REPORT

METEORITES

Carbonaceous chondrite meteorites experienced fluid flow within the past million years

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Carbonaceous chondritic meteorites are primordial Solar System materials and a source of water delivery to Earth. Fluid flow on the parent bodies of these meteorites is known to have occurred very early in Solar System history (first <4 million years). We analyze short-lived uranium isotopes in carbonaceous chondrites, finding excesses of 234-uranium over 238-uranium and 238-uranium over 230-thorium. These indicate that the fluid-mobile uranium ion U^{6+} moved within the past few 100,000 years. In some meteorites, this time scale is less than the cosmic-ray exposure age, which measures when they were ejected from their parent body into space. Fluid flow occurred after melting of ice, potentially by impact heating, solar heating, or atmospheric ablation. We favor the impact heating hypothesis, which implies that the parent bodies still contain ice.

Carbonaceous chondritic meteorites are composed of material that formed in the early Solar System, perhaps beyond the orbit of Jupiter (*1*). Their elemental composition is similar to that of the Sun (*2*). Carbonaceous chondrites probably contain a record of early Solar System processes that have been modified or obscured to varying degrees by later processing (*3*). Some carbonaceous chondrites (the CI and CM subtypes) have undergone aqueous alteration (reactions with liquid water) as demonstrated by the presence of minerals, including hydrated silicates, magnetite, carbonates, sulfates, and halite (*3*, *4*). Extinct radionuclide (^{53}Mn-^{53}Cr) dating of carbonates has shown that aqueous alteration began a few million years after the beginning of Solar System formation and persisted for at least 2 million to 4 million years (*3*, *5*). Water ice has been observed on the surface of the asteroid (24) Themis (*6*), and the presence of radiogenic ^{87}Sr/^{86}Sr in meteorite calcium sulfates indicates that fluid flow may have occurred recently (*7*).

The Th/U ratio of many chondrites and in the early Solar System (the current Solar System has a ratio Th/U = 3.72) are a subject of debate (*8*, *9*). The present-day relative abundance of these radioactive elements provides information on the time-integrated isotope evolution of the Solar System. However, variation of the Th/U ratio has been found even within chondrules from an individual sample of the carbonaceous chondrite Allende (*10*).

Radioactive uranium decays through a chain of reactions that produces a series of short-lived daughter nuclides. In a closed system, the activity ratio—the relative decay rates of two nuclides in the U-series chain—is exactly unity. The hexavalent uranium ion is fluid mobile, whereas thorium is not, so aqueous alteration is expected to fractionate U isotopes and the Th/U activity ratios, which we denote using parentheses—e.g., (^{230}Th/^{238}U)—to distinguish them from the weight or number ratios. Any resulting disequilibria persist for a time that depends on the half-life of the nuclides involved (*11*). The accessible time resolution of U-series isotope disequilibria is ~5 times the relevant half-life: ~1.25 million years ago for ^{238}U-^{234}U and ~350,000 years ago for ^{230}Th-^{238}U nuclide pairs. Many carbonaceous chondrites have cosmic-ray exposure ages <1 million years ago, indicating that they were only recently detached from their parent bodies (*12*, *13*). Any fluid flow that occurred during their ejection, subsequent transport to Earth, and/or entry into the atmosphere is therefore potentially detectable using U-series disequilibria (*14*, *15*).

We performed U-series measurements for a range of CV, CM, CI, and unclassified C-type meteorites (*16*). These include the Murray and Orgueil breccias, which contain numerous millimeter-sized clasts that have experienced more aqueous alteration than the surrounding host rock. This indicates that brecciation occurred after aqueous alteration. Figure 1A shows that these meteorites all have lower Th/U ratios than the bulk value of 3.72 inferred for the Solar System from time-integrated Pb isotope ratios (*17*). Some of the carbonaceous chondrites have higher Th/U values than Earth's ratio of 3.4, as inferred from the geoneutrino flux (*18*). We measure a range of Th/U ratios from 3.60 to 1.64, with the lowest value being the Tagish Lake meteorite, which has a lower ratio than previously reported (*19*). We confirmed this result by repeating the Tagish Lake measurement. Variation in Th/U both between and within individual carbonaceous chondrites has been documented previously—for example, ratios in Orgueil range from 1.06 to 3.79, Murchison from 2.85 to 4.06, and Allende from 2.05 to 3.46 (*8*). We conclude that the Th/U ratios of carbonaceous chondrites vary substantially even within an individual meteorite (*10*).

We performed sequential leaching experiments (*16*), finding that the disequilibria are pervasive but heterogeneous. These experiments also confirm earlier studies that found that >50% of excess ^{234}U is labile and located in low-temperature phases like calcium sulphates, whereas Th was likely contained in phosphates (*20*). We analyzed ~1-cm-sized samples that we expect to be representative of the bulk meteorites. This rules out an α-particle recoil origin for the disequilibria because that effect occurs on ~50-μm length scales. We find that carbonaceous chondrites all have excesses of ^{234}U that range from 9 to 38%. The largest ^{234}U excesses occur in those meteorites with the lowest U contents (Fig. 1B). Most samples have 4 to 8% deficits of ^{230}Th, whereas the Tagish Lake sample has a 37% deficit of ^{230}Th. Figure 1C compares these carbonaceous chondrites to the U-Th equiline [the line of equal activity ratios, where (^{230}Th/^{232}Th) = (^{238}U/^{232}Th)]. They all have excesses of ^{238}U, implying either (i) variable recent U addition to materials that had a wide range of initial ^{230}Th/^{238}U ratios or (ii) U addition over a range of times to materials with the same ^{230}Th/^{238}U. We conclude that the carbonaceous chondrites we analyzed have all gained ^{234}U relative to ^{238}U and gained ^{238}U relative to ^{230}Th and that the resulting disequilibria have had insufficient time to decay back to equilibrium (Fig. 2A). Because many of the samples lie close to the equiline on Fig. 1C and the initial Th/U ratio is debated, it is difficult to estimate the timing of U addition from these results. An absolute age constraint is imposed by the half-lives of the nuclides. However, an upper limit for the age of the last U migration within these meteorites can be calculated by assuming no initial Th (*21*). This model is shown in Fig. 2A and indicates maximum ages of 80,000 to 180,000 years (assuming a single addition event followed by decay). In some cases, this estimate is less than the meteorite cosmic-ray exposure ages (Fig. 2B).

There are several possible explanations for the observed U-series disequilibria. We first consider terrestrial contamination, even though all of the samples were from meteorite falls collected soon after landing on Earth. Uranium

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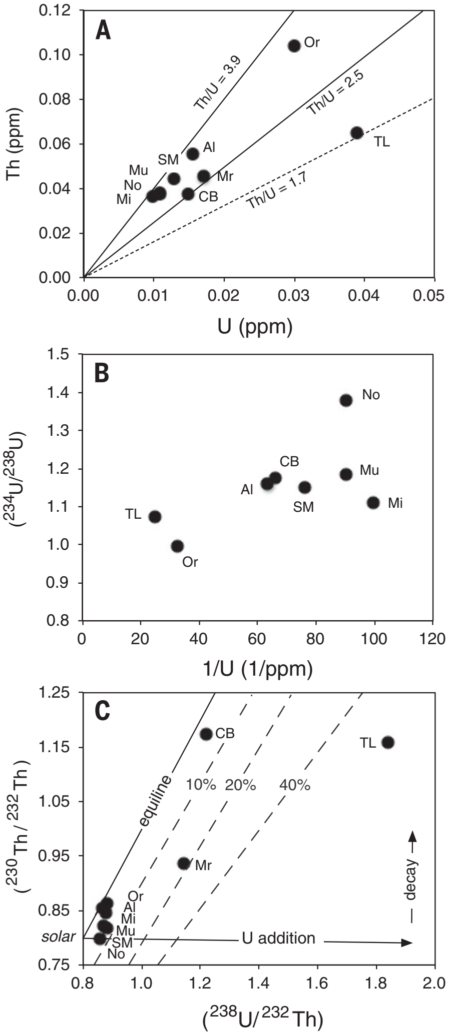

Fig. 1. Elemental and isotopic U and Th variations. (**A**) Abundances of U and Th in the nine carbonaceous chondrites we measured (black dots). The solid and dashed lines indicate constant Th/U ratios as labeled. Four samples have similar Th/U ratios, but Murray, Cold Bokkeveld, Orgueil, and Tagish Lake are enriched in U, i.e., appear below the solid line. (**B**) (^{234}U/^{238}U) ratios plotted against 1/U. The largest ^{234}U excesses [(^{234}U/^{238}U) >1] occur in meteorites with the lowest U contents. (**C**) U-Th isotope diagram with equiline indicating U-Th secular equilibrium. Vectors illustrate the effects of U addition to a composition the same as that of the Solar System by fluid flow and subsequent decay of ^{230}Th with time. Dashed lines indicate percentage of ^{238}U excess. ppm, parts per million; Al, Allende; Mu, Murchison; CB, Cold Bokkeveld; SM, Sutter's Mill; No, Nogoya; TL, Tagish Lake; Or, Orgueil; Mi, Mighei; Mr, Murray. Error bars are smaller than the symbol sizes in all panels.

is fluid mobile in the hexavalent state and ^{234}U resides in recoil-damaged crystallographic sites, but rain does not contain U (*11*). Therefore, if some of the samples were exposed to rainfall before collection (which we cannot exclude), then the result would be loss of U relative to Th, forming a horizontal distribution on the equiline diagram and preferential loss of ^{234}U. However, neither effect is observed in Fig. 1C. River and ground waters contain trace abundances of ^{234}U > ^{238}U > Th (*11*), but none of the samples are known to have interacted with such waters. Allende has excess ^{234}U yet equilibrium ^{238}U-^{230}Th, which is not consistent with acquiring U from surface waters, and it was recovered from a desert environment (*22*). The Tagish Lake sample was collected from a frozen lake surface within a few days of falling and so was never in contact with terrestrial soils, rainfall, or melt water. This sample has the largest disequilibrium we measured [and replicated (*16*)]. Calcium sulfate veins in Orgueil (CI) have been suggested to have a terrestrial origin, in reactions with atmospheric humidity that lead to repeated dissolution and precipitation during sample storage (*23*). Such veins are not a common feature of the other meteorites we have analyzed, and this process would result in alteration areas similar in size to the dimensions of the sulfate veins (<1 mm), with both excesses and deficits of ^{234}U, whereas at the larger scale (~1 cm) measured in our experiments, the U-series would be in equilibrium. We do not measure any samples with deficits of ^{234}U or in equilibrium, though Orgueil is close. We conclude that the disequilibria do not have a terrestrial origin.

Cosmic-ray irradiation cannot produce ^{234}U either by spallation or by neutron capture (*24*). Nor can it explain the observed excesses of ^{238}U activity over ^{230}Th activity. We verified this using numerical calculations and measurements of ^{150}Sm/^{149}Sm as a neutron dosimeter (*16*). Our results indicate that fluid-assisted U migration occurred. This implies that ice persists on carbonaceous chondrite parent bodies to the present day. This is consistent with the occurrence of evaporite minerals like halite in carbonaceous chondrites (*4*) and the presence of water ice on the surface of the asteroid (24) Themis (*6*).

Our results demonstrate that recent fluid flow extended beyond the local scale (>1 cm, the size of our samples). We favor fluid flow similar to that proposed for primordial alteration (*3*). We estimate the scale of recent fluid flow as greater than that of the present diameters of carbonaceous chondrites (>1 m). Continuous fluid flow on the parent bodies of these meteorites since their formation would have destroyed the carbonates that formed in the first 10 million years after Solar System formation (*3*, *5*). We have not found any materials with ^{234}U deficits and ^{230}Th excesses. We consider three alternative hypotheses to explain the U-series disequilibria.

The first possibility is solar heating (*25*), in which meteoroids are warmed above the melting point of ice when their orbits pass close to the Sun (Fig. 3A). We calculate (*16*) that some carbonaceous parent asteroids may retain primordial ice but find that an object that spends a few months warming up at perihelion (the point of closest approach to the Sun) would quickly melt its ice. This would result in a film of fluid evaporating at the surface of the meteoroid, transporting any dissolved U to the exterior of the object. This exterior would be lost during atmospheric entry, so the observed low Th/U, high ^{234}U, and high ^{238}U would not be expected in any chondrite.

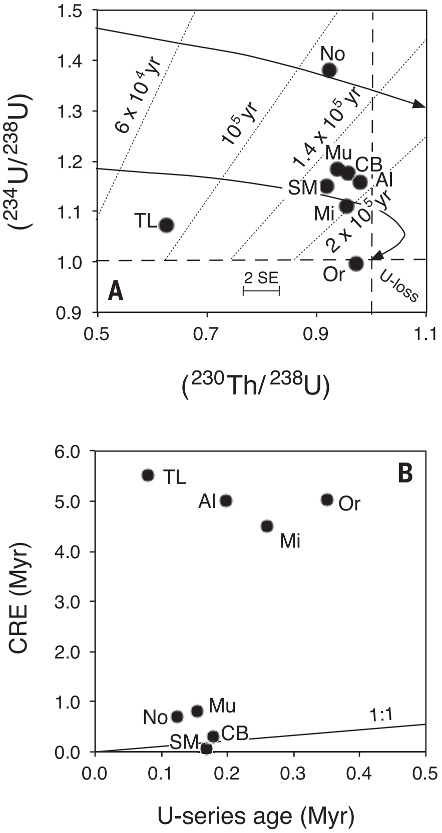

Fig. 2. Uranium-series disequilibria diagrams for carbonaceous chondrites. (**A**) (^{234}U/^{238}U) ratios plotted against (^{230}U/^{238}U) ratios with age calculations; dotted diagonal lines indicate ages for samples that have the same initial disequilibria. Curved lines with arrows indicate time-evolution for samples with different initial activity ratios, and dashed lines indicate equilibrium values [after (*21*)]. The data imply that U mobility (addition) occurred between 140,000 and 200,000 years ago (maximum ages). yr, years. (**B**) U-Th isotope ages inferred from (A) plotted against cosmic-exposure ages (CRE) (table S1). For most meteorites, the CRE age is well above the 1:1 ratio (solid line). Meteorite labels are the same as in Fig. 1. Error bars are smaller than the symbol size, except for (^{230}Th/^{238}U) in (A), which is shown as 2 standard errors (2 SE). Myr, million years.

The second possibility is ablation of a porous carbonaceous chondrite during atmospheric entry, which could drive an ice melt front ahead of the ablation front before the fusion crust forms, leading to $(^{234}\mathrm{U}/^{238}\mathrm{U}) > 1$ and $(^{230}\mathrm{Th}/^{238}\mathrm{U}) < 1$ (compare Figure 2A) in the meteorite interior (Fig. 3B). Although atmospheric entry is too short (a few seconds) for substantial heating to occur, the total mass loss by ablation could be 50 to 80% in the atmosphere. We assume that the porous object has ice partially filling the pores. As the fusion crust migrates inward, silicate melting would be restricted to the outer ~1 mm, but a warm region with liquid water (melted ice) would extend inward about 2 to 3 mm behind the fusion crust. This moving boundary layer (melt front) would eventually traverse about 50 to 80% of the mass of the meteoroid, transporting ^{234}U and ^{238}U inward ahead of the fusion crust. This would leave behind a residue with complementary fractionations that is then removed by ablation, forming ablation melt spherules. These ablation melt spherules would show systematic deficits of ^{234}U and excesses of ^{230}Th. Fusion crusts would show similar disequilibrium, complementary to the chondritic interior. To produce variable disequilibria throughout the chondrite, the U-enriched fluid would need to percolate throughout the interior of the meteorite after the fall, as it warms up. The fluid flow and U migration would then date from entry into the atmosphere. Testing this scenario would require analysis of very short-lived U-series isotopes such as ^{210}Po (half-life of 138 days) in samples collected within days of falling, which was not possible. This hypothesis could explain why the apparent U-series ages are in some cases less than the cosmic-ray exposure ages (Fig. 2B). However, it should also result in a horizontal array on Fig. 1C for Tagish Lake and Murray [using the $(^{230}\mathrm{Th}/^{232}\mathrm{Th})$ ratio, we calculate that the original Th/U = 2.6 and 3.3, respectively]. Most other chondrites lie close to the equiline, i.e., show no disequilibria in $(^{230}\mathrm{Th}/^{238}\mathrm{U})$, implying that the $(^{234}\mathrm{U}/^{238}\mathrm{U})$ ratio must have been perturbed more than 350,000 years ago. Solar heating would result in loss of any ice during the multiple perihelion approaches made by a meteoroid before Earth atmospheric entry, precluding the ablation hypothesis (*16*).

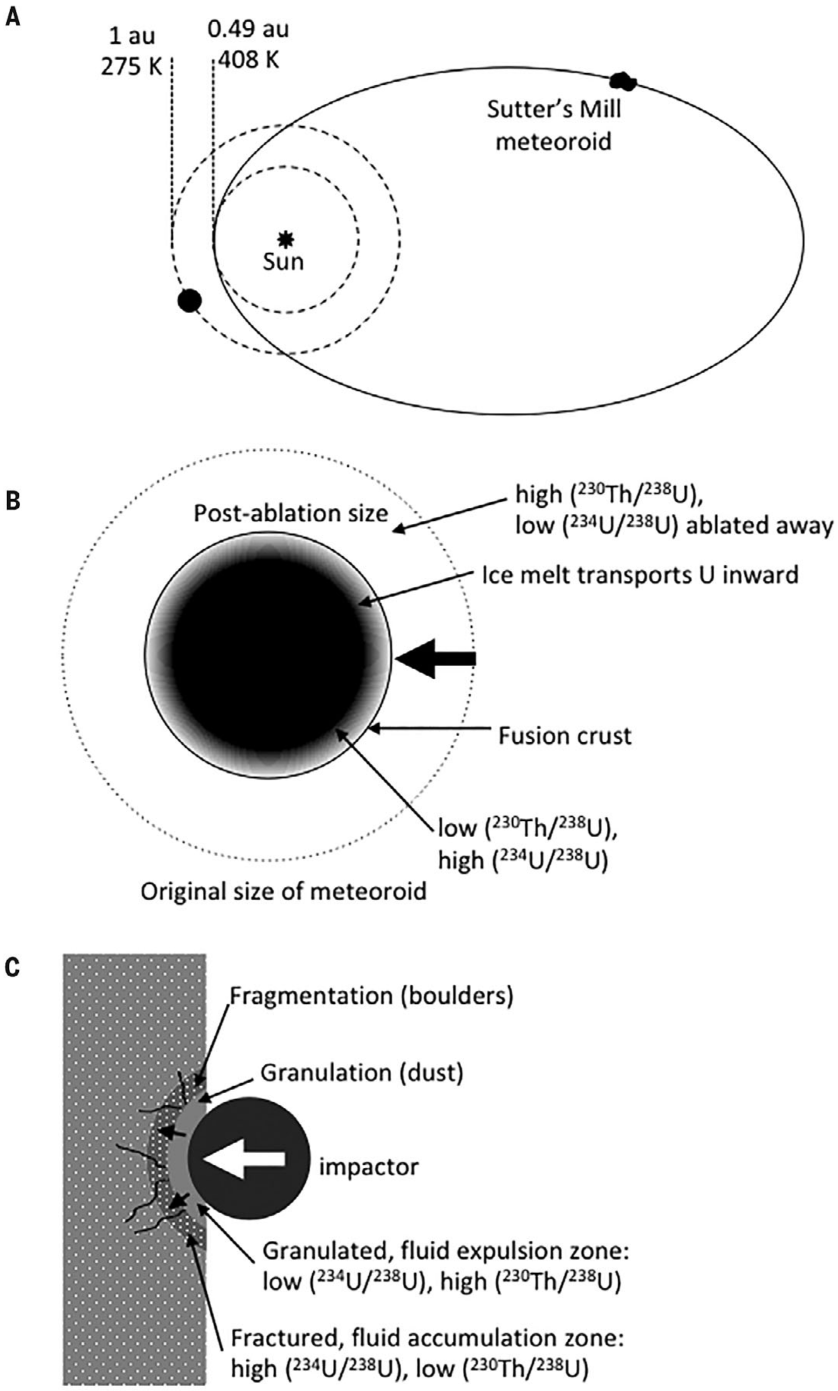

Fig. 3. Hypotheses that could explain the uranium-series disequilibria in carbonaceous chondrites. (**A**) Solar heating scenario with illustration of the orbit of the Sutter's Mill meteoroid, the orbit of Earth, and their respective perihelion temperatures. au, astronomical units. (**B**) Atmospheric ablation scenario. (**C**) Collisional ice melting scenario. See text for descriptions of each scenario.

The third scenario is collisional melting (Fig. 3C) by impact heating when these meteorites were broken off their parent bodies (*26*), melting ice and producing the fluid flow required to explain the observed disequilibria. There is petro-fabric (petrologic alignment of minerals) evidence for impact-related deformation in these meteorites, and the degree of aqueous alteration correlates with the strength of, and may in some cases postdate, fabric formation (*27*). Moderate velocity (~1 km/s) impacts are sufficient to melt ice (*14*). Our observations are consistent with postaccretional surface regolith fluid flow on the parent bodies (*28*) and indicate that they still contain substantial quantities of ice. Large carbonaceous asteroids may have initially experienced convection of their interiors (*29*). After an early sharp burst of aqueous activity (indicated by the ^{53}Mn-^{53}Cr data), the exhaustion of radioactive heat-producing ^{26}Al would have left them frozen, ending fluid flow until episodic impacts ejected parts of the parent bodies. This collisional melting hypothesis relates the observed U-series disequilibria to their ejection from the parent body. Material immediately in front of the impactor would be crushed to dust (granulated), and any ice in it would melt. The resulting fluid would be driven into the porous fractured rock behind the granulated zone. As illustrated in Fig. 3C, this process transports U (and other materials, including organics) into the porous fragments that are then accelerated away from the parent body, some into Earth-crossing orbits. Because impacts can occur at any time, this scenario could explain the wide and nonzero range of apparent U-series ages. Direct samples of solid carbonaceous asteroid material would then show no U-series disequilibria.

Recent fluid transport of U implies potential recent effects on other fluid-mobile species, including organics, alkalis, boron, and other elements. This could potentially affect the radiogenic parent-daughter Rb/Sr ratios—for example, a 10% recent change in a present-day Rb/Sr ratio would be equivalent to a 0.01 difference in a time-integrated ^{87}Sr/^{86}Sr ratio at 4.56 billion years ago (*7*). Differences in initial U isotope and U/Pb ratios could change the dating of the age of the Solar System by 1 million to 2 million years (*13*, *14*). The continuing presence of ice in carbonaceous chondrites could deliver water (*30*) and methane (*31*) to Earth.

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
Figs. S1 to S3
Tables S1 to S3
References (*32*–*43*)

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Carbonaceous chondrite meteorites experienced fluid flow within the past million years

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Recent fluid flow in ancient meteorites

Carbonaceous chondritic meteorites are thought to be fragments broken off parent bodies that orbit in the outer Solar System, largely unaltered since their formation. These meteorites contain evidence of reactions with liquid water that was thought to have been lost or completely frozen billions of years ago. Turner *et al.* examined uranium and thorium isotopes in several carbonaceous chondrites, finding nonequilibrium distributions that imply that uranium ions were transported by fluid flow. Because this signature disappears after several half-lives of the radioactive isotopes, the meteorites must have been exposed to liquid within the past million years. The authors suggest that ice may have melted during the impacts that ejected the meteorites from their parent bodies.

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