Oxygen isotopic heterogeneity in the early Solar System inherited from the protosolar molecular cloud

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INTRODUCTION

The oxygen isotopic composition of the Sun inferred from the measurements of the solar wind returned by the NASA Genesis spacecraft is $\delta^{16}$O-enriched ($\Delta^{17}$O = −28.4 ± 3.6‰) relative to the terrestrial planets, asteroids, and chondrules (−7‰ < $\Delta^{17}$O < 3‰). CaAl-rich inclusions (CAIs), the oldest Solar System solids, approach the Sun's $\Delta^{17}$O. Ultraviolet CO self-shielding resulting in formation of $^{18}$O-rich CO and $^{17}$,18O-enriched water is the currently favored mechanism invoked to explain the observed range of $\Delta^{17}$O. However, the location of CO self-shielding (molecular cloud or protoplanetary disk) remains unknown. Here we show that CAIs with predominantly low ($^{26}$Al/$^{27}$Al)$_\infty$ < $5 \times 10^{-6}$ exhibit a large inter-CAI range of $\Delta^{17}$O, from −40‰ to −5‰. In contrast, CAIs with the canonical ($^{26}$Al/$^{27}$Al)$_\infty$ of $\sim 5 \times 10^{-5}$ from unmetamorphosed carbonaceous chondrites have a limited range of $\Delta^{17}$O, −24 ± 2‰. Because CAIs with low ($^{26}$Al/$^{27}$Al)$_\infty$ are thought to have predated the canonical CAIs and formed within first 10,000–20,000 years of the Solar System evolution, these observations suggest oxygen isotopic heterogeneity in the early solar system was inherited from the protosolar molecular cloud.

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Refactory inclusions are the oldest solids formed in the Solar System. The U-corrected Pb-Pb absolute ages of four CAIs from CV carbonaceous chondrites, 4567.3 ± 0.16 million years (Ma), define a cosmochemical time 0 ($t_0$) of the Solar System evolution (20). Refractory inclusions formed by evaporation, condensation, aggregation, and occasionally melting processes in a gas of approximately solar composition, in a hot protoplanetary disk region exposed to irradiation by solar energetic particles, most likely near the protosun (6, 21, 22). The presence of the mineralogically and isotopically distinct populations of CAIs in different carbonaceous chondrite groups (22–24) suggests that refractory inclusions formed episodically, potentially providing snapshots of isotopic evolution of the protoplanetary disk.

Refactory inclusions recorded heterogeneous distribution of $^{26}$Al, a short-lived radionuclide that decays to $^{26}$Mg with a half-life of ~0.7 Ma, in the early Solar System (8, 25–28). $^{26}$Al is thought to have been injected by a stellar wind from a nearby Wolf-Rayet star (29, 30) or another massive star (31) into the initially $^{26}$Al-poor protosolar molecular cloud shortly before or contemporaneously with its collapse. Most CAIs and AOAs in unmetamorphosed carbonaceous chondrites are characterized by the inferred initial $^{26}$Al/$^{27}$Al ratio ([$^{26}$Al/$^{27}$Al]$_\infty$) of $\sim 5 \times 10^{-5}$, named the canonical ratio (8–10, 22, 32, 33), hereafter referred to as $^{26}$Al-rich CAIs. However, some refractory inclusions in the same meteorites [e.g., platy hibonite crystals (PLACs),...
FUN (fractionation and unidentified nuclear effects) CAIs, and some CAIs rich in corundum (Al$_2$O$_3$), hibonite (CaAl$_{12}$O$_{19}$), or grossite (CaAl$_4$O$_7$) have much lower ($^{26}$Al/$^{27}$Al) than the canonical value, <5 × 10$^{-6}$ [e.g., (25–28, 33, 34)], hereafter referred to as $^{26}$Al-poor CAIs. The $^{26}$Al-poor CAIs are thought to have formed before injection and homogenization of $^{26}$Al in the protoplanetary disk at the canonical level (25–28, 33, 34), i.e., before $t_0$. Because $^{26}$Al appears to have been heterogeneously distributed in the CAI-forming region, the $^{26}$Al-$^{26}$Mg relative chronology cannot be used for defining the age difference between the $^{26}$Al-poor and $^{26}$Al-rich inclusions and the duration of a CAI-forming epoch (27). Astrophysical modeling of a collapse of a protosolar molecular cloud externally polluted by stellar $^{26}$Al suggests that $^{26}$Al-poor CAIs may have formed within the first 10,000 to 20,000 years of the disk evolution, whereas the entire duration of CAI-forming epoch could have lasted ~200,000 to 300,000 years, when the Sun was a class 0-I star (35).

Grossite is one of the first minerals predicted to condense from a gas of solar composition (36). Although grossite-bearing CAIs are
quite rare in most chondrite groups, they are rather common in CH carbonaceous chondrites (37–40). The CH chondrites avoided thermal metamorphism and aqueous alteration on their parent body and therefore must have preserved nebular records largely unchanged (39). Most grossite-bearing CH CAIs are characterized by low \( \frac{^{26}\text{Al}}{^{27}\text{Al}} \), typically <10⁻⁶, consistent with being very ancient refractory inclusions (24, 37, 38, 40). To understand the location of CO self-shielding (molecular cloud versus outer disk), we measured oxygen isotopic compositions (see Materials and Methods) of about 30 randomly selected grossite-bearing refractory inclusions in the paired CH chondrites Acfer 182 and Acfer 214. In the following discussion, we combine these data with the previously reported oxygen-isotope compositions of several grossite-bearing CAIs and AOAs in the genetically related CH3.0 and CB3.0 metal-rich carbonaceous chondrites (24, 40–43).

RESULTS

Most grossite-bearing CAIs measured for oxygen-isotope compositions are isolated inclusions composed of grossite, ±hibonite, ±krotite, perovskite, spinel, and ±gehlenitic melilite, and surrounded by single (melilitic)–layered or multilayered Wark-Lovering rims of spinel, ±hibonite, ±perovskite, melilite, and Al-diopside (Fig. 1, A to D). A grossite-bearing CAI 4C from the CB chondrite QUE 94627 is a fine-grained inclusion composed of concentrically zoned objects made of (from core to edge) ±grossite, spinel, melilite, ±anorthite, diopside, and forsterite with inclusions of Fe,Ni-metal (Fig. 1, E and F). Two grossite-bearing CAIs, 1573-3-13 and I-3, are constituents of AOAs (fig. S1, A to D) (40). Several grossite-bearing CAIs occur as relict inclusions surrounded by a monomineralic spinel layer within the CH porphyritic chondrules (fig. S1, E and F) (43). All CAIs studied are mineralogically pristine and show no evidence for parent body metasomatic alteration or thermal metamorphism.

Oxygen isotopic compositions of the grossite-bearing CAIs studied are listed in table S1 and shown in Fig. 2. Only 2 of 41 (~5%) grossite-bearing inclusions measured from the CH, CH/CB, and CB chondrites, 1573-4-11 (Fig. 1C) and 1573-3-19 (fig. S1G), have heterogeneous oxygen isotopic compositions. In the CAI 1573-4-11, the spinel layer of the Wark-Lovering rim is \(^{16}\text{O}\)-enriched relative to grossite core \((\Delta^{17}\text{O} \sim −24 \text{ and } −17\% \), respectively; because of a small size, the outermost melilitic layer has not been analyzed). In the CAI 1573-3-19, Al,Ti-diopside near the CAI edge is \(^{16}\text{O}\)-enriched relative to grossite core and the spinel layer of the Wark-Lovering rim \([\Delta^{17}\text{O} \sim −27\% \text{ versus } −22\% \), respectively; the anorthite and diopside layers are too thin for our secondary ion mass spectrometry (SIMS) measurements]. The remaining inclusions are isotopically uniform within the analytical uncertainties of our SIMS measurements \((2 \sigma \sim ±2\% \). There is, however, a large range of \(\Delta^{17}\text{O} \) between individual inclusions, from −40 to −10\%. The fine-grained grossite-bearing CAI 4C (Fig. 1, E and F) is uniformly \(^{16}\text{O}\)-depleted \((\Delta^{17}\text{O} \sim −10\% \). The relict grossite-rich CAIs MB4-1-5 (fig. S1E) and MB1-1-1 (fig. S1F) inside porphyritic chondrules are isotopically uniform \((\Delta^{17}\text{O} \sim −33 \text{ and } −5\% \), respectively) and compositionally distinct from the host chondrule phenocrysts \((\Delta^{17}\text{O} \sim +1 \text{ and } −2\% \), respectively).

Only 3 of 59 (~5%) grossite-bearing CH CAIs measured for Al-Mg isotope systematics (24, 37, 38, 40, 43), including CAIs in AOAs and relict CAIs in chondrules, show high excesses of radiogenic \(^{26}\text{Mg}\) corresponding to approximately the canonical \((\frac{^{26}\text{Al}}{^{27}\text{Al}}) \) of ~4 to 5) \(\times10^{-5}\). The remaining CAIs show either no evidence for resolvable excess of radiogenic \(^{26}\text{Mg}\) or they show excesses corresponding to \((\frac{^{26}\text{Al}}{^{27}\text{Al}}) < 5 \times 10^{-6}\) (Fig. 3A).

DISCUSSION

The observed range of \(\Delta^{17}\text{O} \), from −40 to −10\%, among the isotopically uniform grossite-bearing CH and CB CAIs surrounded by Wark-Lovering rims and the uniformly \(^{16}\text{O}\)-depleted grossite-bearing, fine-grained spinel-rich inclusion in QUE 94627 provide a strong evidence for the existence of gaseous reservoirs with different oxygen isotopic composition in the CH and CB CAI-forming region (6). (i) The fine-grained spinel-rich CAIs formed by aggregation of nebular
condensates and avoided subsequent melting (22). (ii) Wark-Lovering rim layers are thought to have formed by melt evaporation and condensation in the CAI-forming region (42). Therefore, the presence of Wark-Lovering rims around the isotopically uniform grossite-bearing CAIs studied, both having similar $\Delta^{17}$O, precludes late-stage reprocessing of these CAIs and oxygen-isotope exchange outside the CAI-forming region. For example, melting of CAIs in an $^{16}$O-poor nebular gas during chondrule formation resulted in partial or complete destruction of Wark-Lovering rims and extensive oxygen-isotope exchange in the melted CAI minerals; the unmelted, relict CAI minerals, however, preserved their initial oxygen isotopic compositions (43). Relict grossite-bearing CAIs MB4-1-5 and MB1-1-1 inside porphyritic chondrules are surrounded by a monomineralic layer of spinel, which probably represents the innermost Wark-Lovering rim layer that survived melting during chondrule formation; the missing mellitite and diopside layers were probably dissolved in the host chondrule melts (42, 43). The rim spinels are compositionally very similar to grossite and mellitite of the relict CAI cores. The host chondrule phenocrysts are $^{18}$O-depleted compared to the relict CAIs (Fig. 3A). We infer that MB4-1-5 and MB1-1-1 largely preserved their original $\Delta^{17}$O, $~$−33 and $~$−5‰, respectively. Therefore, the entire range of $\Delta^{17}$O of nebular gas recorded by the grossite-bearing CH and CB CAIs is from $~$−40 to $~$−5‰.

Two grossite-bearing CAIs showing variations in $\Delta^{17}$O, 1573-4-11 and 1573-3-19, appear to have recorded fluctuations of oxygen isotopic composition of nebular gas during their formation. In the CAI 1573-4-11, the spinel layer of the Wark-Lovering rim is $^{16}$O-enriched relative to grossite core. We infer that oxygen-isotope composition of the nebular gas changed from $^{16}$O-depleted to $^{16}$O-rich during crystallization of spinel. Bodénan et al. (44) have reported fluctuation of oxygen-isotope composition of nebular gas in the opposite direction during condensation of a diopside rim ($\Delta^{17}$O $~$−25‰) around corundum-hibonite-spinel CAI ($\Delta^{17}$O $~$−33‰) from the ALH 77307 (CO3.0) chondrite. Cyclical fluctuations of O-isotope composition of nebular gas during growth of a spinel-rich layered CAI from the MIL 090019 (CO3) chondrite have been inferred by Simon et al. (45). In the CAI 1573-3-19, Al,Ti-diopside near the CAI edge is $^{16}$O-enriched relative to the grossite core and the spinel layer of a discontinuous spinel-anorthite-diopside Wark-Lovering rim. In this case, we cannot exclude change in oxygen-isotope composition of the nebular gas after formation of the Wark-Lovering rim, e.g., during partial melting.

There are three major oxygen-bearing species in the protosolar molecular cloud: CO, H$_2$O, and silicates. The initial oxygen isotopic compositions of these species in the early Solar System are not known. FUV (far ultraviolet) CO self-shielding in the protosolar molecular cloud (16) or in the outer protoplanetary disk (17) is expected to produce $^{16}$O-rich CO and $^{16}$O-poor H$_2$O. The primordial silicates in molecular clouds are largely amorphous (46), and their oxygen isotopic compositions are not directly affected by the CO self-shielding at low temperatures ($<$50 K) expected for these environments, because oxygen-isotope exchange between amorphous silicates and water requires much higher temperatures, $>$500 K (47). Assuming that primordial silicates had a solar $\Delta^{17}$O of −28.8‰, whereas CO and H$_2$O were $^{16}$O-rich ($\Delta^{17}$O $=$ −116‰) and $^{16}$O-depleted ($\Delta^{17}$O $=$ +24‰), respectively, Alexander et al. (48) calculated that evaporation of disk regions enriched in primordial silicates and water-dominated ice relative to CO gas [dust/gas ratio $~$1 to 5] could produce nebular gas with $\Delta^{17}$O ranging from $−$28.8 to $−$5‰; evaporation of dust-depleted regions ($~$0.6 × solar) could produce nebular gas enriched in $^{16}$O relative to the Sun, with a $\Delta^{17}$O of $−$40‰. Therefore, this process could explain the entire range of $\Delta^{17}$O values recorded by the grossite-bearing CH studied. We note that two other types of $^{26}$Al-poor refractory inclusions, PLACs (28) and FUN CAIs (49), also show a larger range of $\Delta^{17}$O compared to the $^{26}$Al-rich CAIs in CR (8), CM (9), and CO (10, 11) chondrites (Fig. 3, A and B).

Quantitative modeling of the CO self-shielding in the parent molecular cloud illustrates how the self-shielding process may have operated. Model results from Lee et al. (50) for CO self-shielding in a collapsing cloud core predict massive $^{16}$O enrichments and depletions in the CO and H$_2$O ice reservoirs, respectively. The protostellar core

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**Fig. 3.** The inferred initial $^{26}$Al/$^{27}$Al ratios and $\Delta^{17}$O values for CAIs from the CV3, CR2, CM2, CH3.0, and CO3.0-3.1 + Acfer 094 (C3.0 ungrouped) carbonaceous chondrites. (A) The initial $^{26}$Al/$^{27}$Al ([$^{26}$Al/$^{27}$Al]$_{i}$) ratios inferred from the internal and model $^{26}$Al/$^{28}$Mg isochrons in CAIs from the CV3, CR2, CM2, CH3.0, and CO3.0-3.1 + Acfer 094 (C3.0 ungrouped) carbonaceous chondrites. The CM SHIBs and CAIs in CR, CO, and CO-like chondrites have approximately the canonical ($^{26}$Al/$^{27}$Al)$_{0}$ of (5.25 ± 0.02) × 10$^{-5}$. The CM PLACs, most FUN/F (fractionation ± unidentified nuclear isotope effects) CAIs from CV, CM, CR, and CO chondrites, and ~95% of the grossite-bearing CH CAIs have much lower ($^{26}$Al/$^{27}$Al)$_{0}$ than the canonical value, <5 × 10$^{-5}$. (B) Average $\Delta^{17}$O of the isotopically uniform CAIs in CM2, CR2, CO3.0, CH3.0, and Acfer 094, and FUN/F CAIs from CV3, CO3, CM2, and CR2 chondrites. FUN CAIs from metamorphosed CV chondrules experienced postcrystallization oxygen-isotope exchange and are isotopically heterogeneous (49). To calculate the average $\Delta^{17}$O of the CV FUN CAIs, only minerals that avoided postcrystallization oxygen-isotope exchange were used; these include spinel, hibonite, forsterite, and most Al,Ti-diopside grains. The predominantly $^{26}$Al-poor CAIs show a much larger range of $\Delta^{17}$O than the $^{26}$Al-rich CAIs. Note that not all CAIs measured for oxygen isotopes were analyzed for Al-Mg isotope systems. Data from (8–10, 24, 26, 32, 34, 37, 38, 49, 40–43) and this study. CH, grossite; SW, Genesis solar wind (1); TF, terrestrial fractionation line.
collapse is modeled as a sequence of Bonnor-Ebert spheres that undergo densification with each step in the sequence, with seven steps total. At time \( t = 0 \) in the collapse model, growth of the protosun commences. After \( 4.6 \times 10^4 \) years, the protosun has reached a mass of \( 1 M_{\odot} \). Figures 4 and 5 show volume fractions and delta values for several important chemical species in the collapse model, including gaseous CO, CO\(_{ce} \), and H\(_2\)O\(_{ice} \) at the inner edge of the model (125 AU) and for a FUV radiation field that is \( 100x \) the local interstellar field \( (G_\odot = 100) \). Most of the collapsing material goes to the formation of the protosun, and all self-shielding–derived isotope signatures are erased in this material owing to high-temperature exchange. Higher angular momentum material falls further out onto the growing disk (35). If the temperature of the infall material accreted onto the disk is sufficiently low that trapping of \( ^{16}\)O-poor \((^{17,18}\)O-rich) H\(_2\)O ice on dust particles occurs, subsequent gravitational settling of icy grains creates an \( ^{16}\)O-poor zone (H\(_2\)O ice) near the developing midplane and an \( ^{16}\)O-rich zone near the surface (CO). Ice/gas separation is an essential component of the CO self-shielding theory for the distribution of oxygen isotopes in inner Solar System materials (16, 17) and provides a natural explanation for the \( ^{17}\)O values of the grossite-rich CAIs reported here. The isotopic result of material fractionation of H\(_2\)O ice and CO gas for the inner edge of the collapse model is shown in Fig. 6. For this model run, which assumes a FUV radiation field that is \( 100x \) the local interstellar medium field, approximately 10% of H\(_2\)O ice must be sequestered on grains during inward migration of nebular material. A coupled collapse-disk model with all oxygen isotopologues and chemical kinetics is needed to fully quantify this scenario. Ideally, a three-dimensional simulation is needed with an anisotropic radiation field and possible dynamical effects to account for possible in-cloud variations in self-shielding isotope signatures.

CO self-shielding also likely occurred early in the outer solar nebula. Because the formation of \(^{26}\)Al-poor CAIs may have occurred within the first 20,000 to 30,000 years of the protoplanetary disk evolution (35), nebular self-shielding would require rapid transfer of CO to the inner disk (~\( 10^4 \) years for a turbulent viscosity parameter \( \alpha \sim 0.1 \); fig. S2) to account for the very low \( ^{17}\)O values of CAIs reported here. In this scenario, a zone of \( ^{16}\)O-enriched CO is formed between the UV disk surface and the deeper x-ray surface, with FUV photons creating the \( ^{12}\)C\(^{16}\)O enrichment and x-rays generating enough ionization to form the high-\( \alpha \) region (fig. S3A). H\(_2\)O derived from CO self-shielding must first be trapped on grains and fall out of the high-\( \alpha \) UV active zone. Calculations show that this may be possible on a time scale of ~\( 10^4 \) years, but this assumes a disk structure similar to the post-infall accretion disk, a structure unlikely to be valid shortly after the start of infall. In addition, the observation that the range of \( ^{17}\)O of CAIs with the canonical \( ([26\text{Al}/27\text{Al}])_0 \) is very limited (Fig. 3) suggests that CO and H\(_2\)O were isotopically homogenized with time until the arrival of \( ^{16}\)O-poor H\(_2\)O brought in by centimeter-scale ice-rich objects. We hypothesize that this contradicts continuous generations of oxygen isotopic effects by CO self-shielding in the outer disk but is consistent with inheritance of isotopic effects in CO and H\(_2\)O from the molecular cloud. A time-evolving coupled collapse-disk model with vertical and radial transport, chemical kinetics, and all oxygen isotopologues is needed to fully evaluate this scenario.

We conclude that the origin of the oxygen isotope variations seen in Solar System materials most likely derives from self-shielding of CO in the parent molecular cloud from which the Solar System formed. Photodissociation and material fractionation (i.e., H\(_2\)O ice sequestration on grains) were primary determinants of the distribution

![Fig. 5. Time evolution of delta values (\( \delta^{17}\)O and \( \delta^{18}\)O) for CO and H\(_2\)O at the inner boundary of the collapse model of Lee et al. Time evolution of delta values (\( \delta^{17}\)O and \( \delta^{18}\)O) for CO and H\(_2\)O at the inner boundary of the collapse model of Lee et al. (50). Delta values are computed relative to SMOW (Standard Mean Ocean Water) would be approximately 60‰ lighter. CO\(_{tot} \)= CO + CO\(_{ce} \), and O\(_{tot} \)= CO\(_{tot} \)+ H\(_2\)O\(_{ice} \)+ atomic O. At time \( t = 0 \), the protosun begins to accumulate mass. At the end of the model run, the protosun is 1 solar mass. Photodissociation of CO is accompanied by a massive isotope enrichment in \( ^{17}\)O and \( ^{18}\)O in the newly formed H\(_2\)O due to self-shielding by \(^{12}\)C\(^{16}\)O. CO isotope fractionation during photodissociation is computed using shielding functions.](http://advances.sciencemag.org/)

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**Fig. 4. Volume fractions of key O-bearing species in the protostellar collapse model of Lee et al. at the inner boundary of the model (125 AU).** Volume fractions of key O-bearing species in the Lee et al. (50) protostellar collapse model at the inner boundary of the model (125 AU). The radiation field is 100x the local interstellar medium FUV radiation field \( (G_\odot = 100) \). Collapse is defined to start at \( t = 0 \). The first CAIs are believed to have formed ~\( 10^3 \) years after the start of the collapse. The photochemical loss of CO releases oxygen atoms that are converted to H\(_2\)O.
of oxygen isotopes in planetary materials formed in the solar nebula. Future modeling of isotopic exchange reactions during accretion of cloud core material will very likely yield a strong constraint on the size of the stellar nursery that gave birth to our Solar System.

MATERIALS AND METHODS

To search for grossite-bearing CAIs, multiple polished sections of Acfer 182 and Acfer 214 were mapped in Mg, Ca, Al, and Ti Kα rays using a 10-μm electron beam, a 15-kV accelerating voltage, a 50-nA beam current, an acquisition time of 10 ms per pixel, and a resolution of 10 μm per pixel with wavelength-dispersive spectrometer detectors with the University of Hawaii’s field-emission electron microprobe JEOL JXA-8500F. Elemental maps in Mg, Ca, and Al Kα were combined using an RGB color scheme (Mg, red; Ca, green; and Al, blue). In these maps, the grossite-bearing CAIs have a dark-blue color. The identified inclusions were studied in backscattered electrons and analyzed for chemical compositions using the UH JEOL JXA-8500F. Quantitative wavelength-dispersive analyses were obtained at 15 kV with counting times of 30 s for peak and for background measurements for each analysis. Natural minerals were used as standards. Electron probe data were reduced via the modified ZAF (Z is the atomic number correction, A is the absorption correction, F is the fluorescence correction) correction procedure PAP (51).

Oxygen isotopic compositions were analyzed in situ with the UH Cameca ims-1280 ion microprobe using the previously described method (52). Briefly, a primary Cs⁺ ion beam focused to ~1 to 2 μm with ~25 pA was used. Three oxygen isotopes were measured simultaneously: $^{16}\text{O}$ was measured on a Faraday cup, and $^{17}\text{O}$ and $^{18}\text{O}$ were measured on electron multipliers. Instrumental fractionation was corrected using terrestrial standards, including San Carlos olivine (for melilite, olivine, and low-Ca pyroxene), augite (for Al-diopside and high-Ca pyroxene), spinel (for hibonite, grossite, krotite, spinel, and perovskite), and Miyake-jima anorthite (for plagioclase).

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/42/eaay2724/DC1

REFERENCES AND NOTES

depleted hibonite-rich CAIs from M.-C. J. absolute chronology and chondrites. 

Multiple generations of aluminum-rich inclusions from Geochim. Cosmochim. Acta


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