Previous searches for an extraterrestrial component in suevite and melt rock samples at the Rochechouart impact structure in France have yielded a variety of different results and interpretations. Siderophile trace element data (mostly for PGEs) were interpreted by Janssens et al. [4] and Lambert [52] to indicate an iron meteorite projectile, most probably a group IIA iron meteorite. This was questioned by Horn and El Goresy [53], who found metal veinlets and particles in the crater basement. These particles are rich in Fe, Ni, Cr, and Co and have Ni/Co ratios close to chondritic values. These authors also argued that the high Cr contents cannot be explained by an iron meteorite source. At the same time, Palme et al. [7] reported siderophile and chalcophile element data for a set of melt rock samples from Rochechouart that show clear enrichments in, for example, Ir, Co, Ni, and Cr compared to target rocks (and other breccias). These authors concluded that their data agreed better with a chondritic projectile, and that the interelement ratios are strongly disturbed, probably because of weathering. A similar conclusion was reached by Lambert [28], who stated that “siderophile trace elements are a good indicator for meteoritic contamination, but there may be a serious limitation regarding their use for projectile identification and projectile mass calculations when contaminated formations have been exposed to hydrothermal alteration and weathering”. This problem – that siderophile element ratios, including those of the PGEs, can be subject to post-impact alteration – has been encountered at various other impact structures (see discussion in the reviews by [2,11]). Reimold and Oskierski [29] also favored a stony meteorite projectile.

Due to the apparent enrichment of Cr in the melt rocks compared to the target rocks, the Cr isotope method is ideal to address the question of identifying the projectile type at Rochechouart. We analyzed melt rock sample VR1 from the Valette region [7] and found a strong ^{53}Cr excess of 0.32 ± 0.05 ε ($2\sigma_{mean}$ uncertainty). As above this value was obtained by repeat measurements of the Cr isotopic composition. Again this relatively large excess is inconsistent with both a carbonaceous chondrite type projectile and an enstatite-type meteorite. The data clearly indicate an ordinary chondrite projectile; together with the elemental abundances of Cr and those of the PGEs about 3 wt.% of a chondritic component is present in this particular melt rock sample. Because the interelement ratios, and the correlations between the various elemental abundances are severely disturbed, it is not possible to constrain the type of ordinary chondrite any further.

5. Summary and conclusions

The Cr isotopic method not only allows for the verification that an extraterrestrial component is present in impactites, it also provides information regarding the type of projectiles [16]. The ^{53}Cr relative abundances are measured as the deviations of the ^{53}Cr/^{52}Cr ratio in a sample relative to the standard terrestrial ^{53}Cr/^{52}Cr ratio by high-precision thermal ionization mass spectrometry. Terrestrial rocks do not show any variation in the ^{53}Cr/^{52}Cr ratio, whereas most meteorite groups [16,33,35] show a variable excess of ^{53}Cr relative to terrestrial samples. The range for meteorites is about +0.1 to +1.3 ε, depending on the meteorite type, except for carbonaceous chondrites, which show an apparent deficit in ^{53}Cr of about −0.4 ε after second order correction. Compared to both the use of Os isotopes or PGE abundance ratios the Cr isotope method is, thus, selective not only regarding the Cr source (terrestrial vs. extraterrestrial), but also concerning the meteorite type.

To expand the database of meteorite type identifications at terrestrial impact craters, we analyzed impact melt rock and glass samples from four structures: the 10.5-km diameter, 1.07 Ma Bosumtwi crater (Ghana), the 22-km diameter, 290 Ma Clearwater East structure (Canada), the ca. 21-km diameter, 77 Ma Lappajärvi structure (Finland), and the ca. 25-km diameter, 214 Ma Rochechouart structure (France).

In the case of Bosumtwi, no layers or large bodies of melt rock have been found at or within the crater, and, thus, we analyzed an Ivory Coast tektite (derived from Bosumtwi), which previously was shown to have an extraterrestrial component based on osmium isotope data; however, these data did not allow distinction between chondritic and iron meteorite types. The abundances of the PGEs and other siderophile elements in Ivory Coast tektites and impactites and target rocks from the Bosumtwi crater, the source crater of the Ivory Coast tektites, are all relatively high and did not allow the presence or nature of any meteoritic component in the impactites at Bosumtwi to be determined. Our new Cr isotope data show a distinct ^{53}Cr excess of 0.30±0.05 ε in Ivory Coast tektite IVC3395, which indicates that the Bosumtwi impactor was an ordinary chondrite. Due to the high indigenous abundances of the siderophile elements, it is not possible to use PGE abundances or interelement ratios to further constrain the meteorite type.

At Clearwater East, a melt rock sample revealed also a clear ^{53}Cr excess of 0.21±0.05 ε. This value excludes carbonaceous chondrites (as had been suggested by [49]) and also enstatite chondrites and indicates an ordinary chondrite projectile. Interelement correlations indicate either an H (this work) or an L chondrite affiliation [14], although due to data limitations, and possibly irregular distribution of the different siderophile elements, it is difficult to distinguish between the different types of ordinary chondrite.

The ^{53}Cr isotopic excess in an impact melt rock sample from Lappajärvi was found to be 0.17±0.05 ε, which again excludes carbonaceous and enstatite chondrites. Together with correlations between various siderophile element contents, we conclude that an H-chondrite is the best match for the data; a similar result was obtained by Tagle et al. [51] based on PGE interelement abundance rations (although their data allow for both, carbonaceous chondrites and H chondrites).

A melt rock sample from Rochechouart yielded a ^{53}Cr excess of 0.32±0.05 ε. The amount of extraterrestrial Cr present, as well as interelement ratios, eliminate the possibility of an iron meteorite projectile as had been suggested earlier [4,52]. Our data also eliminate the possibility of a carbonaceous or enstatite chondrite projectile and clearly indicate that Rochechouart was formed by an ordinary chondrite impactor.

Thus so far the Cr isotope method showed, or confirmed, that five mid-size to large impact structures on Earth (the four studied here, and Morokweng — [31]) were formed by ordinary chondrite projectiles. In addition, at the late Eocene impact layer (most likely related to Popigai), Cr isotope data also indicate an ordinary chondrite source [56], as well as for the late Archean Hamersley Basin (Western Australia) impact spherule layer [54]. In contrast, for some much larger impact events – Chicxulub structure/K–T boundary impact event, and some of the Early Archean spherule layers in South Africa and Australia – carbonaceous chondritic impactors are indicated. Despite the complicated and time-consuming analytical procedure, the Cr isotope method has great potential for further projectile identifications at impact structures where elemental abundances and interelement ratios yield ambiguous results.

Acknowledgments

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