EXTRATERRESTRIAL MATTER ON EARTH: EVIDENCE FROM THE CR ISOTOPES. Shukolyukov¹ and G.W. Lugmair². ¹Scripps Institute of Oceanography, Univ. of California, San Diego, La Jolla CA 92093-0212, ²Max-Planck-Inst. for Chemistry, Cosmochem., PO 3060, 55020 Mainz, Germany.

The recent studies of the $^{53}\text{Mn}$-$^{53}\text{Cr}$ isotope systematic in various solar system objects [1] has provided a method for unequivocally demonstrating the existence of an extraterrestrial component (ETC) in impact ejecta with high concentrations of meteoritic Cr. The method is based on the study of the relative abundances of $^{53}\text{Cr}$, daughter product of the extinct radionuclide $^{53}\text{Mn}$ ($T_{1/2}=3.7$ Ma). The isotopic variations are measured as the deviations of the $^{53}\text{Cr}/^{52}\text{Cr}$ ratios from the standard terrestrial $^{53}\text{Cr}/^{52}\text{Cr}$ ratio, which are usually expressed in $\varepsilon$-units (1 $\varepsilon$ is 1 part in $10^4$, or 0.01%). We developed a technique for high precision mass spectrometric analysis of the Cr isotopic composition in rocks and minerals, which allows measurement of $^{53}\text{Cr}/^{52}\text{Cr}$ variations of less than 1 $\varepsilon$ with an uncertainty of 0.05 to 0.10 $\varepsilon$-units [1].

Because Earth homogenized long after $^{53}\text{Mn}$ had fully decayed, no variation of $^{53}\text{Cr}/^{52}\text{Cr}$ ratios is expected for any terrestrial samples. Indeed, all examined terrestrial samples exhibit the same $^{53}\text{Cr}/^{52}\text{Cr}$ ratio ($\equiv 0$ $\varepsilon$) (Fig. 1). In contrast, most meteorite classes studied so far, including ordinary and enstatite chondrites, primitive achondrites, and various differentiated meteorites are characterized by a variable excess of $^{53}\text{Cr}$ relative to terrestrial samples (Fig. 1), reflecting a heterogeneous distribution of $^{53}\text{Mn}$ in the early solar system [1]. The $^{53}\text{Cr}/^{52}\text{Cr}$ variations in the bulk samples of differentiated meteorites are due to processes of an early Mn/Cr fractionation within their parent bodies. The carbonaceous chondrites show an apparent deficit of $^{53}\text{Cr}$ of $\approx -0.40 \varepsilon$ [2]. However, due to the presence of a pre-solar component the bulk carbonaceous chondrites have an excess of $^{53}\text{Cr}$. Since in our method the $^{54}\text{Cr}/^{52}\text{Cr}$ ratio is used for a second order fractionation correction [1] and, for this purpose, is assumed to be normal, the elevated $^{54}\text{Cr}/^{52}\text{Cr}$ ratio translates into apparent deficit of $^{53}\text{Cr}$. The measured “raw” $^{53}\text{Cr}/^{52}\text{Cr}$ and $^{54}\text{Cr}/^{52}\text{Cr}$ ratios of the bulk CV meteorite Allende (that is, without application of the second order fractionation correction) are $+0.1\pm0.1 \varepsilon$ and $+0.9\pm0.2 \varepsilon$. Preliminary data for bulk Orgueil show that the actual $^{53}\text{Cr}/^{52}\text{Cr}$ ratio is comparable with that for the other undifferentiated asteroid belt meteorites (i.e., an excess of $^{53}\text{Cr}$) and the $^{54}\text{Cr}/^{52}\text{Cr}$ ratio is elevated up to $\approx +1.5 \varepsilon$. Thus, an apparent deficit of $^{53}\text{Cr}$ in the carbonaceous chondrites (when the “normalized” $^{53}\text{Cr}/^{52}\text{Cr}$ ratios are used) is actually due to an excess of $^{54}\text{Cr}$. Nevertheless, because the use of the “raw” $^{53}\text{Cr}/^{52}\text{Cr}$ ratio would drastically decrease the precision, and thus the resolution of our measurements, we prefer to apply the second order fractionation correction, even for the samples with an elevated $^{54}\text{Cr}/^{52}\text{Cr}$ ratio.

The observed difference in $^{53}\text{Cr}/^{52}\text{Cr}$ ratios between Earth and meteorites represents a direct experimental fact that does not involve any models or assumptions. This allows us to unequivocally demonstrate an extraterrestrial component in geological samples on Earth that contain a significant proportion of meteoritic Cr, based on measurements of the Cr isotopic composition.

The Cretaceous/Tertiary (K/T) boundary. The K/T boundary sediments from Stevns Klint, Denmark, and Caravaca, Spain, were found to have a Cr isotopic signature which is very similar to that of the carbonaceous chondrites: from $-0.33$ to $-0.40 \varepsilon$, while the background clays were found to have a normal Cr isotopic composition (Fig. 1) [2]. The “raw” $^{53}\text{Cr}/^{52}\text{Cr}$ and $^{54}\text{Cr}/^{52}\text{Cr}$ ratios of the carbonaceous chondrites and the K/T sediments were also found to be similar. These results indicate that more than 80% of Cr in the K/T sediments originated from a carbonaceous chondrite type impactor. This is the first isotopic evidence for the cosmic origin of the K/T layer and the type of the impactor.

Archean impact deposits in Barberton, South Africa. Criteria that distinguish these beds from typical volcanic and clastic sediments include widespread geographic distribution in a variety of depositional environments, relict quench textures, absence of juvenile volcanioclastics debris, and extreme enrichment of Ir and other platinum group elements [3]. However, some workers argued for terrestrial origin, possibly related to volcanism or gold mineralization [4]. This controversy has been solved based on the recent studies of the Cr isotopes [5]. The Cr isotopic compositions of samples from spherule bed S4 are unquestionably non-terrestrial (Fig. 1). Samples D-4 and C were found to have a clearly anomalous “normalized” $^{53}\text{Cr}/^{52}\text{Cr}$ ratios of $-0.32\pm0.06 \varepsilon$ and $-0.26\pm0.11 \varepsilon$, respectively. The less precise “raw” data for the $^{54}\text{Cr}$ abundances in bed S4 indicate only a small enrichment in this isotope, a result most consistent with a CV-type chondrite source. The background sediments yield normal terrestrial $^{53}\text{Cr}/^{52}\text{Cr}$ ratios. The “normalized” $^{53}\text{Cr}/^{52}\text{Cr}$ ratios in samples 10B and 10G (-
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0.37±0.07 ε and -0.41±0.08 ε, respectively) from another Archean bed from the Sheba Mine (probably bed S3, northern location) also clearly indicate the presence of abundant ETC from a carbonaceous chondrite type source (Fig. 1). The background sample (3 m below the bed) yields normal terrestrial 53Cr/52Cr ratio of +0.01±0.06 ε. These results imply that essentially all Cr in samples 10B and 10G is extraterrestrial. The “raw” 53Cr/52Cr and 54Cr/52Cr ratios in sample 10G (+0.08±0.18 ε and +0.89±0.23 ε) are consistent with this conclusion and suggest a CV type projectile. The sample 5B from the southern part of bed S3 indicates a similar “normalized” 53Cr/52Cr ratio of -0.41±0.10 ε. Thus, the Cr isotope signature in the samples from beds S4 and S3 provide unequivocal evidence of at least two major accretion events at ~3.24 Ga. These deposits are considerably thicker than the K/T deposits and a simple interpretation of the data leads to the conclusion that the projectiles that formed them were at least 20 km in diameter and possibly considerably larger.

ETC in impactites. We have studied impact melt samples from the Morokweng, Clearwater East, Lappajarvi, and Rochechouart impact structures. Based on the chemical analysis [6-8] the projectiles of all these craters are of chondritic origin. The 53Cr excesses (from ~+0.2 ε for Lappajarvi up to ~+0.3 for Rochechouart ) indicate the presence of an ETC. Using the Cr isotope data and the correlations in the siderophile element concentrations, the type of the Morokweng projectile was recently determined to be an L-chondrite [9]. Similarly, the Clearwater East and Lappajarvi projectiles were found to be a H-chondrite type objects. In the Rochechouart melts the interelement ratios among the siderophile elements are heavily disturbed [7]. This makes the assignment to a specific class difficult. However, based on the measured 53Cr/52Cr ratio the enstatite and carbonaceous chondrites can be excluded and, thus, the most likely candidate for the projectile is an ordinary chondrite.

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Fig. 1. Chromium isotope systematics in terrestrial samples, various meteorites, K/T boundary samples, Archean impact deposits, and impactites.