The Meteoritical Society

Meteoritics & Planetary Science 58, Nr 5, 722–736 (2023)
doi: 10.1111/maps.13982

Chromium isotopes identify the extraterrestrial component in impactites from Dhala impact structure, India

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(Received 16 June 2022; revision accepted 31 March 2023)

Abstract–The Dhala structure in north-central India is a confirmed complex impact structure of Paleoproterozoic age. The presence of an extraterrestrial component in impactites from the Dhala structure was recognized by geochemical analyses of highly siderophile elements and Os isotopic compositions; however, the impactor type has remained unidentified. This study uses Cr isotope systematics to identify the type of projectile involved in the formation of the Dhala structure. Unlike the composition of siderophile elements (e.g., Ni, Cr, Co, and platinum group elements) and their inter-element ratios that may get compromised due to the extreme energy generated during an impact, Cr isotopes retain the distinct composition of the impactor. The distinct $\varepsilon^{54}Cr$ value of -0.31 ± 0.09 for a Dhala impact melt breccia sample (D6-57) indicates inheritance from an impactor originating within the non-carbonaceous reservoir, that is, the inner Solar System. Based on the Ni/Cr ratio, Os abundance, and Cr isotopic composition of the samples, the impactor is constrained to be of ureilite type. Binary mixing calculations also indicate contamination of the target rock by 0.1–0.3 wt% of material from a ureilite-like impactor. Together with the previously identified impactors that formed El'gygytgyn, Zhamanshin, and Lonar impact structures, the Cr isotopic compositions of the Dhala impactites argue for a much more diverse source of the objects that collided with the Earth over its geological history than has been supposed previously.

INTRODUCTION

Like all rocky bodies of the Solar System, the surface of the Earth has also been modified by extraterrestrial (asteroidal or cometary) impacts, the frequency of which has changed over time (e.g., Mazrouei et al., 2019; Zellner, 2017). A large portion of these impactors (e.g., the Late Heavy Bombardment) has also influenced significantly the present-day chemical makeup of the Earth. Some small- and large-scale impacts throughout the Earth's geological history are evident in the form of variably sized craters, layers of impact ejecta (tektites or spherules), and shock deformation features on its surface (French & Koeberl, 2010). Due to the presence of an atmosphere and geological activities on Earth (e.g., tectonic recycling of the crust, volcanism, weathering, erosion, and sedimentation), most evidence of meteorite impacts are erased over time, but their chemical contributions to the Earth are still significant, especially for the giant and abundant impacts during its accretionary phase (e.g., Fegley et al., 1986; Parkos et al., 2018). Of special interest is the origin of volatile components, particularly water, to the Earth, as the early Earth accreted most likely as a dry rocky body (e.g., Wänke & Dreibus, 1988). The origin of water and other volatiles on Earth has been attributed to the material that was added to the proto-Earth as part of the giant impact and as the late veneer. The relative abundances and isotopic compositions of H, N, and noble gas suggest that the later added material must have had some similarity to carbonaceous (CC) chondrites (e.g., Marty, 2012; Mezger et al., 2021; Piani et al., 2020). This late addition of volatiles by impacts may have been essential in creating a habitable planet that ultimately enabled the emergence of life on Earth. It is ironic that a similar collision of an extraterrestrial projectile with the Earth has been identified as the cause of the Cretaceous-Paleogene (K-Pg, formerly K-T) mass extinction about ~65.5 million years ago (Alroy, 2008). The anomalously high abundance of Ir and other platinum group elements (PGEs) in the K-Pg boundary clay and impact ejecta (spherules, shocked minerals, etc.) deposits led to the discovery of the ~180- to 200-km-diameter Chicxulub impact structure on the Yucatan Peninsula, Mexico (Alvarez et al., 1982; Hildebrand et al., 1991; Smit & Klaver, 1981).

Till date, there are ca. 208 impact structures on the Earth, each confirmed by the presence of mesoscopic (shatter cones) and/or microscopic (e.g., planar deformation features, planar fractures, feather features, diaplectic glasses, etc.) unambiguous evidence of shock metamorphism, ultrahigh-pressure mineral polymorphs (e.g., coesite, stishovite), and/or the chemical traces of extraterrestrial projectile within the impactites (French & Koeberl, 2010; Kenkmann, 2021; Schmieder & Kring, 2020). However, the identification of the projectile is limited to only a very few impact structures (Tagle & Hecht, 2006; see the review by Goderis et al., 2013). The characterization of the projectile component in impactites could help in constraining the source and composition of the impactors and better comprehend the chemical evolution of the Earth. These variations in the nature/type of impactor can also be linked with the modification in dynamics of small asteroid bodies in the solar system. However, tracing and identifying an extraterrestrial impactor is a challenging task since the extreme energy released during an impact event almost completely melts and/or evaporates the impactor. Nevertheless, common impactites such as impact melt breccia, diaplectic glasses, and impact spherules incorporate small amounts ($\leq$1 wt%) of projectile components that can be identified due to their distinct geochemical signature relative to the local or average crustal abundances (e.g., Koeberl, 1998; Koeberl et al., 2007). Most of the conventional methods of detecting and identifying the projectiles are based on recognizing the concentrations of siderophile elements including the PGEs, since meteorites (chondritic or achondritic) have several orders of magnitude higher concentrations of these elements than the target rocks in the upper continental crust (UCC; e.g., French & Koeberl, 2010; Koeberl et al., 2012; Schmidt et al., 1997; Tagle, 2004; Tagle & Berlin, 2008). However, these studies assume that the elemental abundances of siderophile elements (including PGEs) are not significantly fractionated during the impact cratering process which is often not the case. The high thermal energy released during an impact event can change the behavior of the elements and their chemical compounds and in addition, impact-induced heating can also produce hydrothermal systems that may significantly alter the elemental abundances of the impactites (e.g., Déhais et al., 2022; Koeberl et al., 2012; Naumov, 2002), thus obscuring their presence. One way of bypassing the impact-related fractionation and/or hydrothermal alteration of the geochemical signatures is the analysis of Cr and Os isotopes. Almost all meteorite groups have high Cr concentrations and distinct Cr isotopic compositions due to nucleosynthetic variations and radiogenic ingrowth (Lugmair & Shukolyukov, 1998; Trinquier et al., 2007). The high Cr concentration in most meteorites helps in detecting the extraterrestrial component while the distinct isotopic compositions of meteorite groups can help with the identification of the nature of the projectile. Cr shows variations in ^{53}Cr and ^{54}Cr among meteorite classes. The variation in ^{53}Cr composition is the result of the decay of the now-extinct parent radionuclide ^{53}Mn (half-life $T_{1/2}$ = 3.7 $\pm$ 0.4 Ma, Honda & Imamura, 1971) into stable ^{53}Cr in early-accreted Solar System matter. Whereas variations in ^{54}Cr abundances are due to nucleosynthetic heterogeneity (Lugmair & Shukolyukov, 1998; Trinquier et al., 2007) inherited from the solar nebula. The latter is characteristic of different meteorite classes. The studies of Shukolyukov and Lugmair (1998) and Trinquier et al. (2006) were the first that exploited Cr isotopes as tracers of the extraterrestrial impactor at the K-Pg boundary. Shukolyukov and Lugmair (1998) used ^{53}Cr as a tracer for extraterrestrial impactors and identified a CC chondrite-like impactor material. Later, Trinquier et al. (2006) used ^{54}Cr as a tracer for the impactor material and narrowed down the earlier identified CC chondrite-like impactor to CM chondrites. The precise measurements of Cr isotopic abundances in other impact-related materials from different impact sites have successfully provided evidence for the presence of cosmic materials in impactites and have, in some cases, allowed the identification of the type of impactor (Morokweng, Bosumtwi, Clearwater East, Lappajarvi, Rochechouart: Koeberl et al., 2007; El'gygytgyn: Foriel et al., 2013; Zhamanshin: Magna et al., 2017 and Lonar: Mougel et al., 2019). Thus, the Cr isotopes are used in this study to evaluate the presence of extraterrestrial component in impactites and the type of impactor involved in the formation of the 1.7–2.5 Ga old Dhala impact structure, Madhya Pradesh, India.

GEOLOGICAL SETTING OF THE DHALA IMPACT STRUCTURE

The ~11-km diameter Dhala impact structure (N25°17′59.7″; E78°8′3.1″) is located on the western part of the Archean Bundelkhand Craton, India (Pati, Reimold, Koeberl, et al., 2008; Figure 1). The crystalline basement of this impact structure is predominantly composed of granitoids (~2.5 Ga) except few minor enclaves of older metasupracrustal rocks and tonalite–trondhjemite–granodiorite gneisses. The target lithology and impactites of the Dhala structure are overlain by post-impact sediments of the Dhala Formation and the Kaimur Group of Vindhyan Supergroup, which jointly constitutes a mesa-like central elevated area (CEA) of about 5 km^2 in its central part (Figure 1c). Nevertheless, the precise age of the Dhala impact event is not determined so far and is stratigraphically constrained between 2.5 and 1.7 Ga corresponding to the ages of the target granitoids and the post-impact sediments of the Vindhyan Supergroup, respectively (Pati et al., 2010). In the Dhala area, the CEA is surrounded by a ring of >200 monomict breccia outcrops occurring in the outermost annular region of the structure (Singh et al., 2021). These breccia bodies mostly comprise sheared, fractured, and extensively brecciated fragments of the target rocks. Toward the inner edge of the breccia ring, at least eight irregular outcrops of impact melt breccia are observed in a semi-circular pattern between the villages of Maniar and Pagra (Pati et al., 2019). These impact melt rocks are largely of granitic composition. The annular region between the CEA and the monomict breccia ring is mainly occupied by sub-horizontal intercalated purplish-brown siltstone and greenish-white sandy siltstone, together with pockets of conglomerate lenses within a clastic matrix (Pati, Reimold, et al., 2008). Despite the prolonged erosion and multiple post-impact tectono-thermal activities in the region, the impactites of the Dhala structure are relatively well preserved as revealed by the field observations (Pati et al., 2010; Singh et al., 2021). The presence of shatter cones, impact-diagnostic microscopic shock metamorphic features, and the chemical signature of extraterrestrial components within impact melt breccia also confirmed the well-shielded nature of the Dhala structure (Pati et al., 2019 and references therein).

The earlier attempt to identify the projectile component in Dhala impactites was based on concentrations of siderophile elements (Ni, Cr, and Ir) as well as Os isotopic measurements (Pati et al., 2017). The obtained results agreed with the 0.3 wt% contribution of an extraterrestrial component as revealed by one impact melt breccia sample ($^{187}Os/^{188}Os = 0.133$; chondritic impactor?) from the Dhala structure even though the Ir content was below the detection limit for most of the analyzed samples (Pati et al., 2017). More importantly, the study did not exclude the possibility of an iron projectile and therefore could not confirm the precise nature and type of the impactor. This study provides new constraints on the origin of the impactor that led to the formation of the Dhala structure using Cr isotope systematics of the impactites.

SAMPLES AND ANALYTICAL METHODS

Surface and subsurface rock samples from the Dhala impact structure were collected from the CEA, vicinity of the CEA (Pati, Raju, et al., 2008; Pati, Reimold, et al., 2008), impact melt breccia outcrops, and the country rocks. It comprises impact melt/lithic breccia samples (D6-57, D6-33, MCB-7, and MCB-7/2) and a biotite-granitoid (MCB-10/5) and a gabbro (JAD-8/4) from the target lithologies. The meso- and microscopic characterization, trace element content, and Os isotopic composition of the samples were previously published in Pati et al. (2017). For this study, Cr isotope analyses were performed on samples D6-57, MCB-7/2, MCB-7, and JAD-8/4.

A fragment from each sample was powdered with an agate mortar and pestle, homogenized, and dried in an oven at 80°C for about 1 h. Afterwards, 250–300 mg whole-rock material from each sample as well as geostandard IAG OKUM (Certified Reference Materials, International Association of Geoanalysts, Peters & Pettke, 2017) was weighed in 14 mL Teflon® vials and subjected to a concentrated HF-HNO_3 mixture on a hot plate at 90°C for 24 h. Subsequently, the samples were dried and completely digested in concentrated inverse aqua regia on a hot plate at 120°C for 72 h.

The procedure for Cr purification is adopted from Schoenberg et al. (2016) and described in Anand, Pape, Wille, Mezger, Hofmann, et al. (2021) and Anand, Pape, Wille, and Mezger (2021). Each sample was split into two aliquots corresponding to ~10 μg Cr and loaded onto two separate ion exchange columns. The two aliquots were recombined after the elution of Cr from the column. This ensured a low matrix load on a single chromatographic column (Mougel et al., 2019). Typical recovery of Cr was in excess of 90% for the whole column chemistry and total chemistry blanks were below 20 ng, which are negligible compared to the amount of Cr processed through the columns.

Purified Cr samples were analyzed using a Thermo Scientific TRITON Plus Thermal Ionization Mass Spectrometer at the Institute of Geological Sciences, University of Bern. Two micrograms of Cr from each sample was loaded onto an outgassed Re filament and measured at a signal intensity between 7 and 10 V

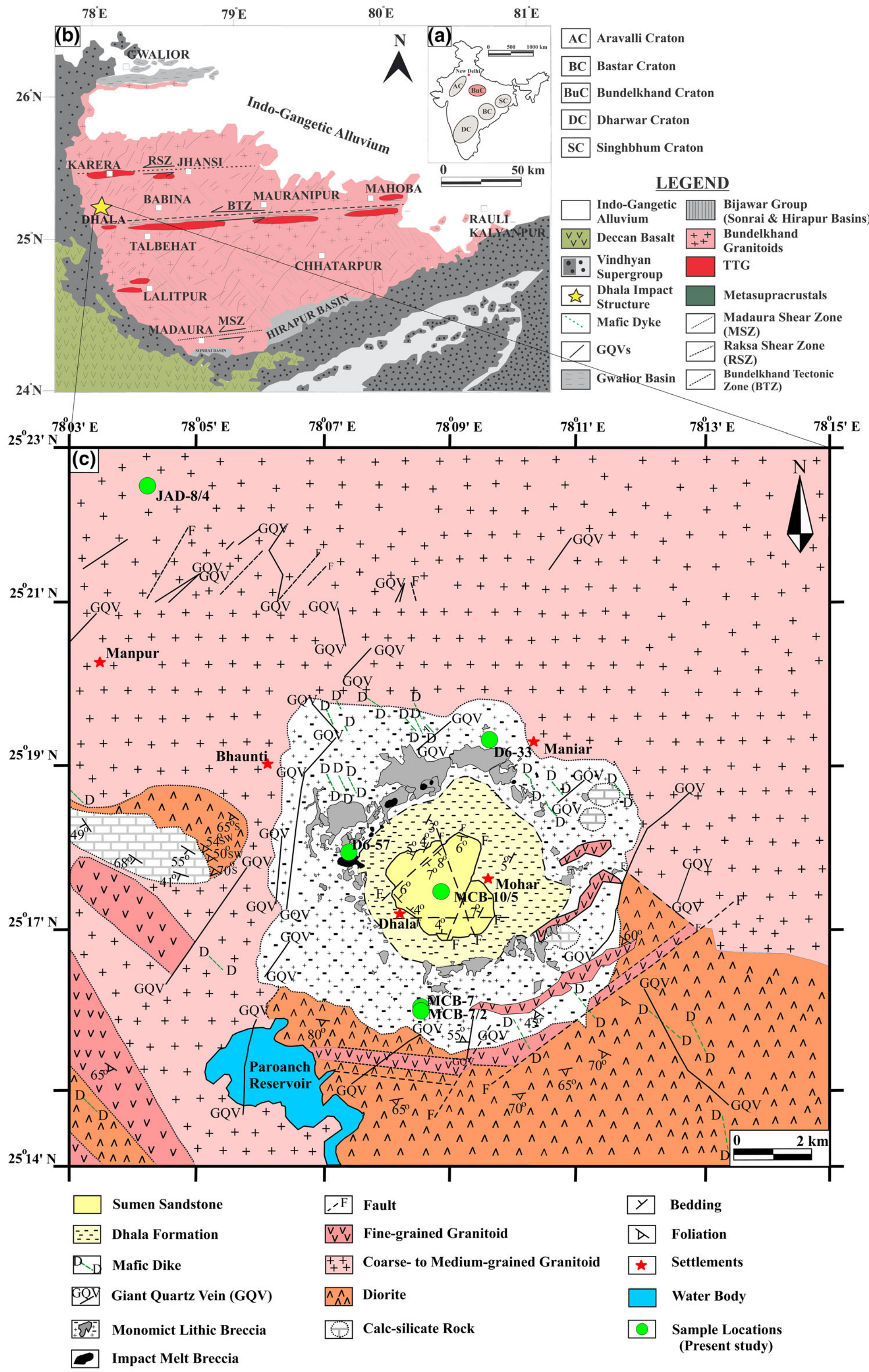

FIGURE 1. (a) The inset map shows five major cratons of India with the Bundelkhand Craton highlighted in pink. (b) Geological map of the Bundelkhand Craton showing different lithological units. The yellow star marks the location of Dhala impact structure. (c) Enlarged geological map of the Dhala impact structure showing the sample locations (green solid circles) and exposures of different rock types around the structure. (Color figure can be viewed at wileyonlinelibrary.com.)

(10^{-11} Ω resistor) for ^{52}Cr. Intensities of ^{50}Cr, ^{51}V, ^{52}Cr, ^{53}Cr, ^{54}Cr, ^{55}Mn, and ^{56}Fe were measured on the Faraday cups L3, L2, L1, C, H1, H2, and H3, respectively (Trinquier, Birck, & Allègre, 2008). Isobaric interference from ^{54}Fe on ^{54}Cr was corrected by monitoring ^{56}Fe and applying the natural $^{56}Fe/^{54}Fe$ ratio. The isotope ^{51}V was measured and ^{49}Ti was monitored to correct for isobaric interference of ^{50}V and ^{50}Ti on ^{50}Cr. However, both ^{49}Ti and ^{51}V intensities remained indistinguishable from background intensities for all samples, verifying the successful separation of V and Ti from Cr during column chromatography. A typical run for a single filament load consisted of 24 blocks with 20 cycles each (integration time = 8.389 s, idle time 3 s), obtained in a static acquisition mode. Gain calibration was done, at the beginning of each analytical session. Amplifiers were rotated and the baseline was measured after every block. The Cr standard reference material NIST SRM 979 was used as a terrestrial reference material. The $^{53}Cr/^{52}Cr$ and $^{54}Cr/^{52}Cr$ ratios were normalized to $^{52}Cr/^{50}Cr = 19.28323$ (Shields et al., 1966) by applying the exponential mass fractionation law and are reported as $\varepsilon^{i}Cr$, where $\varepsilon^{i}Cr = ([^{i}Cr/^{52}Cr]_{sample}/[^{i}Cr/^{52}Cr_{NIST\ SRM\ 979}] - 1) \times 10^4$ (i = 53 or 54).

The $\varepsilon^{i}Cr$ reported for any one sample represents the mean of 2–4 replicate measurements. The replicate measurements for each sample are used to determine the external precision reported as 2 SE (Table 1). The isotope compositions ($\varepsilon^{53}Cr$ and $\varepsilon^{54}Cr$) of each sample are reported relative to the respective mean values of the standard reference material (NIST SRM 979) measured along with the samples in the measurement session (single turret). The external precision (2 SE, $n = 5$) for the standard reference material (NIST SRM 979) in each measurement session was ~0.05 for $\varepsilon^{53}Cr$ and ~0.10 for $\varepsilon^{54}Cr$.

RESULTS

The Cr isotopic compositions of the samples are provided in Table 1. The Cr isotopic compositions of the geostandard IAG OKUM and meteorite sample Allende are consistent with the literature data confirming the analytical accuracy of the Cr isotope measurements (Zhu, Moynier, Schiller, Alexander, et al., 2021 and references therein). Figure 2 shows a comparison between the Cr isotopic composition of the rock samples from the Dhala impact structure, and a compilation of the Cr isotopic composition of the terrestrial rock samples published in the previous studies (Mougel et al., 2018; Trinquier et al., 2007; Trinquier, Birck, Allègre, Göpel, et al., 2008; Zhu, Moynier, Schiller, Alexander, et al., 2021). The Cr isotopic composition of sample JAD-8/4 is indistinguishable compared to the values for terrestrial rock samples. Among the three impact melt/lithic breccia samples, D6-57 plots significantly outside the terrestrial array. This sample shows a resolved negative $\varepsilon^{54}Cr$ anomaly (Figure 2). The Cr isotopic compositions of the other two impact melt/lithic breccia samples MCB-7/2 and MCB-7 are within the uncertainties of the Cr isotopic composition of terrestrial rocks.

DISCUSSION

Figure 2 shows a comparison between the Cr isotopic composition of the rock samples from the Dhala impact structure and different terrestrial rock samples published in the earlier studies (Mougel et al., 2018; Trinquier et al., 2007; Trinquier, Birck, Allègre, Göpel, et al., 2008; Zhu, Moynier, Schiller, Alexander, et al., 2021). Although the Cr isotopic composition of the terrestrial rocks is considered constant due to the homogenization effects of melting and planetary differentiation, a slightly positive correlation between $\varepsilon^{53}Cr$ and $\varepsilon^{54}Cr$ of the terrestrial samples can be observed (Figure 2). This correlation (slope of ~2) has been linked to the "residual" variability in terrestrial rock analyses highlighting the natural Cr isotope fractionation due to geological differentiation processes and/or during Cr purification and TIMS analysis that is not completely removed after correction for mass-dependent variability using the kinetic law for Cr (Zhu, Moynier, Schiller, Alexander, et al., 2021).

The bulk Earth Cr isotopic composition, that is, the average of the Cr isotopic composition of the terrestrial rocks ($\varepsilon^{53}Cr = 0.04 \pm 0.02$, $\varepsilon^{54}Cr = 0.09 \pm 0.03$, Zhu, Moynier, Schiller, Alexander, et al., 2021) can be used to compare with the Cr isotopic composition of the impactites to identify the projectile contribution in the impactites. The low Cr concentration of the granitoid target rocks, best represented by the sample MCB-10/5 ([Cr] = 11.1 ppm, Pati et al., 2017), indicates that the country rocks at the Dhala impact structure are sensitive to detecting and identifying contamination from a chondritic impactor using Cr isotopes due to the high contrast in Cr abundances (Table 1) between the impactor and target lithologies. Relative to the bulk Earth Cr isotopic composition, the Cr isotopic composition of the impactite D6-57 is distinct and shows a resolvable Cr isotope composition that is not terrestrial but rather represents at least partial contribution from the impactor. The Cr isotopic composition of the impactite sample D6-57 does not follow the positive correlation between $\varepsilon^{53}Cr$ and $\varepsilon^{54}Cr$ observed in the terrestrial rocks and hence, its anomalous $\varepsilon^{54}Cr$ value cannot be the result of the "residual" Cr isotope fractionation during Cr purification and/or TIMS

TABLE 1. Chromium isotope compositions and Cr, Ni, and Co abundances (ppm) of the bulk rock samples from Dhala impact structure and meteorites.

Sample	$\varepsilon^{53}Cr \pm 2SE$	$\varepsilon^{54}Cr \pm 2SE$	*n*	Ref.	[Cr] (ppm)	[Ni] (ppm)	[Co] (ppm)	Ni/Cr	Ni/Co	Cr/Co
Dhala										
JAD-8/4	0.01 ± 0.01	0.04 ± 0.05	2		96.6	95	58.4	0.98	1.63	1.65
MCB-7/2	−0.04 ± 0.01	0.14 ± 0.03	2		84.4	58	16.9	0.69	3.43	4.99
MCB-7	0.06 ± 0.03	0.09 ± 0.08	3		68.6	43	18.8	0.63	2.29	3.65
D6-57	0.07 ± 0.04	−0.31 ± 0.09	4		66.8	82	3.99	1.23	20.55	16.74
D6-33	—	—	—		33.0	31	2.05	0.94	15.12	16.10
MCB-10/5	—	—	—		11.1	18	24.2	1.62	0.74	0.46
Standards										
SRM 979	0.00 ± 0.05	0.00 ± 0.10	7		—			—	—	—
OKUM	0.03 ± 0.07	0.01 ± 0.11	5		~2631	~954	~95	~0.4	~10.1	~27.8
Allende	0.07 ± 0.07	0.88 ± 0.12	4		~3407	~13,404	~645	~3.9	~20.8	~5.3
Meteorites										
H	0.17 ± 0.01	−0.38 ± 0.02	13	[1]	~3500	~17,100	~830	4.38 ± 0.42	22.07	4.54
L	0.19 ± 0.04	−0.40 ± 0.04	4	[1]	~3690	~12,400	~580	3.22 ± 0.19	22.67	6.40
LL	0.25 ± 0.02	−0.40 ± 0.08	6	[1]	~3680	~10,600	~480	2.64 ± 0.21	21.19	7.78
EH	0.17 ± 0.03	0.02 ± 0.04	6	[1]	~3300	~18,400	~870	5.79 ± 0.36	21.04	3.59
EL	0.16 ± 0.02	0.02 ± 0.06	6	[1]	~3030	~14,700	~720	4.77 ± 1.03	20.18	4.33
CI	0.28 ± 0.01	1.56 ± 0.04	2	[1]	~2650	~11,000	~505	3.87 ± 0.25	20.87	5.37
CM	0.17 ± 0.03	0.92 ± 0.07	13	[1]	~3050	~12,300	~560	4.01 ± 0.30	21.27	5.25
CO	0.11 ± 0.03	0.90 ± 0.19	5	[1]	~3520	~14,200	~680	3.96 ± 0.09	19.73	5.09
CV	0.11 ± 0.02	0.89 ± 0.12	6	[1]	~3480	~13,200	~640	3.76 ± 0.12	21.26	5.55
Eucrite	0.69 ± 0.05	−0.72 ± 0.02	6	[2]	~2070	~4.5	~7	~0.024	~3.9	~363
Diogenite	0.12 ± 0.13	−0.75 ± 0.05	3	[2]	~5683	~23.24	~16.88	0.004 ± 0.007	1.65 ± 2.97	424.1 ± 406.6
Angrite (plutonic)	0.21 ± 0.06	−0.41 ± 0.14	2	[3]	~2308	~185	~54	~0.080	~2.9	~43
Aubrite	0.00 ± 0.02	−0.12 ± 0.04	1	[4]	~909	~1182	~16	~1.3	~22	~57
Winonaite	0.06 ± 0.01	−0.53 ± 0.01	2	[5]	~1964	~16,300	~345	~8.3	~47	~5.7
Acapulcoite	0.12 ± 0.04	−0.70 ± 0.08	1	[6]	3889	13,095	725.3	3.61 ± 2.67	18.20 ± 1.84	5.36 ± 3.23
Ureilite	0.18 ± 0.04	−0.90 ± 0.04	20	[8]	4383	1315	89	~0.25	11–12	49
Mesosiderite	0.11 ± 0.01	−0.72 ± 0.03	3	[2]	—	—	—	—	—	—

Note: Uncertainties associated with Cr isotopic compositions of the rock samples from the Dhala impact structure are reported as 2 SE. of repeat analyses (*n*) for each sample. Uncertainties associated with Cr isotopic compositions of the meteorite groups are reported as 2 SE of multiple samples (*n*) analyzed within each group. Reference for Ni/Cr, Ni/Co, Co/Cr, and Cr isotopic composition of Dhala rock samples: Pati et al. (2017). References for Cr isotopic composition of meteorites: [1] Zhu, Moynier, Schiller, Alexander, et al. (2021) and references therein; [2] Trinquier et al. (2007); Trinquier, Birck, Allègre, Göpel, et al. (2008); [3] Zhu et al. (2019); [4] Shallowater, Zhu, Moynier, Schiller, Becker, et al. (2021); [5] average of samples NWA 725 and Villabeto de la Pena clast, Schmitz et al. (2016); [6] Ramlat Fasad 529, Anand, Pape, Wille, and Mezger (2021); [7] Li et al. (2018); [8] Zhu et al. (2020), and references therein. References for Ni/Cr, Ni/Co, Co/Cr compositions of meteorites: Allende—Makishima and Nakamura (1997), Chondrites—Tagle and Berlin (2008), Achondrites—Tagle (2004), Patzer et al. (2004), Barrat et al. (2008). References for [Cr], [Ni] and [Co]: Makishima and Nakamura (1997), Wasson and Kallemeyn (1988), Mittlefehldt (2003), Lodders (2003), Tagle (2004), Zhu et al. (2019), Patzer et al. (2004), Yamakawa et al. (2010).

analysis. In contrast to sample D6-57, the Cr isotopic compositions of the other two analyzed impact melt/lithic breccia samples, MCB-7/2 and MCB-7, are within the uncertainties of the sample JAD-8/4 and the Cr isotopic composition of terrestrial rocks. This indicates undetectable presence of meteoritic Cr in these two samples. These results are in agreement with the Re and Os abundances and the Os isotopic composition of the same samples presented in Pati et al. (2017). The authors showed that Re-Os isotopic system identifies terrestrial values for samples MCB-7/2 and MCB-7, but a distinctly low Os isotope composition and abundance for sample D6-57.

The mass-independent isotope variations in ^{54}Cr have been established as a fundamental genealogical tool exhibiting a dichotomy between the NC and CC reservoirs, which most likely represent materials from the inner and outer Solar System, respectively (Kruijer et al., 2020; Warren, 2011 and references therein). The bulk CC reservoir is characterized by excesses in the neutron-rich isotopes of ^{54}Cr, whereas the materials originating in the NC reservoir have depleted ^{54}Cr

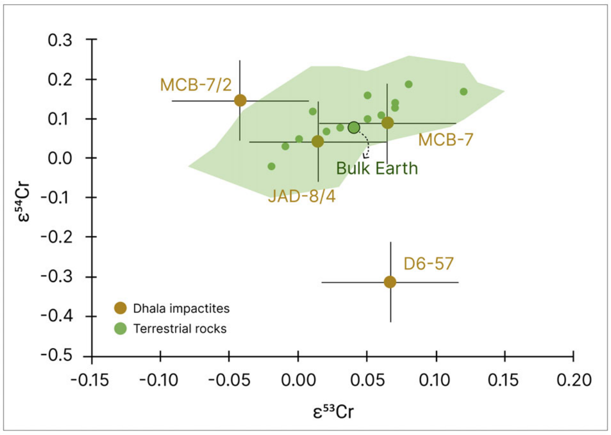

FIGURE 2. A comparison between Cr isotopic composition of the rock samples from the Dhala impact structure and terrestrial rocks (Mougel et al., 2018; Trinquier et al., 2007; Trinquier, Birck, Allègre, Göpel, et al., 2008; Zhu, Moynier, Schiller, Alexander, et al., 2021). Uncertainties associated with the Cr isotopic compositions of the Dhala rock samples are reported as 2 SE of the repeat analyses of the terrestrial Cr standard NIST SRM 979. The shaded region represents an error envelope covering the terrestrial rock Cr isotopic compositions. The bulk Earth Cr isotopic composition refers to the average of the Cr isotopic composition of terrestrial rocks (ε^{53}Cr = 0.04 ± 0.02, ε^{54}Cr = 0.09 ± 0.03, Zhu, Moynier, Schiller, Alexander, et al., 2021). (Color figure can be viewed at wileyonlinelibrary.com.)

signatures with respect to a terrestrial standard (Kleine et al., 2020 and references therein). The depleted ε^{54}Cr isotope composition of the Dhala impactite D6-57 is clearly inherited from an impactor originating in the NC reservoir and this disqualifies all the CC reservoir material (e.g., CC chondrites, ungrouped achondrites) as an impactor for the Dhala structure. In the NC reservoir, enstatite chondrites (or enstatite achondrites or aubrites) have identical ^{54}Cr isotopic composition to that of the Earth and hence, any contamination from an enstatite chondrite-like impactor would not be manifested as an anomalous Cr isotopic composition in the impactites. Ordinary chondrites have been identified to dominate the number of the impactors forming craters on Earth (e.g., Koeberl, 2014; McDonald, 2002; McDonald et al., 2001; Tagle & Claeys, 2005). They are assumed to have originated from the S-type asteroids which are abundant in the main asteroid belt and among the Near-Earth Objects (Ivezic et al., 2001; Morbidelli et al., 2002; Stuart & Binzel, 2004). However, the Cr isotopic compositions (ε^{53}Cr and ε^{54}Cr) of the impactite D6-57 do not match contamination by an ordinary chondrite impactor.

Figure 3 presents binary mixing models between the bulk Earth Cr isotopic composition and the Cr isotopic composition of the meteorite groups within the NC reservoir. The Cr abundances of these meteorite groups

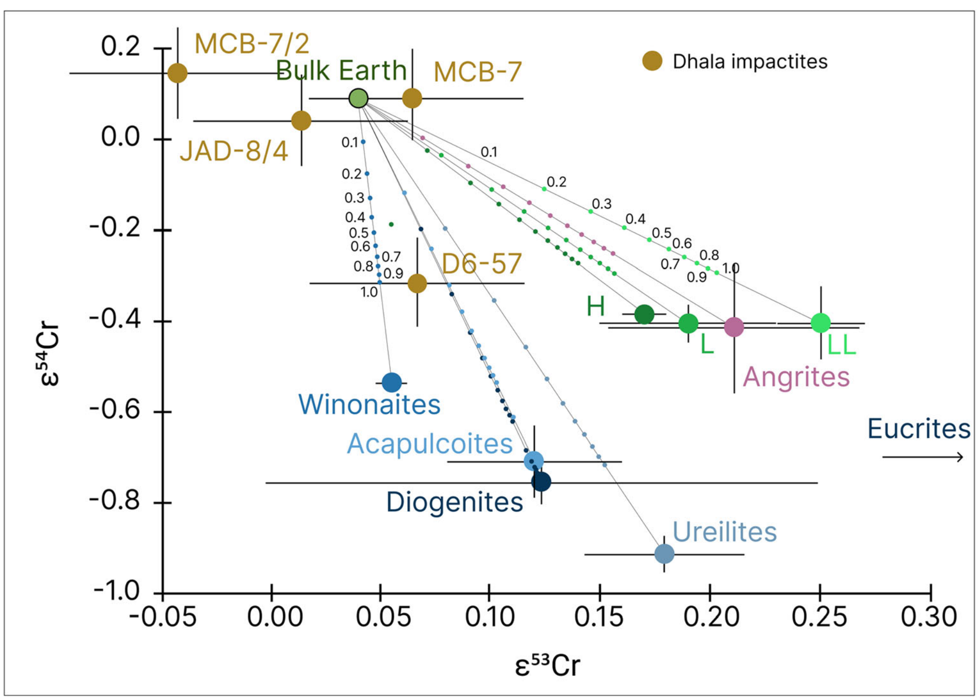

FIGURE 3. Comparison between the Cr isotopic composition of the rock samples from Dhala impact structure and meteorite groups within "non-carbonaceous (NC)" reservoir (references for the data are in Table 1). Solid lines with dots represent binary mixing models between the bulk Earth Cr isotopic composition and different meteorite groups. Each dot on the mixing line represents the amount (wt%) of impactor component considered in the admixture (0.1%, 0.2%, 0.3%, and 0.1%). Confidence envelopes on the mixing lines are removed for the sake of clarity. (Color figure can be viewed at wileyonlinelibrary.com.)

are given in Table 1. The Cr abundance of a biotite granite sample MCB-10/5 is used as an estimate for the Cr abundance of the Dhala target lithology because it is the most abundant litho type in the area. The first-order mixing calculations indicate that the ε^{54}Cr isotopic composition of the impact melt breccia D6-57 would require up to 1 wt% mixing with an ordinary chondrite impactor. For most of the terrestrial impact structures, the impact melt rocks contain meteoritic contamination much lower than 1 wt% (e.g., Koeberl, 1998; Tagle & Hecht, 2006). Ordinary chondrites have a higher Mn/Cr ratio (Mn/Cr = 0.74, Zhu, Moynier, Schiller, Alexander, et al., 2021) than the bulk silicate Earth (Mn/Cr = 0.38, Trinquier, Birck, Allègre, Göpel, et al., 2008) which should be reflected in the radiogenic ε^{53}Cr composition of the impactite sample. However, the sample D6-57 shows contamination from an impactor with a sub-chondritic evolved ε^{53}Cr composition. Within the analytical uncertainties of the Cr isotopic compositions of the Dhala impactites and variations observed in different meteorites groups, an ordinary chondrite-like impactor although unlikely, cannot be completely ruled out as a source of the meteoritic component solely based on the Cr isotope results. However, the combined ε^{53}Cr and ε^{54}Cr compositions of D6-57 are in a much better agreement with an achondritic impactor from the ureilites, diogenites, winonaites, and acapulcoite meteorite groups (Figure 3). The binary mixing calculations indicate that impact melt sample D6-57 is compatible with a contamination of 0.1–0.3 wt% projectile material from a ureilite, diogenite, or an acapulcoite-like impactor (Figure 3). This result is also in good agreement with the estimated 0.3 wt% extraterrestrial (average chondritic) component from the Cr versus Ir concentrations of the sample D6-57 reported in Pati et al. (2017).

The Cr isotopic compositions of the Dhala impactites reported in Table 1 correlate with the Os isotopic compositions of the samples presented in Pati et al. (2017). These authors noted that the impact melt breccias, D6-57 and D6-33, which are dominated by a granitic target component, have distinct ^{187}Re/^{188}Os and ^{187}Os/^{188}Os ratios that nearly overlap with the chondritic and iron meteorites-like compositions (Chen et al., 1998; Horan et al., 1998).

Figure 4 presents the Os data of the rock samples from the Dhala impact structure and different meteorites groups on a ^{187}Os/^{188}Os versus Os abundances plot (references for the data are provided in Table 1). The Os

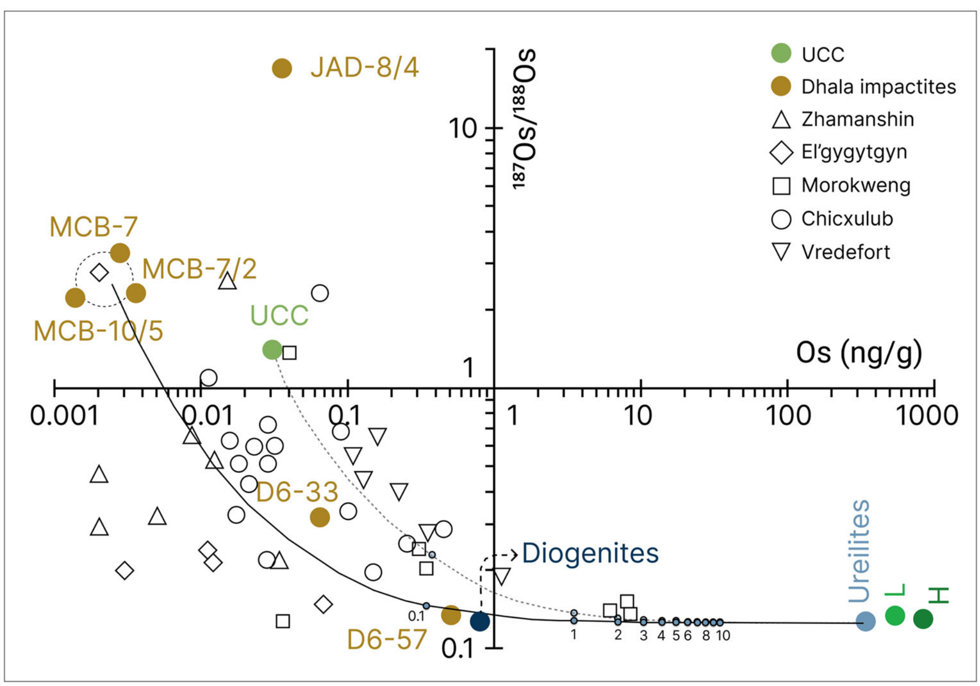

FIGURE 4. ^{187}Os/^{188}Os versus Os composition of the meteorite groups (references for the data are in Table 2), rock samples from the Dhala impact structure, and impactites from the Vredefort (Koeberl et al., 1996), Morokweng (Koeberl et al., 2002), Chicxulub (Gelinas et al., 2004), El'gygytgyn (Goderis et al., 2013), and Zhamanshin (Jonášová et al., 2016; Schulz et al., 2020) impact structures. The solid line represents binary mixing between ureilites and average of the uncontaminated Dhala rock samples, MCB-7/2, MCB-7, and MCB-10/5. The dotted line represents binary mixing between ureilites and the average UCC compositions. Each dot on the mixing lines represents the amount (wt%) of impactor component considered in the admixture (0.1%, 1%, 2%, and 0.10%). (Color figure can be viewed at wileyonlinelibrary.com.)

abundance and Os isotope composition of the impactite D6-57 plot within a region constrained by two binary mixing lines: (i) between ureilites and average of the uncontaminated Dhala rocks, MCB-7/2, MCB-7, and MCB-10/5 and (ii) between ureilites and average UCC compositions. The binary mixing calculation yields 0.1–0.2 wt% projectile contamination in the impactite D6-57 which agrees with 0.1–0.3 wt% projectile contamination as determined from the Cr isotopic compositions (Figure 3). Diogenites, which originate from differentiated parent bodies, are a possible impactor type for the Dhala structure based on Cr isotope compositions (Figure 3). However, based on their extremely low Os abundance (0.81 ± 0.88 ng g^{-1}, Table 2), they can be ruled out because an unrealistic >10 wt% contribution from the projectile would be needed to account for the contamination in D6-57.

TABLE 2. Os concentration and $^{187}Os/^{188}Os$ isotopic composition of the bulk rock samples from the Dhala impact structure, meteorites, and upper continental crust (UCC).

Sample	Rock type	[Os] ± 2SE (ng g^{-1})	$^{187}Os/^{188}Os$ ± 2SE
Dhala			
JAD-8/4	Gabbro	0.0359 ± 0.0004	17.05 ± 0.19
MCB-7/2	Impact melt breccia	0.0036 ± 0.0001	2.33 ± 0.06
MCB-7	Lithic breccia	0.0028 ± 0.0001	3.31 ± 0.09
D6-57	Impact melt breccia	0.5090 ± 0.0064	0.103 ± 0.002
D6-33	Impact melt breccia	0.0654 ± 0.0009	0.317 ± 0.006
MCB-10/5	Biotite granite	0.0014 ± 0.0001	2.228 ± 0.181
UCC		0.0310	1.4 ± 0.3
Meteorites			
CC		612.40 ± 71.63	0.125 ± 0.002
H		837.25 ± 101.36	0.129 ± 0.001
L		544	0.133
LL		391.0 ± 41.6	0.127 ± 0.004
Diogenite		0.81 ± 0.88	0.132 ± 0.013
Acapulcoite		833.3 ± 190.7	—
Ureilite		347.0 ± 161.7	0.126

Note: References for Os concentration and $^{187}Os/^{188}Os$ isotopic composition of Dhala rock samples: Pati et al. (2017), Upper continental crust (UCC): Rudnick and Gao (2003), Peucker-Ehrenbrink and Jahn (2001), Meteorites: CC, H, L, LL—Fischer-Gödde et al. (2010), Diogenites—Dale et al. (2012), Day et al. (2012), Acapulcoites—Patzer et al. (2004), Ureilites—Janssens et al. (1987), Spitz and Boynton (1991), Rankenburg et al. (2007). Uncertainties are reported as 2 SE.

Abundances of moderately siderophile elements (Ni, Cr, and Co) and their inter-element ratios (Ni/Cr, Ni/Co, and Co/Cr) are proven effective tools for the identification of the impactor type (e.g., Koeberl, 1998, 2014). In a few cases, strong enrichments of these siderophile elements in impact melt rocks can be used to distinguish between a chondritic or iron impactor. But, the elemental fractionation during impact melting, vaporization, condensation, and crystallization of the melt rocks are also known to modify or mask chemical signatures of the projectile (e.g., Evans et al., 1993; Mittlefehldt et al., 1992).

Figure 5 shows Ni, Cr, and Co elemental abundances plotted against the SiO_2 composition of the rock samples from the Dhala impact structure. The Ni and Cr abundances of the impactite samples MCB-7/2, MCB-7, and D6-33 and target rock samples MCB-10/5 and JAD-8/4 show a good correlation with their SiO_2 compositions; however, the impact melt breccia sample D6-57 has an anomalous enrichment in Cr and Ni, which may indicate contamination from the impactor. This is also in good agreement with the anomalous Cr isotopic composition of the sample D6-57 shown in this study and its Os elemental and isotopic composition presented in Pati et al. (2017). Furthermore, the Ni/Cr ratio of the impact melt breccia sample D6-57 as shown in Figure 6 is in good agreement with 0.1 wt% admixture of a ureilite-like impactor (and also 0.1–0.3 wt% of other differentiated achondrites such as angrites, diogenites, and eucrites) to the target lithology of the Dhala structure, predominantly represented by sample MCB-10/5. The 0.1 wt% mixing between a ureilite-like impactor and the target rock is also in agreement within uncertainties with the binary mixing results of Cr isotopic composition (~0.3 wt%), Os abundance, and the Os isotopic compositions (0.1–0.2 wt%). Although the Ni/Cr ratio of the impactite D6-57 also agrees with ≤0.1 wt% mixing of an angrite/diogenites/eucrites-like impactor with the basement rock (MCB-10/5); however, these meteorite groups have already been ruled out based on the Cr and Os isotopic compositions of impactites and their Os enrichment. In addition, acapulcoites (and also ordinary chondrites), which were identified as probable impactor type for the Dhala structure, can be ruled out based on the combined Cr and Os isotopic data, plus Os abundances and Ni/Cr ratio of the impact melt breccia sample D6-57 (see forbidden zone in Figure 6), leaving ureilite as the most appropriate type of impactor for Dhala structure (Figure 7).

Chromium isotopes have successfully been used for the detection and identification of the impactor involved in the formation of the Morokweng, Bosumtwi, Clearwater East, Lappajarvi, Rochechouart, El'gygytgyn, Zhamanshin, and Lonar impact structures (Foriel

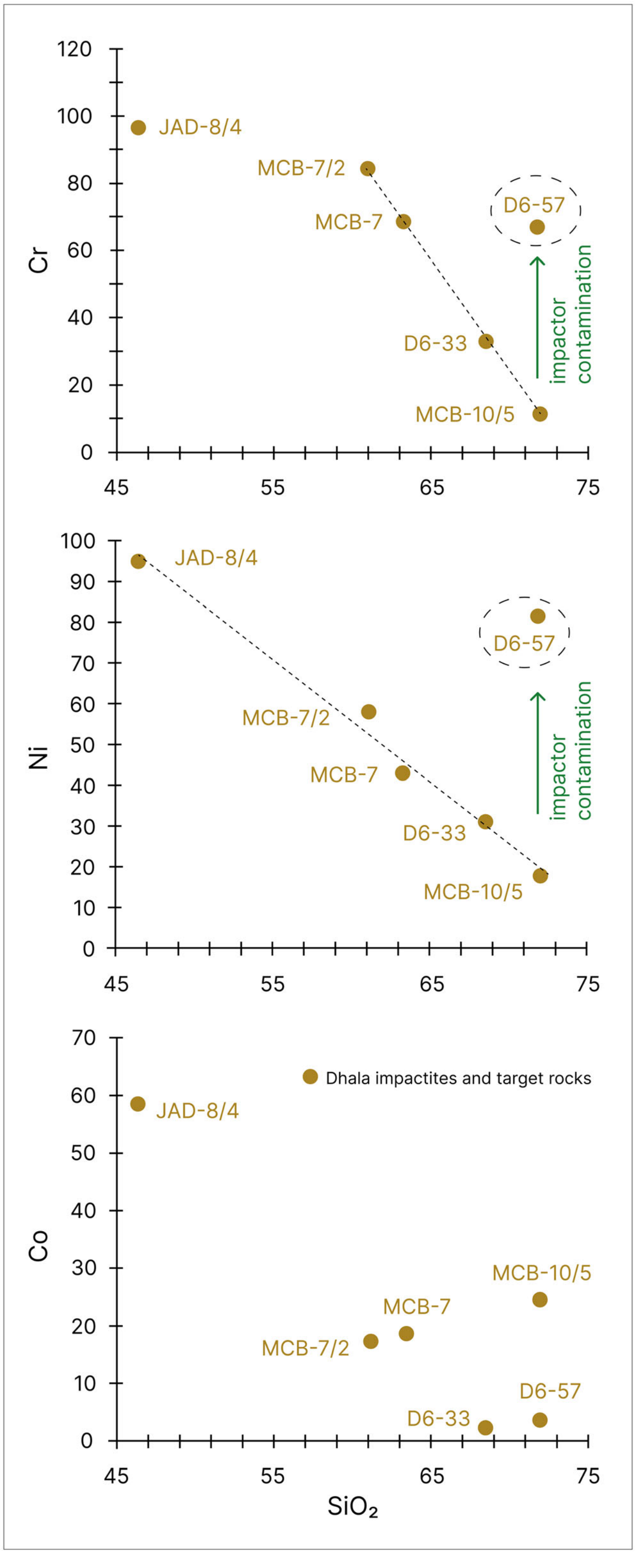

FIGURE 5. Moderately siderophile (Ni, Cr, and Co) elemental abundances versus SiO_2 composition of the rock samples from the Dhala impact structure. The Ni and Cr abundances of the impactite samples MCB-7/2, MCB-7, and D6-33 and target rock samples MCB-10/5 and JAD-8/4 correlate with their SiO_2 concentrations; however, the impact melt breccia sample D6-57 shows an anomalous enrichment in Cr and Ni indicating contamination by the impactor. (Color figure can be viewed at wileyonlinelibrary.com.)

et al., 2013; Koeberl et al., 2007; Magna et al., 2017; Mougel et al., 2019). The identification of the impactors at Morokweng, Bosumtwi, Clearwater East, Lappajarvi, and Rochechouart impact structures is only based on ^{53}Cr (Koeberl et al., 2007). The method relies on the argument that most of the meteorite groups, such as ordinary and enstatite chondrites, primitive achondrites, and other differentiated meteorites, show a variable excess of ^{53}Cr relative to the terrestrial standards, except CC chondrites, which show an apparent deficit in ^{53}Cr of about -0.4 ε after second-order mass fractionation correction based on the $^{54/52}Cr$ ratio, which is assumed to be constant (i.e., $\varepsilon^{54/52}Cr = 0$, Lugmair & Shukolyukov, 1998). These authors analyzed the impact melt rock and glass samples from Morokweng, Bosumtwi, Clearwater East, Lappajarvi, and Rochechouart impact structures and based on the excesses of ^{53}Cr relative to the terrestrial standard (and occasional correlations with siderophile abundances), they concluded that the impactors are related to ordinary chondrites. However, taking the advantage of improved precision on the less abundant and mass independently varying ^{54}Cr and a richer Cr isotope dataset for meteorite groups, recent studies (e.g., Foriel et al., 2013; Magna et al., 2017; Mougel et al., 2019; Trinquier et al., 2006) could improve the efficacy of the Cr isotope approach to reveal the source of impactors at the level of individual meteorite groups within the CC and NC reservoirs. Based on the combined ^{53}Cr-^{54}Cr analyses of the impactites, Foriel et al. (2013), Magna et al. (2017), and Mougel et al. (2019) identified a ureilite-like impactor at the El'gygytgyn structure, a CI chondrite-like impactor for the Zhamanshin structure, and a CM-chondrite-like projectile for the Lonar crater, respectively. The Cr isotopic composition of the Dhala impactites investigated in this study, in conjunction with their Os and Ni/Cr data (Pati et al., 2017), constrain the origin of the impactor to ureilites, which originate from the NC reservoir (Figure 7). Foriel et al. (2013) argued that the discovery of asteroid 2008 TC_3 that exploded after entering the Earth's atmosphere, producing numerous ureilite fragments in a strewn field in Sudan, indicates that such similar bodies may also cause impact structures (see also Goodrich et al., 2019). Together with the identified

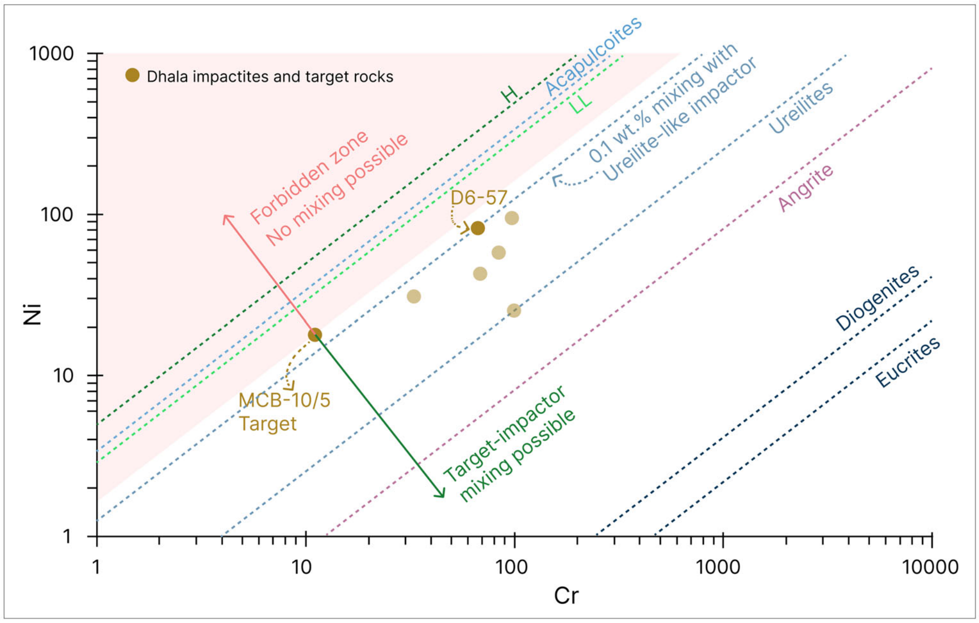

FIGURE 6. Nickel versus Cr abundances of the rock samples from the Dhala impact structure and Ni/Cr correlation trends (dashed lines) for different meteorite groups (references for the data are in Table 1). The impact melt breccia sample D6-57 lies on a Ni/Cr trend line defined by 0.1 wt% binary mixing between ureilite-like impactor and target lithology represented by sample MCB-10/5. The shaded region represents a forbidden zone where mixing between impactors and target lithology cannot explain the Ni/Cr ratio of the impact melt breccia D6-57. (Color figure can be viewed at wileyonlinelibrary.com.)

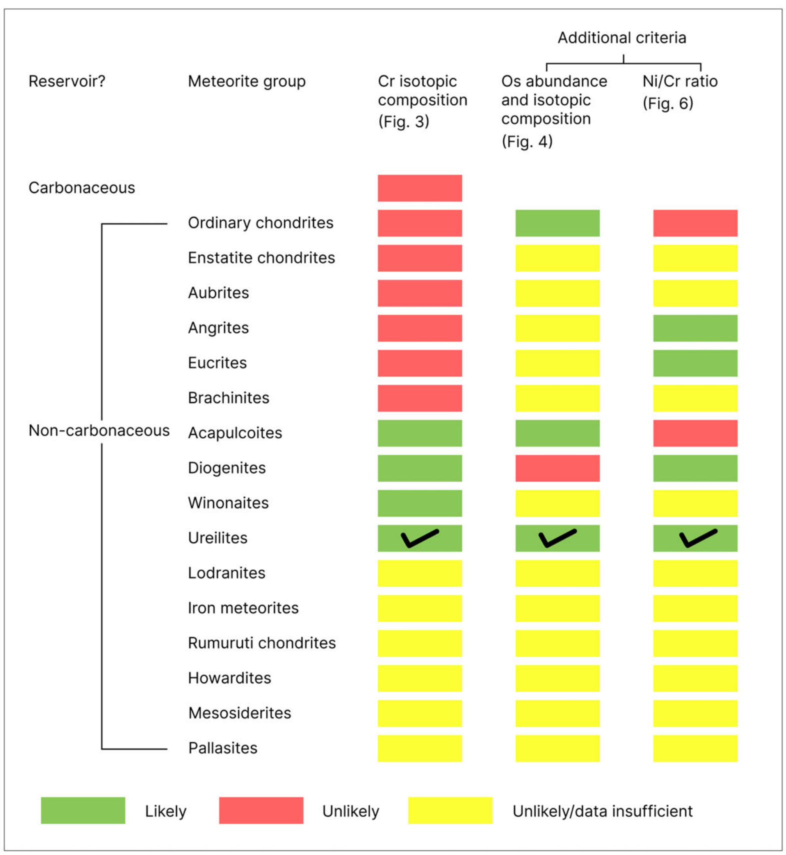

FIGURE 7. Summary of the criteria used to single out the potential type of impactor involved in the formation of the Dhala structure. (Color figure can be viewed at wileyonlinelibrary.com.)

nature of impactors involved in the formation of the El'gygytgyn structure, Zhamanshin structure, and the Lonar crater (Foriel et al., 2013; Magna et al., 2017; Mougel et al., 2019), the ^{53}Cr-^{54}Cr results of the Dhala impactites argue for a much diverse source of the objects that collided with the Earth over its geological history than has been supposed previously. Although ordinary chondrites dominate the current collection of meteorites on Earth, there could be a shift in the origin and composition of the influx of material accreting on Earth. Records of many smaller impacts could have been lost within the background rain of chemical, pyroclastic, and detrital sediments or episodes of volcanic activities (Lowe & Byerly, 2018). There could be an extensive record of the impact history of the early Earth in terrestrial rocks yet to be discovered and with the improvement in analytics, previously determined records should be updated.

Acknowledgments—This study was partially funded by the "Swiss Government Excellence Scholarship (2018.0371)". We acknowledge funding within the framework of the NCCR PlanetS supported by the Swiss National Science Foundation grant no. 51NF40-141881. A.K.S. acknowledges the Council of Scientific and Industrial Research, New Delhi, Government of India, for providing financial assistance through a Shyama Prasad Mukherjee Fellowship (SPM-09/001(0328)/2020-EMR-I). We thank Gopalan Srinivasan for editorial handling and Anne Trinquier and an anonymous reviewer for their constructive comments that helped to improve the manuscript. Open access funding provided by Universitat Bern.

Conflict of Interest Statement—The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability Statement—The data that support the findings of this study are available in the supplementary material of this article.

Editorial Handling—Dr. Gopalan Srinivasan

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