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Chondrites and their main components, chondrules, are our guides into the evolution of the Solar System. Investigating the history of chondrules, including their volatile element history and the prevailing conditions of their formation, has implications not only for the understanding of chondrule formation and evolution but for that of larger bodies such as the terrestrial planets. Here we have determined the bulk chemical composition—rare earth, refractory, main group, and volatile element contents—of a suite of chondrules previously dated using the Pb–Pb system. The volatile element contents of chondrules increase with time from ∼1 My after Solar System formation, likely the result of mixing with a volatile-enriched component during chondrule recycling. Variations in the Mn/Na ratios signify changes in redox conditions over time, suggestive of decoupled oxygen and volatile element fugacities, and indicating a decrease in oxygen fugacity and a relative increase in the fugacities of in-fluxing volatiles with time. Within the context of terrestrial planet formation via pebble accretion, these observations corroborate the early formation of Mars under relatively oxidizing conditions and the protracted growth of Earth under more reducing conditions, and further suggest that water and volatile elements in the inner Solar System may not have arrived pairwise.

cosmochemistry | planetary formation | pebble accretion | Solar System evolution | meteorites

Primitive undifferentiated meteorites—chondrites—provide glimpses into the nascent Solar System, as their parent bodies are thought to have accreted within the first ∼5 My after its formation at ∼4,567 Ma, as taken from the age of Ca- and Al-rich inclusions (CAIs) (1). However, bulk chondritic meteorites are complex mélanges of materials that possibly did not form at the same time and/or under the same conditions (2), and, as such, they can provide only generic, fragmented views into the origin and evolution of Solar System solids. Furthermore, when taking both elemental and isotopic composition into account, bulk chondrite origin stories fall short of comprehensively explaining the details of Solar System evolution and terrestrial planet formation (3–8).

Chondrules on the other hand, which are the main constituents of chondrites and, like Earth, are generally depleted in moderately volatile and volatile elements (hereafter collectively termed volatile) compared with bulk chondrites (9–14), may provide more promising pathways toward terrestrial planet formation (15, 16). Pb–Pb ages indicate that most chondrules formed within 1 My after the birth of the Solar System (1, 17). Furthermore, Ca isotopes suggest that the bulk of Earth's mass, and that of the inner Solar System, accreted well within the first million years of the Solar System's existence (18). This latter study further proposes secular volatile element enrichment via the influx of outer Solar System materials, and such events are reflected in the younger, recycled chondrules which record complex heating and remelting of precursor materials over at least 4 My (17, 18). Pebble accretion of chondrule-size materials has proven a viable mechanism to reproduce the basic size and mass distribution of the Solar System on appropriate timescales (15, 16), and, furthermore, corroborates the early influx and subsequent removal of water from the inner Solar System (19). Cumulatively, the above observations highlight chondrules as viable source materials for the majority of inner Solar System bodies, and, in this light, chondrules likely record the processes dominating inner Solar System volatile element systematics.

The behavior of elements during melting, evaporation, and condensation depends on the resident thermodynamic conditions. Exploring the compositional evolution of individual chondrules through time, especially for volatile elements that are acutely sensitive to changing conditions, can shed light on prevailing conditions during chondrule formation and recycling. For example, the Mn/Na ratio of Solar System materials is an established gauge of the prevailing oxygen fugacity conditions (fO_2) during volatilization (20, 21). This is because Mn condenses as a divalent species and Na condenses monovalently, and, as such, they express differential sensitivity to fO_2, i.e., Mn is relatively less volatile than Na at higher fO_2. Taken together, volatile element contents and Mn/Na ratios for individual, Pb–Pb dated chondrules remark on the prevailing history of the early inner Solar System and provide time-specific, detailed information on evolving volatile contents and redox conditions during the planet-forming epoch.

Here, we report the only elemental abundance data that currently exist for individually dated chondrules, all of which have been extracted from two of the most primitive chondrites, the L3.10 ordinary chondrite (OC), NWA 5697, and the CR2 carbonaceous chondrites, NWA 6043 (17). Element abundance data fill important informational gaps for the rare earth element

Significance

We present time-anchored elemental abundance data for some of the Solar System's first solids by tracking Pb–Pb dated chondrule compositions. Volatile element contents generally rise, while redox conditions (based on chondrule Mn/Na ratios) decline beginning ∼1 My after Solar System formation (∼4,567 Ma). These results reflect a continued rise in volatile element contents and their fugacities during chondrule recycling, and early water influx to the inner Solar System followed by its express removal. These observations support the early formation of Mars under oxidizing condition and Earth's protracted growth under more reducing conditions in an environment increasing in volatile contents with time, while also calling into question the coupling of water and volatile elements during Solar System evolution.

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(REE) patterns of L chondrite chondrules, as well as the volatile element contents of both L and CR chondrite chondrules. Volatile element abundances and Mn/Na ratios have then been investigated as a function of age to constrain the evolution of volatile contents and provide first-order insights into the prevailing redox conditions during chondrule formation for the first ~4 My of the inner Solar System. These observations, in turn, are placed within the context of Solar System evolution and terrestrial planet formation to provide time-anchored constraints on the conditions of formation for Mars and Earth, and to assess the paradigmatic view that water and volatile elements arrived in the inner Solar System contemporaneously.

Trace Element Results for Individual Chondrules

Eight chondrules from L3.10 NWA 5697 and three chondrules from CR2 NWA 6043 were analyzed for REEs, six refractory and main component elements—W, Zr, Mo, Ti, Nb, and Cr—and nine volatile elements—Mn, Ag, Sb, Na, Rb, Cs, Zn, Sn, and Cd. REEs have been conventionally listed in order of decreasing atomic radius for clarity and ease of comparison with other studies; all other elements have been reported in order of increasing volatility under solar nebula conditions (22). All element data are reported herein as La- and CI chondrite-normalized abundances (see *Materials and Methods*). Host meteorite, Pb–Pb age, and other relevant details for all chondrules can be found in Table 1 (17), along with calculated Mn/Na ratios for each chondrule. La- and CI-normalized concentrations for volatile elements are reported in Fig. 1 and Table 2, and La- and CI-normalized data for all elements are reported in *SI Appendix*, Table S1 along with literature data, with La-normalized abundances in *SI Appendix*, Table S2 and literature data in *SI Appendix*, Table S3.

The REE abundances for CR chondrite chondrules are in good agreement with literature values (*SI Appendix*, Fig. S1 and Table S1), validating the choice of La as a normalizing agent and, more importantly, the data set as a whole. REE abundance data for L chondrite chondrules are by-and-large novel, and, as such, there are very limited data for comparison (9); however, the few available literature values are in good agreement, and the data presented herein shed light on L chondrite chondrule composition (*SI Appendix*, Fig. S1 and Table S1). In both the L and CR chondrite chondrules, refractory and main group elements are relatively more enriched than volatile elements, especially those with 50% condensation temperatures (T_{50}) below ~800 K, consistent with the generally volatile-depleted nature of chondrules relative to bulk meteorites (13) (Fig. 1).

Table 1. Host meteorite, Pb–Pb age and information for individual chondrules

Chondrule	Pb–Pb age	Type	Ol. Fa#	Mn/Na
NWA 5697 (L3.10)				
2-C1	4,567.57 ± 0.56	I	6.1	0.5
5-C2	4,567.54 ± 0.52	II	19	2.2
5-C10	4,567.41 ± 0.57	II	19	2.7
D-C3	4,566.58 ± 0.57	II	27	3.3
5-C4	4,566.56 ± 0.53	II	12	2.9
3-C5	4,566.20 ± 0.63	II	19	2.5
11-C2	4,564.65 ± 0.46	II	22	0.8
3-C2	4,563.64 ± 0.51	II	15	1.2
NWA 6043 (CR2)				
1-C2	4,567.26 ± 0.37	I	9.8	1.5
2-C2	4,565.06 ± 0.40	II	23.9	1.9
2-C4	4,563.64 ± 0.51	II	13.5	1.5

Pb–Pb ages, chondrule type, and olivine fayalite number (Ol. Fa#) from ref. 17. Manganese-to-sodium ratios (Mn/Na) are from the current study.

The majority of data are for chondrules from L3.10 NWA 5697, providing a more complete picture of L chondrite chondrule evolution. Therefore, discussion focuses largely on these chondrules, and, where possible, complementary observations are presented for the chondrules of the CR2 chondrite NWA 6043.

Comparison with Other Chondrules, Bulk Chondrites, and the Bulk Earth

REE patterns for both L and CR (to a lesser degree) chondrite chondrules are similar to that of their host meteorites (*SI Appendix*, Fig. S1), indicating that the REE contents of chondrules largely set the budget of these elements in the bulk. This is likely owing to chondrules constituting over half the bulk or more by volume in both cases (>60% for OCs) (23, 24). Bulk chondritic patterns are more closely mirrored by older chondrules in CR chondrite chondrules for refractory and main group elements (Fig. 1). For L chondrite chondrules, La- and CI-normalized data indicate that refractory and main group elements are similar to bulk L chondrites and chondrules (where data exist) (Fig. 1). For both L and CR chondrite chondrules, these observations suggest that chondrules generally control bulk chondrite element systematics, as has been previously suggested (25).

Volatile element contents for L chondrite chondrules from the current study generally fall in line with the so-called volatility trend (e.g., Na, Rb, and Cs), while bulk L chondrite values for these elements display enrichments relative to this trend, suggesting an enriched component for these elements in the host meteorite. The general volatile depletion pattern of the L chondrite chondrules is in good agreement with that for the bulk Earth (Fig. 1). Data for CR chondrules in the current work, as well as in the literature, display relatively higher variations, making further discussion largely untenable.

Age-Dependent Element Systematics

The current study provides compositional data for chondrules that are anchored in time. The abundances of Ag and Sb, and, to a lesser degree, Zn, display stochastic variations concomitant with CAI formation at ~4,567 Ma (1), followed by a general depletion and subsequent increase in abundance (Fig. 2 and Table 2). Fig. 2 reports the abundances of Ag and Sb as a function of chondrule age, as these elements display the most coherent positive trend with time, while elements of higher volatility show more variability. These data provide observational evidence for a general increase in volatile contents in the inner Solar System through time. This volatile element evolution pathway for the inner Solar System is supported by a wealth of literature across a broad range of methodological approaches, e.g., experimental petrology (26), isotope geochemistry (18, 27), and astrophysicochemical modeling (19, 28, 29). These lines of evidence, along with the likely early formation of most chondrules and their later recycling in a volatile-enhanced environment (17, 18), support the view that a fraction of the chondrule population in OCs were formed (recycled) under noncanonical conditions marked by increased volatile contents (30), and, moreover, provide time constraints for the generation of such chondrule populations. The agreement between chondrule volatile element contents and that of the bulk Earth (Fig. 1 and Table 2) further evidence a robust compositional relationship between the two, strengthening the argument that chondrule accretion played an integral role in terrestrial planet formation. More broadly speaking, the current data suggest that increased volatile flux to the inner Solar System began ~1 My after CAI formation and proceeded for at least the next 3 My (Fig. 2 and Table 2), corroborating and elaborating upon previous observations for these same chondrules (18).

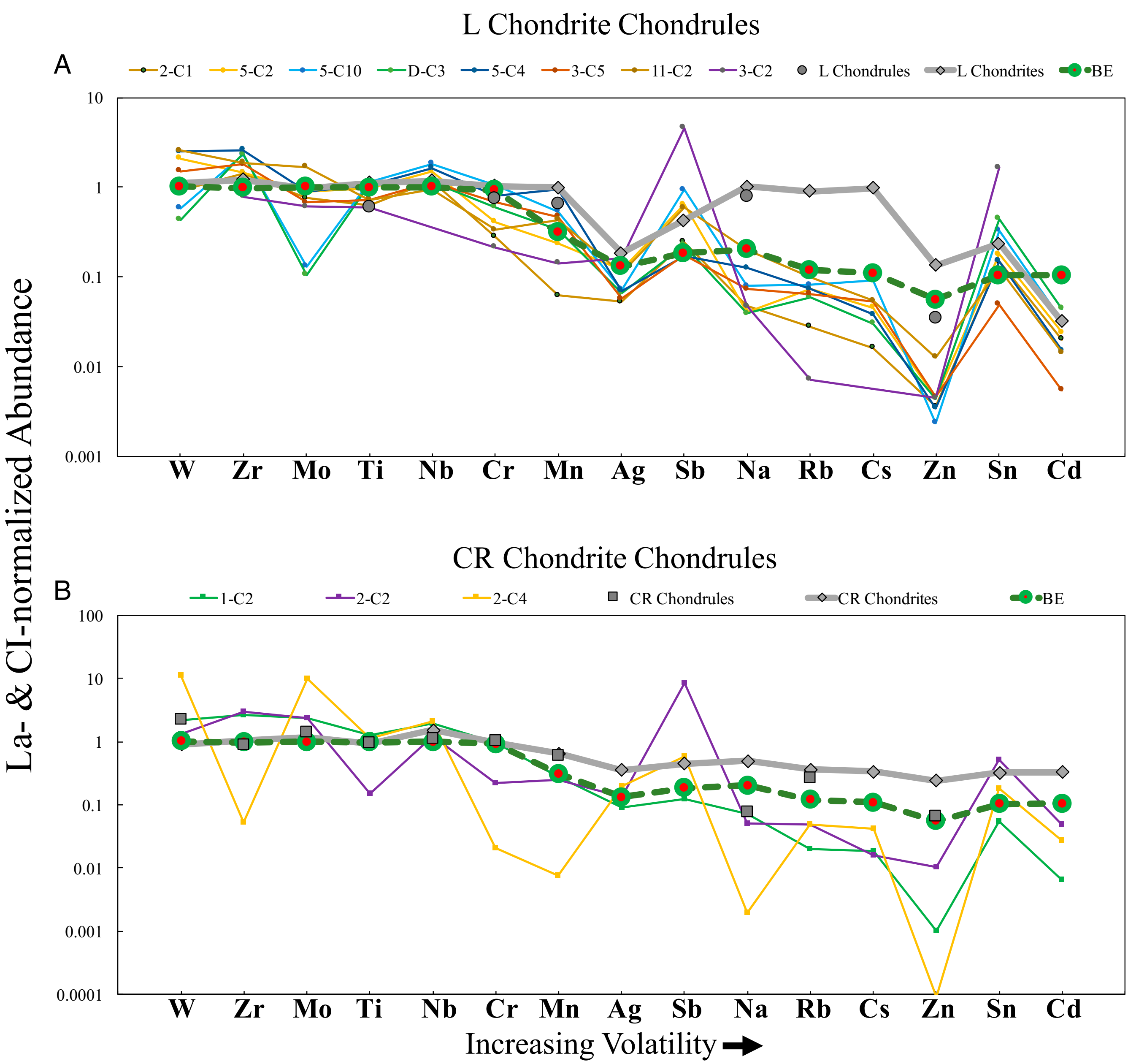

Fig. 1. La- and CI-normalized abundances for refractory, main group, and volatile elements for (*A*) L and (*B*) CR chondrite chondrules. Where available, literature data have been included for both L and CR chondrite chondrules and their bulk meteorite hosts. Bulk Earth estimates have been averaged from refs. 40 and 42. All data are as indicated in the legend.

Manganese-to-Sodium Ratios of Individual Chondrules

The Mn/Na ratios of the L chondrite chondrules increase from ~0.5 to over 3 within approximately the first 1 My of the Solar System, and then decline to nearly the starting value (final Mn/Na ≈ 1) over the next 3 My (Fig. 3). Chondrule olivine fayalite contents (17) for L chondrite chondrules increase concomitant with Mn/Na ratios up to their highest values, further validating Mn/Na as an appropriate first-order gauge of prevailing fO_2 conditions. These results indicate an increase in fO_2—or at least variable fO_2—beginning with the formation of CAIs and peaking at ~4,566.5 Ma, followed by a return to more reducing conditions by ~4,564.5 Ma. This redox path is seemingly at odds with the general enrichment of volatile elements observed (Fig. 2 and Table 2), indicating coeval forces.

Reconciling Volatile Element and Mn/Na Ratio Systematics

The overall redox state of a gas−melt system and the subsequent (re)distribution of volatile elements is not only governed by oxygen fugacity but also by the fugacities of other influential elements that may be present in the gas phase, and it is likely that such mechanisms are at play here. There is ample evidence to indicate that high partial pressures of volatile elements (e.g., K, Na, and S) are behind the simultaneously reduced and volatile-enriched nature of OCs and their chondrules (30, 31). A volatile-rich but water-poor (dry) environment after ~1 My is consistent with the observed general increase in volatile element contents in the younger chondrules and the subsidence of more oxidizing conditions. Dry volatile influx beginning at ~4,566.5 Ma readily explains the enhanced volatile contents of L chondrite chondrules relative to CR chondrite chondrules (Table 2),

Table 2. La- and CI-normalized volatile element abundances for individual chondrules

	NWA 5697 (L3.10)								NWA 6043 (CR2)		
Element	2-C1	5-C2	5-C10	D-C3	5-C4	3-C5	11-C2	3-C2	1-C2	2-C2	2-C4
Mn	0.06	0.23	0.54	0.34	0.95	0.47	0.43	0.14	0.29	0.25	0.01
Ag	0.05	0.12	0.07	0.07	0.07	0.06	0.11	0.16	0.09	0.14	0.20
Sb	0.25	0.64	0.94	0.23	0.17	0.18	0.59	4.59	0.13	8.39	0.58
Na	0.05	0.04	0.08	0.04	0.13	0.07	0.20	0.05	0.07	0.05	0.002
Rb	0.03	0.07	0.08	0.06	0.07	0.06	0.10	0.01	0.02	0.05	0.05
Cs	0.02	0.05	0.09	0.03	0.04	0.05	0.05	0.00	0.02	0.02	0.04
Zn	0.004	0.004	0.002	0.004	0.003	0.005	0.013	0.004	0.001	0.010	0.000
Sn	0.15	0.18	0.33	0.44	0.15	0.05	0.12	1.66	0.05	0.51	0.18
Cd	0.02	0.02	0.03	0.04	0.02	0.01	0.01	0.00	0.01	0.05	0.03

Elements ordered from least to most volatile under solar nebula conditions (22). Please see *SI Appendix* for La- and CI-normalized abundances for all elements (*SI Appendix*, Table S1); La-normalized values and calculated Mn/Na ratios (*SI Appendix*, Table S2); and CI abundances used for normalization, along with literature data for the bulk Earth, L, and CR chondrites and their chondrules (*SI Appendix*, Table S3).

and explains the same observation for their bulk chondrite hosts, as well as the generally water-poor nature of OCs (and enstatite chondrites) relative to carbonaceous ones (5, 13, 23, 32). Moreover, the timing of the volatile element and redox pathway herein is fully consistent with that suggested by astrophysicochemical models, wherein water levels in the inner Solar System increase dramatically within the first ∼200 Ky (and thus fO_2), followed by decline to dry conditions after about 1 My, leading to a relative increase in the fugacities of other volatile species (19, 33). Furthermore, these results indicate a decoupling

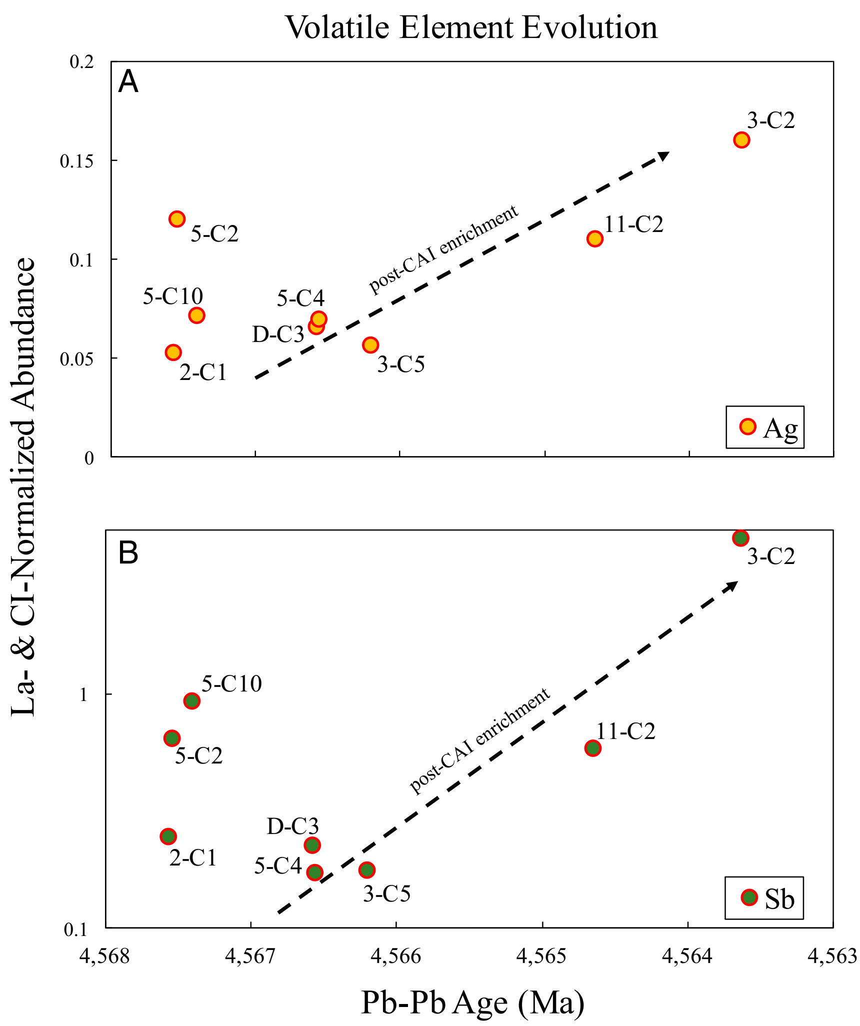

Fig. 2. La- and CI-normalized (*A*) Ag and (*B*) Sb contents for L chondrite chondrules as a function of Pb–Pb age (17). General volatile enrichment through time indicates interaction of chondrules with a gas phase that is increasingly volatile-rich during recycling. Note the logarithmic scale for Sb in *B*.

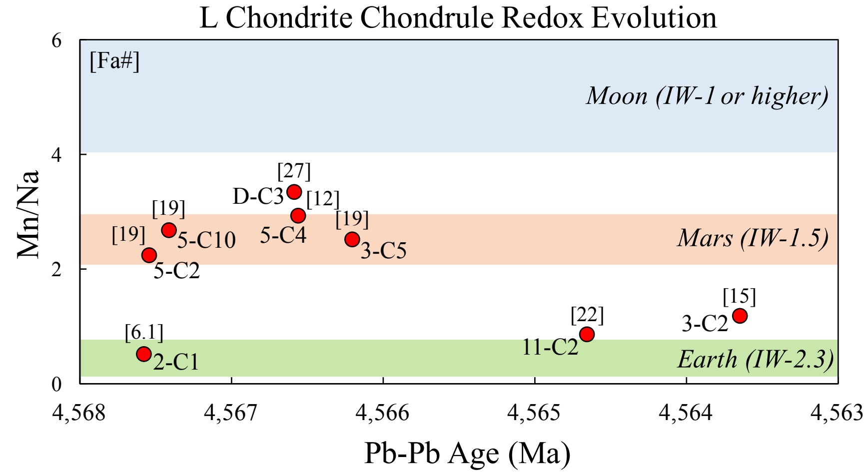

Fig. 3. Calculated Mn/Na ratios for L chondrite chondrules as a function of Pb–Pb age, with individual olivine fayalite number, Fa#, reported in brackets next to individual chondrules (17). Olivine chondrule Fa# is strongly correlated to Mn/Na for the first 1 My of the Solar System ($R^2 = 0.97$), corroborating an increase in oxygen fugacity during this time. After ∼4,566.5 Ma, Mn/Na and Fa# decouple ($R^2 = 0.29$), likely due to incomplete melting of chondrules, gas interaction, and/or variable Fe retention. Typical error for Pb–Pb dating is approximately ±0.50 My, i.e., less than the time spanned here, further validating the observed trends for Mn/Na and Fa#. For reference, Mn/Na ratios for Earth, Mars, and the Moon are reported as green, red, and blue shaded regions, respectively, along with present-day bulk fO_2 estimates (20, 26, 43, 44).

of water and volatile element evolution in the early inner Solar System, calling for a reexamination of the established notion that water and volatile elements arrived in lockstep to the inner Solar System (34).

On the Formation of Larger Solar System Bodies

Vesta and the angrite parent body are thought to have formed no later than 0.4 My after CAIs (35, 36), and the angrite parent body has the highest reported Mn/Na ratio (∼12) of any Solar System body (20), coincident with the timing of high Mn/Na ratios determined in the current study (Fig. 3). Mars, at least half of which is thought to have accreted within the first 2 My of the Solar System (37), has an estimated Mn/Na ratio of 2 to 3 (20), a range strikingly similar to OC chondrules of the same age within the current study, suggesting that a major fraction of Mars could have accreted from OC chondrules (or at least OC-like) resident during this time and thus under similarly oxidizing conditions and increasing volatile contents (Figs. 2 and 3). Proto-Earth is thought to have experienced protracted growth up to ∼5 My after CAIs, with ∼40% of its accreting material potentially coming from volatile-enriched outer Solar System reservoirs (18). This growth history for Earth suggests significant accretion from a reservoir of relatively reduced chondrules, with subsequent volatile content augmentation through time via remelting/recycling in an increasingly volatile-rich environment (Figs. 2 and 3). Moreover, the volatile element and redox pathway determined in the current study indicate a need to explore the possible decoupling of water and other volatiles in petrochemical models of terrestrial planet formation and differentiation. Finally, it is possible that bodies accreting outward of Earth's orbit may have acted as volatile element sinks, based on the relatively elevated moderately volatile element contents of ordinary and enstatite chondrites and their chondrules (13, 38), the high volatile contents (e.g., S) estimated for bulk Mars (38, 39), the formation of these bodies outward of the orbit of Earth (8, 38), and the lack of such enrichments for Earth and that presumed for Venus and Mercury (5, 38, 40).

Conclusions

The compositional evolution of some of the Solar System's first solids, chondrules, has been mapped out as a function of time. These results display a general increase in chondrule volatile contents with age, likely due to chondrule recycling under changing conditions that became more enriched in volatile elements over time. Redox conditions evolved over time, and this is recorded in chondrule Mn/Na ratios. Oxygen fugacity generally increased with time up to ∼4,566.5 Ga, concomitant with volatile enrichments, and afterward displayed a more protracted decline, while volatile contents continued to rise. These observations suggest that this time (∼4,566.5 Ma) marks the onset of inner Solar System dry-out and the subsequent rise in fugacities of other volatiles such as K, Na, and S. These results provide time-anchored evidence for the early formation of proto-Mars from relatively oxidized materials, and evidence that proto-Earth formed via accretion of variably oxidized, volatile-poor chondrules, followed by appreciable addition of recycled, volatile-enhanced material under more reducing conditions. Such an accretion pathway for Earth elicits the need for formation and differentiation models which consider the decoupling of volatile element and water (redox) evolution. Lastly, the timing of formation, relative distance from the Sun, and chemical composition of Mars and the ordinary (and enstatite) chondrites indicate that perhaps these bodies incorporated the bulk of incoming volatile-rich material from cooler outer regions of the Solar System, depriving Earth and other bodies accreting inside its orbit of their full complement of volatile elements.

Materials and Methods

Dilute sample aliquots were analyzed using the high-resolution inductively coupled plasma mass spectrometer Thermo Scientific Element XR at Laboratoire Géosciences Océan (Université de Brest) for the following isotopes: ^{139}La, ^{140}Ce, ^{141}Pr, ^{146}Nd, ^{147}Sm, ^{151}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{167}Er, ^{169}Tm, ^{174}Yb, ^{175}Lu, ^{184}W, ^{91}Zr, ^{98}Mo, ^{47}Ti, ^{93}Nb, ^{53}Cr, ^{55}Mn, ^{107}Ag, ^{121}Sb, ^{23}Na, ^{85}Rb, ^{133}Cs, ^{66}Zn, ^{118}Sn, and ^{114}Cd. All elements were calibrated using standard solutions (10, 50, 100, and 1,000 ppt), and their count rates were blank-corrected. Indium (^{115}In) was used as an internal standard to correct all data for instrumental mass bias during the analyses. Instrument precision (reproducibility) for all elements was better than 4% (1 relative SD). All analyses within the current study were performed on the same sample dissolutions from which aliquots had previously been taken for Pb–Pb dating and Zn isotopic measurements (17), rendering calculations of absolute element concentrations untenable. Therefore, all data have been normalized to La and to CI chondrites. La was chosen as an ideal normalizing element, as its concentrations in both L and CR chondrite chondrules have been previously established and anchor the results (9, 12). Furthermore, La is a

EARTH, ATMOSPHERIC, AND PLANETARY SCIENCES relatively refractory ($T_{50} \approx$ 1,580 K) (22) and fluid-immobile element and thus is not redistributed during secondary alteration processes (41), and it is sufficiently abundant in all samples for precise measurement.

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