Nickel isotopic evidence for late-stage accretion of Mercury-like differentiated planetary embryos

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Earth's habitability is closely tied to its late-stage accretion, during which impactors delivered the majority of life-essential volatiles. However, the nature of these final building blocks remains poorly constrained. Nickel (Ni) can be a useful tracer in characterizing this accretion as most Ni in the bulk silicate Earth (BSE) comes from the late-stage impactors. Here, we apply Ni stable isotope analysis to a large number of meteorites and terrestrial rocks, and find that the BSE has a lighter Ni isotopic composition compared to chondrites. Using first-principles calculations based on density functional theory, we show that core-mantle differentiation cannot produce the observed light Ni isotopic composition of the BSE. Rather, the sub-chondritic Ni isotopic signature was established during Earth's late-stage accretion, probably through the Moon-forming giant impact. We propose that a highly reduced sulfide-rich, Mercury-like body, characterized with light Ni isotopic composition of its mantle, collided with and merged into the proto-Earth during the Moon-forming giant impact, producing the sub-chondritic Ni isotopic signature of the BSE, while delivering sulfur and probably other volatiles to the Earth.

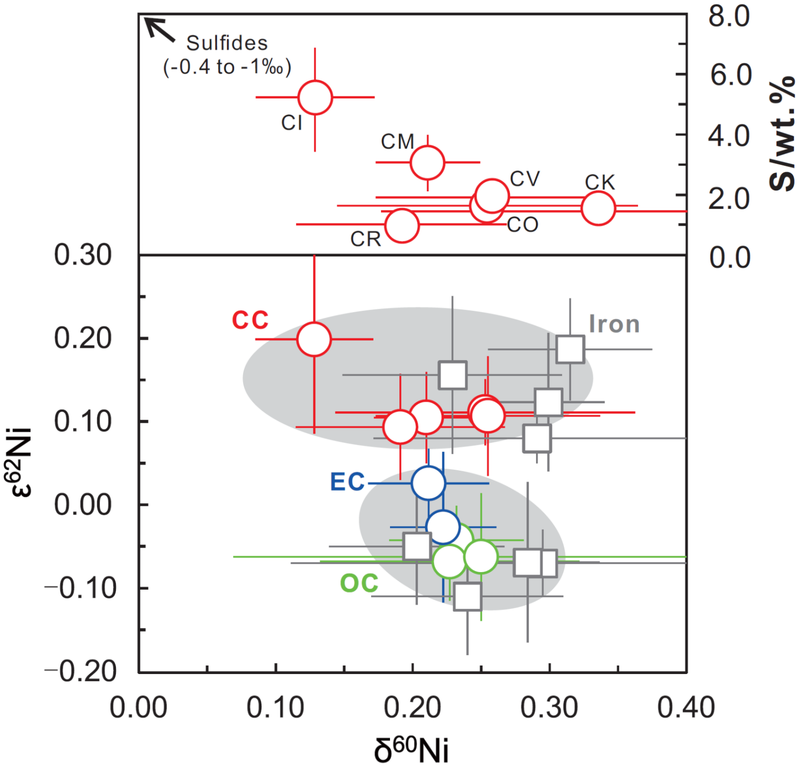
The Earth experienced a protracted accretion history over several tens up to 100 million years, which proceeded by the collision of numerous planetesimals and planetary embryos[1,2]. A fundamental assumption was that the Earth's building blocks as a whole were compositionally similar to undifferentiated chondritic meteorites. Researchers have looked among different classes of chondrites for the closest representative of accreting materials that formed Earth[3-6]. However, emerging evidence points to a mismatch in many crucial elemental and isotopic ratios between chondritic meteorites and the accessible Earth, arguing for the possible accretion of additional materials that are chemically and isotopically different from extant meteorite collections[7-11]. Constraining the nature of these building blocks of Earth is important, because they not only provide fundamental information on terrestrial planet formation, but also help understand how the Earth evolved into its current habitable status.

The late accretion stages, including the Moon-forming giant impact and the late veneer event, likely account for only <10% of Earth's total mass[12], but they represent a critical step for Earth to build its life-essential volatile budgets[13-20]. Dynamical models of Earth's growth suggest that the late accretion stages were highly heterogeneous, consisting of a mixture of materials from two genetically distinct reservoirs in the Solar nebula[20,21]. One end-member may originate from the inner Solar system and contain a reduced, non-carbonaceous component that is probably 'missing' in known meteorites[11,20-24]. The other may be oxidized, carbonaceous chondrite-like material from the outer Solar system[17,19-21,25]. When these materials were added to Earth is still debated[19-27]. The carbonaceous chondrite-like materials are commonly thought to be the source of major volatiles in Earth[28,29]. Recent high pressure-temperature experiments on metal alloy-silicate partitioning of volatiles (*e.g.,* carbon, sulfur and nitrogen), however, suggest that Earth's volatile abundance patterns could have been largely established by impact of a sulfur-rich, differentiated planetary body with minimal contributions from carbonaceous chondrite-like materials[15,16]. Due to the lack of proper meteorite proxies, the nature of late-stage impactors remains poorly known.

Nickel isotopic compositions of meteorites and terrestrial rocks may hold important clues. Nickel in the bulk silicate Earth (BSE) was mostly derived from late-stage impactors, as that from earlier stages was largely removed into the core due to its moderately siderophile nature[23]. Models predict that ~95% of Ni in the BSE derived from the last ~35% of mass that accreted to Earth[23,30]. Nickel is non-volatile and partitioned compatibly into the mantle dominant phase –

olivine – following accretion, such that the BSE can potentially capture the Ni isotopic signature of late-stage accreting materials.

Nickel isotopic variations in meteorites have been well documented, as shown in Fig. 1. Mass-independent nucleosynthetic Ni isotope anomalies arise from the heterogeneous distribution of presolar matter in the Solar protoplanetary disk, and thus trace the provenance of Earth's building blocks. The nucleosynthetic anomalies are present in carbonaceous and ordinary chondrites, with enstatite chondrites largely within error of the BSE[23,31-35], supporting the general idea that the late-stage accreting materials mainly originated from an enstatite-like source region in the inner Solar system[23]. Iron meteorites display similar anomalies, together with different groups of chondrites, forming a dichotomy between carbonaceous and non-carbonaceous meteorites as found in many other isotope systems (*e.g.,* Mo, Cr, Ru, Ti)[36]. Mass-dependent isotopic variations stem from physico-chemical processes in the Solar nebular and on the planetary parent bodies. Nickel isotopic compositions of enstatite, ordinary, and most carbonaceous chondrites exhibit a common value, expressed as $\delta^{60/58}$Ni (the ^{60}Ni/^{58}Ni ratio in parts per thousand, relative to the SRM986 standard; $\delta^{60/58}\text{Ni} = (^{60/58}\text{Ni}_{\text{sample}}/^{60/58}\text{Ni}_{\text{SRM986}} - 1) \times 1000$), with an average of +0.23 ± 0.11‰ (2SD, n = 34)[32,37-40]. The small isotopic variation observed in the carbonaceous chondrites most likely reflects the heterogeneous distribution of an isotopically light sulfide component[41,42], supported by the roughly negative correlation between $\delta^{60/58}$Ni and sulfur content (Fig. 1). It is not surprising that iron meteorites have $\delta^{60/58}$Ni values within the 'chondritic' range (Fig. 1), because they represent fragments of the disrupted cores of planetary bodies, and dominate the Ni budget.


Sulfides
(-0.4 to -1‰)
CI
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CR
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CC
Iron
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OC
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−0.20
0.00
0.10
0.20
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δ60Ni

Fig. 1. A summary of mass-independent and mass-dependent Ni isotopic variations in meteorites. The ε^{62}Ni ($\varepsilon^{62}\text{Ni} = ({}^{62/58}\text{Ni}_{\text{sample}}/{}^{62/58}\text{Ni}_{\text{SRM986}} - 1) \times 10^6$ after internal normalization to ^{61}Ni/^{58}Ni), are from literature[31-33,35,36], and the δ^{60}Ni of chondrites and iron meteorites are from this study and literature[37-41,43]. The carbonaceous chondrites (CC), enstatite chondrites (EC) and ordinary chondrites (OC) have average δ^{60}Ni values of 0.23 ± 0.14‰ (2SD), 0.22 ± 0.02‰ (2SD) and 0.24 ± 0.02‰ (2SD), respectively. A roughly negative correlation between δ^{60}Ni and sulfur abundance is observed in carbonaceous chondrites (upper panel), which may be caused by the presence of various abundances of sulfides with δ^{60}Ni values as low as -1‰[41,42]. The grey areas represent the dichotomy between carbonaceous and non-carbonaceous meteorites[36]. The sulfur abundances are from Ref.[44]. Meteorite data from this study and literature are presented in Supplementary Table S2. Error bars represent 2s.d.

Results and discussion

Non-chondritic Ni isotopic composition of the bulk silicate Earth. The Ni isotopic composition of present BSE is poorly constrained. An earlier report of a few ultramafic rocks yielded $\delta^{60/58}$Ni values indistinguishable from the chondritic average, and they concluded that the BSE has a chondritic Ni isotopic composition[39]. This conclusion is questioned in a recent study combining new and reported peridotite samples[40], which suggests that the BSE has a $\delta^{60/58}$Ni ~0.1‰ lower than the chondrite average, a difference that was attributed to Earth's core formation[40,45]. Central to this debate is the limited Ni isotope data for terrestrial silicate rocks and scant information on Ni isotope fractionation during igneous and core-mantle differentiations.

Our new high-precision, inter-laboratory analyses on 60 terrestrial silicate rocks demonstrate that the present BSE is unambiguously sub-chondritic. Fertile peridotites, whose major element compositions are closest to the Primitive Mantle (*e.g.*, $Mg^{\#} = 89.6 \pm 1.0$; $Al_2O_3 = 3.52 \pm 0.60$ wt.%)[46], have $\delta^{60/58}$Ni values clustering tightly around +0.10 ± 0.07‰ (2SD; n = 13, Fig. 2 and Supplementary Fig. S2). Peridotites overprinted by mantle metasomatism have Ni isotopic compositions shifted towards either heavier or lighter values, but only to a limited degree (Supplementary Section 1). The melting products of mantle have similar or lighter Ni isotopic compositions compared to peridotites (Fig. 2). Komatiites formed by high-degree mantle melting (>45%) record an isotopic signature similar to the fertile peridotites (+0.13 ± 0.09‰, 2SD, n = 15; Fig. 2). Oceanic basalts (OIBs and MORBs), which are produced by relatively low-degree melting (<25%), have Ni isotopic compositions slightly lighter than peridotites (Fig. 2; Student *t*-test; $p < 0.001$), with an average $\delta^{60/58}$Ni value of 0.03 ± 0.16‰ (2SD, n = 15). Eclogites, formed from metamorphism of basalts, display a similar average $\delta^{60/58}$Ni value of 0.02 ± 0.06‰ (2SD, n = 7; Fig. 2). Whether the difference between oceanic basalts and peridotites implies possible Ni isotope fractionation during partial melting or results from the limited dataset of oceanic basalts deserves further investigations. Nevertheless, the present BSE, as best represented by fertile peridotites reported in this study and literature[40], has $\delta^{60/58}$Ni of +0.11 ± 0.06‰, lower than the chondritic average, +0.23 ± 0.11‰ (Student's *t*-test, $p \ll 0.001$; Supplementary Section 1).

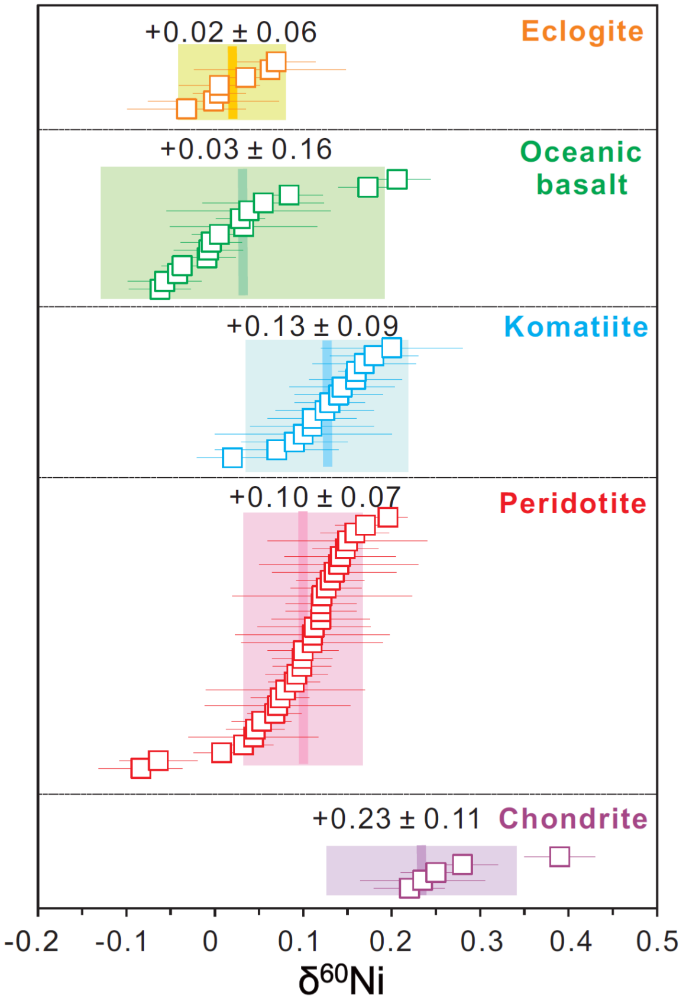

Fig. 2. Mass-dependent Ni isotopic variations of terrestrial silicate rocks and chondrites from this study. The bands represent the average value for each sample category with 2s.d. The $\delta^{60/58}$Ni of five chondrite samples fall within the chondritic average defined by all published data (0.23 ± 0.11‰). The fertile and non-metasomatized peridotites with chemical compositions most close to the BSE[46] have homogeneous Ni isotopic compositions, while those experienced secondary modification have relatively large variations. Therefore, only fertile, non-metasomatized peridotites are used for the average calculation (0.10 ± 0.07‰). Data for chondrites are presented

in Supplementary Table S2; Data for peridotites, komatiites, oceanic basalts and eclogites are presented in Supplementary Table S4-S6. Error bars represent 2s.d.

First-principles calculations on Ni isotope fractionation during core formation. We suggest that the sub-chondritic Ni isotopic composition of the BSE could have resulted from two possible processes: isotope fractionation associated with Earth's differentiation, or the accretion of non-chondritic materials.

The former hypothesis is examined using first-principles calculations on Ni isotope fractionation factors ($10^3\ln\alpha$ of $^{60}Ni/^{58}Ni$) among Earth's major Ni-bearing phases: olivine, wadsleyite, ringwoodite, bridgmanite, and Fe-Ni alloy. Limited differences in $10^3\ln\alpha$ are found between olivine and wadsleyite/ringwoodite in the mantle transition zone and bridgmanite in the lower mantle (*e.g.,* $10^3\ln\alpha < 0.05‰$ at 1500 K and $< 0.03‰$ at 2000 K; Fig. 3a), which excludes the possibility of a hidden reservoir enriched in heavy Ni isotopes in the mantle. This lends credence to the use of accessible mantle and mantle-derived samples as representative of the present BSE Ni isotopic signature. Nickel isotope fractionation between Fe-Ni alloy and silicate (*e.g.,* bridgmanite) under core-formation conditions is also negligible (P = 25-130 GPa; Fig. 3b and Supplementary Section 2). Notably, incorporation of sulfur into the Fe-Ni alloy slightly reduces the force constant of Ni, leading to the enrichment of light Ni isotopes in Fe-Ni alloys relative to the silicates (Fig. 3b).

To directly assess the equilibrium Ni isotope fractionation between silicate and metallic melts during core-mantle differentiation, we performed first-principles molecular dynamic simulations on melt phases of $Fe_{92}Ni_5S_3$ and $Mg_{30}NiSi_{32}O_{96}$ based on the density functional theory. The $10^3\ln\alpha$ between $Fe_{92}Ni_5S_3$ and $Mg_{30}NiSi_{32}O_{96}$ melts is -0.011‰ at ~38 GPa and 3500 K (Fig. 3b; Supplementary Section 2), further confirming the conclusion based on crystals that core-mantle differentiation does not significantly fractionate Ni isotopes.

Two published experimental studies investigated equilibrium Ni isotope fractionation between metal and silicate mineral or melt at lower pressure (< 1.3 GPa) and temperature (< 1623 K)[47,48]. Both studies predict limited Ni isotope fractionation under core-formation conditions (< 0.01‰ in terms of $\delta^{60/58}$Ni at T > 3000 K), a result that is consistent with our first-principles calculations at higher pressures, implying negligible pressure effect on silicate-metal Ni isotope fractionation. If the bulk Earth (BE) has a chondritic Ni isotopic composition, mass balance calculations using high-pressure metal-silicate Ni elemental partition coefficients[49] and isotope fractionation factors obtained from our first-principles calculation demonstrate that core-mantle differentiation cannot explain the sub-chondritic Ni isotopic composition of the BSE (Fig. 3c).

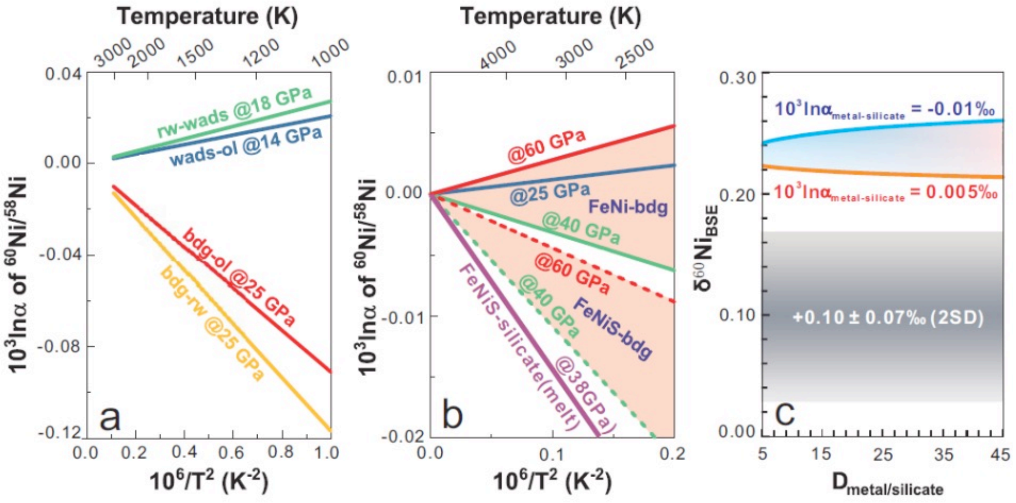

Fig. 3. a, Equilibrium fractionation factors (10^3ln□) between olivine, wadsleyite, ringwoodite, and bridgmanite. **b**, Equilibrium fractionation factors between Fe-Ni (S) alloy and bridgmanite, and between FeNiS melt ($Fe_{92}Ni_5S_3$) and silicate melt ($Mg_{30}NiSi_{32}O_{96}$). **c**, $\delta^{60}Ni_{BSE}$ vs. distribution coefficient of Ni between metal and silicate ($D^{Ni}_{metal\text{-}silicate}$). If the bulk Earth (BE) has a chondritic Ni isotopic composition, the BSE δ^{60}Ni after core formation can be evaluated using the Rayleigh fractionation equation: $\delta^{60}Ni_{BSE} - \delta^{60}Ni_{BE} = \Delta^{60}Ni_{metal\text{-}silicate} \times \ln f_{BSE}$, where f_{BSE} is the fraction of Ni in the BSE. The f_{BSE} can be calculated using the mass balance model: $f_{BSE} = M_{mantle}/(M_{mantle} + D \times M_{core})$. The masses of the mantle and the core are 0.675 and 0.325, respectively. Given that the $D^{Ni}_{metal\text{-}silicate}$ is likely < 45[49], core segregation induced Ni isotope fractionation between metal and silicate cannot account for the light Ni isotopic composition of the BSE. ol, olivine; wads, wadsleyite; rw, ringwoodite; bdg, bridgmanite; FeNi, Fe-Ni alloy; FeNiS, S bearing Fe-Ni alloy or melt.

Other possible events including evaporative loss, collision erosion and core-mantle chemical diffusion can be discounted as causes for the sub-chondritic Ni isotopic signature of BSE[40]. Evaporative loss of Ni is unlikely given the relatively refractory nature of Ni. In addition, kinetic isotope fractionation associated with evaporation would lead to a heavy BSE Ni isotopic composition, opposite to observations (Fig. 2). Collisional erosion during Earth's formation preferentially removed early-formed basaltic crust[50]. The terrestrial oceanic basalts have an average $\delta^{60/58}$Ni (0.03 ± 0.16‰; 2SD) slightly lower than the BSE value, and thus collisional erosion cannot explain the sub-chondritic Ni isotopic composition of BSE. The presence of a Ni chemical gradient between Earth's core and mantle may induce diffusive isotope fractionation, due to the faster diffusivity of light isotopes relative to heavy ones. A one-dimensional diffusion model shows that core-mantle chemical diffusion produces ~0.1‰ variation in the silicate part but is restricted to the lowermost two kilometers of the mantle on a time scale of 10 million years (Supplementary Section 3).

The nature of late-stage accreting materials and its implications

Therefore, the Earth's mantle must have accreted sub-chondritic materials during its growth. In the early stages, metal and silicate melts equilibrate completely in the magma ocean[51], so that the proto-BSE likely has a low Ni concentration and a chondritic Ni isotopic composition. Because of the moderately siderophile nature of Ni[23] and a possible disequilibrium scenario for the late-stage accretion[51,52], the BSE's sub-chondritic Ni isotopic signature was likely established during the late stages of Earth's accretion. The late veneer following the main growth stage added the last <0.5% of mass to Earth and contributed <5% of the Ni budget of the BSE[18]. Hence, it is unlikely to be the event that produced the sub-chondritic Ni isotopic composition of BSE. To account for the observed Ni isotopic value of the present BSE, the late-veneer material accreted to the BSE would have to have had extremely low $\delta^{60/58}$Ni of around -2.5‰ (Supplementary Fig. S8), a value that has not been found in any natural rocks, and would be inconsistent with the Ni isotopic composition of an average carbonaceous chondrite-like material for the late veneer[17,19,24,25]. The last significant stage of Earth's accretion was the Moon-forming giant impact, contributing >20% of Ni budget of the BSE[51]. The Ni isotopic composition of the BSE could have been strongly influenced by the Moon-forming impactor[18,23]. Assuming a chondritic Ni isotopic composition for the proto-BSE as discussed above, mass balance calculation suggests that materials accreted to the proto-BSE have $\delta^{60/58}$Ni values as low as -0.35‰ (Supplementary Fig. S8). Accordingly, the Moon-forming impactor is unlikely to have a composition represented by chondrites.

Instead, we hypothesize that the sub-chondritic Ni isotopic composition of the BSE resulted from the impact and accretion of the sulfide-rich mantle of a highly reduced, differentiated planetary body. It has long been recognized that accretion of planetary embryos that were already differentiated into cores and mantles contributed significantly to the growth of Earth[53-55]. The Moon-forming impactor has been suggested to be a sulfur-rich, differentiated planetary body[15,16,27]; but uncertainties remain as to whether it is a highly reduced, Mercury-like impactor[23,24,27] , or a relatively more oxidized body[20,21,26]. Mercury is the most reduced planet in the inner Solar system and has an abnormally high abundance of sulfides in its mantle[56-58], whereas oxidized planetary embryos have the sulfur dominantly in their cores[59]. This is because sulfur is highly siderophile at high oxygen fugacity (fO_2) and partitions into the metallic core, but becomes lithophile and enters into the silicate melt as sulfide species under low fO_2 (*e.g.,* five units below the iron-wüstite buffer; IW-5)[60,61]. Magmatic sulfides are the only major Ni-bearing phases that are isotopically much lighter than silicates ($\delta^{60/58}\mathrm{Ni}_{\mathrm{sulfide}}$ down to -1‰)[41]. Rocks with high sulfide/silicate ratios have light Ni isotopic compositions, which is most evident in magmatic Ni-sulfide deposits where the bulk $\delta^{60/58}$Ni values are negatively correlated with the sulfur content[41,42]. Therefore, when small planetary embryos (the proto-impactor) were formed in a sulfur-rich early Solar nebular and differentiated into core and mantle under highly reduced environment analogous to the Mercury (mean IW-5.4)[60], the mantle would be sulfur-rich and have a light Ni isotopic composition (Fig. 4). By contrast, in the large proto-Earth, core-mantle differentiation proceeded under much higher pressure and likely more oxidizing condition (>IW-3; Fig. 4)[62], in which sulfur behaves as siderophile[63,64], leading to a sulfur-poor mantle. In this case, limited silicate-metal Ni isotope fractionation is expected (see discussion above), and thus the proto-Earth mantle likely has a chondritic Ni isotopic composition. During the Moon-forming impact, the impactor's core merged directly into the proto-Earth's core due to its limited emulsification, while the remaining parts of the impactor were incorporated into the Earth's mantle[51,52]. The Mercury-like impactor's sulfur-rich mantle would have been completely dissolved in the planet-wide, more oxidizing terrestrial magma ocean[65], and produced the sub-chondritic Ni isotopic signature of Earth's mantle (Fig. 4).

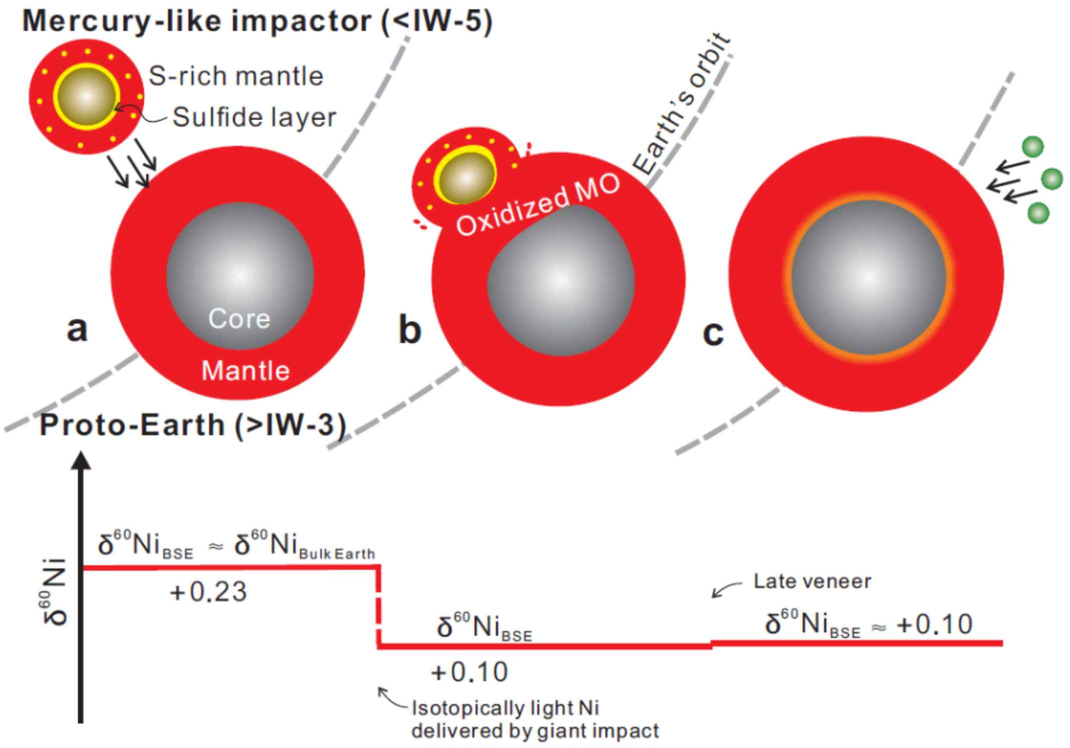

Fig. 4. Cartoon showing the merger of a highly reduced, Mercury-like planetary body with the relatively more oxidizing proto-Earth, and schematic evolution of Ni isotopic composition of the BSE. **a**, The small, proto-impactor from the highly reduced innermost region of the Solar system differentiated into a core, a sulfur rich mantle, and likely a sulfide layer at the mantle-core boundary, because sulfur behaves more lithophile at low fO_2 and pressure[60,61,63,64]. The impactor's mantle likely had a sub-chondritic Ni isotopic composition due to the enrichment of sulfides. Differentiation on the large, proto-Earth partitioned sulfur dominantly into the core because sulfur is more siderophile at relatively high fO_2 and pressure[60,61,63,64]. Therefore, the mantle of the proto-Earth had a Ni isotopic composition close to the chondritic value. **b**, The Moon-forming giant impact would have completely melted the Earth, forming a planet-wide, more oxidizing magma ocean (MO) with high solubility of sulfur[62]. The sulfides in the impactor's mantle were dissolved in the terrestrial magma ocean, producing a sub-chondritic Ni

isotopic composition for the Earth's mantle. **c,** The late veneer following the main growth stage added carbonaceous chondrite-like materials to the Earth with limited effects on the Ni isotopic systematics of the BSE as discussed in the text.

Our findings imply that, instead of an outer Solar system origin[20,21], the Moon-forming impactor may represent a 'missing' Earth's building component originated from a highly reduced reservoir in the inner Solar system. While the absence of nucleosynthetic Ni isotopic anomalies in enstatite chondrites is consistent with an inner Solar system provenance for the Moon-forming impactor[23], the sub-chondritic mass-dependent Ni isotopic composition of the BSE further points towards a sulfide-rich, Mercury-like impactor, likely from closer to the Sun. This is in accordance with variations in nucleosynthetic isotope anomalies of other elements in meteorites, suggesting that the Earth possesses the most *s*-process enriched materials from the inner Solar system[11,22,24,66]. A most recent study found *s*-process enriched ruthenium (Ru) isotopic signatures in Eoarchean rocks, supporting the idea that the pre-late veneer Earth incorporated building materials from the innermost region of the Solar system, most likely through the Moon-forming giant impact[24]. The impactor might be sulfur-rich, such that the highly siderophile Ru was partially retained in the mantle without being completely extracted to the core. Later addition of a carbonaceous chondrite-like late veneer with *s*-process Ru deficits from the outer Solar system ultimately built up the modern mantle Ru isotopic composition[24]. The proposed late-stage accretion of the highly reduced, Mercury-like planetary body may not only explain the broad geochemical similarity between Earth and Moon[27], but also account for the volatile abundance patterns in the BSE[16]. Our study highlights the importance of inner planets, *e.g.*, Mercury and

Venus, in searching for the Earth's 'missing' building blocks that are not present in extant meteorite collections. Future studies on achondrites from the inner Solar system and samples from Venus and Mercury as well as experimental work will shed more light on these issues.

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Supplementary Materials:

Materials and Methods

Figures S1-S8

Tables S1-S9

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