

# Cosmic dust fertilization of glacial prebiotic chemistry on early Earth

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Earth's surface is deficient in available forms of many elements considered limiting for prebiotic chemistry. In contrast, many extraterrestrial rocky objects are rich in these same elements. Limiting prebiotic ingredients may, therefore, have been delivered by exogenous material; however, the mechanisms by which exogeneous material may be reliably and non-destructively supplied to a planetary surface remains unclear. Today, the flux of extraterrestrial matter to Earth is dominated by fine-grained cosmic dust. Although this material is rarely discussed in a prebiotic context due to its delivery over a large surface area, concentrated cosmic dust deposits are known to form on Earth today due to the action of sedimentary processes. Here we combine empirical constraints on dust sedimentation with dynamical simulations of dust formation and planetary accretion to show that localized sedimentary deposits of cosmic dust could have accumulated in arid environments on early Earth, in particular glacial settings that today produce cryoconite sediments. Our results challenge the widely held assumption that cosmic dust is incapable of fertilizing prebiotic chemistry. Cosmic dust deposits may have plausibly formed on early Earth and acted to fertilize prebiotic chemistry.

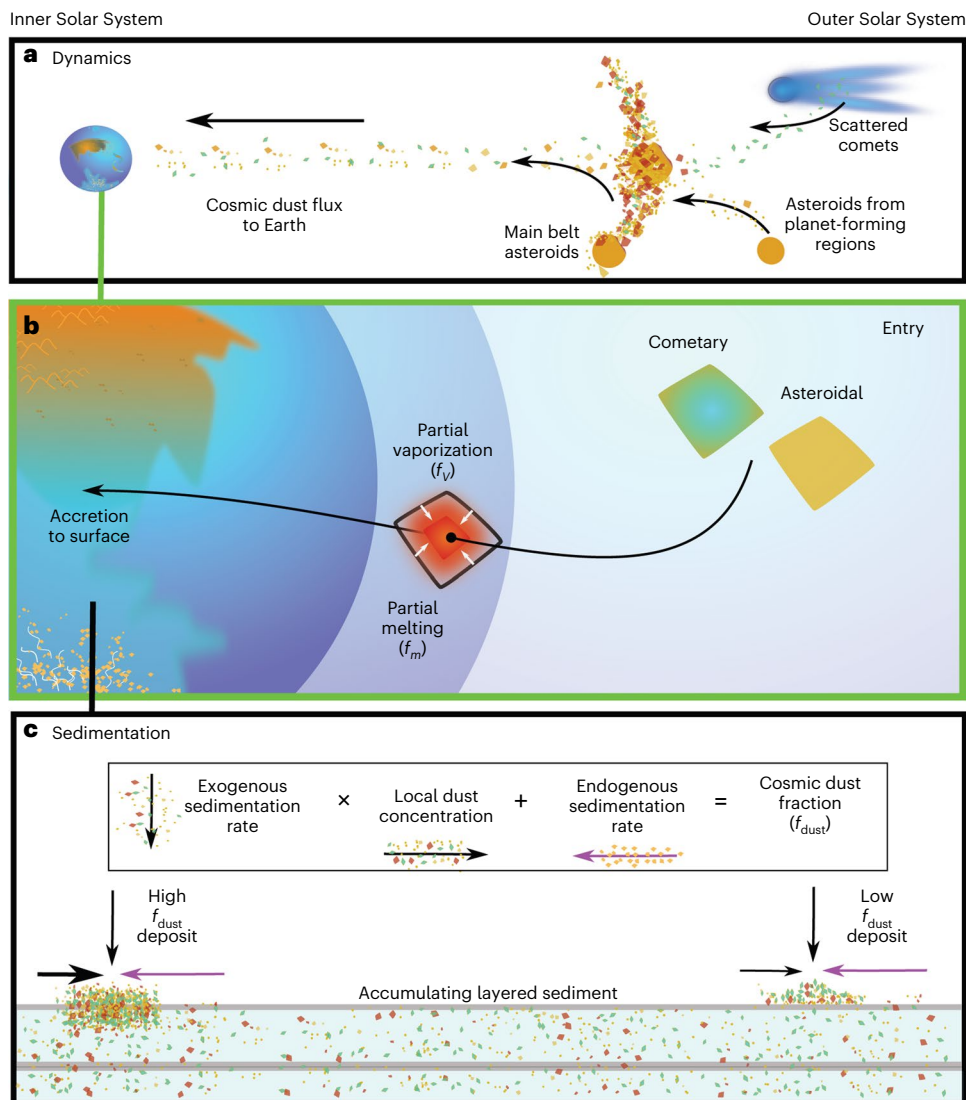
The origin of life on Earth probably resulted from interacting solid, liquid and gaseous reservoirs of bioessential elements in reactive molecular forms<sup>1</sup>. Experimental work demonstrates that high to moderate concentrations (mM to hundreds of mM) of simple species (for example, HCN, PO<sub>4</sub><sup>3-</sup> and HSO<sub>3</sub><sup>-</sup>) can produce high yields of biologically relevant molecules, such as nucleic acids, lipids and peptides<sup>2-6</sup>. However, a remaining gap in our understanding of the geological context of prebiotic chemistry on Earth is the mechanisms by which concentrated feedstocks were produced<sup>7</sup>.

Common terrestrial rocks are relatively poor in reactive and soluble forms of the key elements mentioned above: phosphorus (P), sulfur (S), nitrogen (N) and carbon (C). Indeed, life on Earth is engaged in fierce competition for the limited, endogenous, bioavailable reservoirs of these elements. Complex enzymatic machinery has evolved in response

to this challenge, such that life can extract these species from the environment even when they occur in limited concentration or in largely inert chemical form. The pre-enzymatic world of prebiotic chemistry must have initially lacked such mechanisms to enhance the availability of key species. However, certain processes in Earth's early history may have gone part or all of the way towards solving this apparent paradox. One such possibility is the accretion and surficial sedimentary sorting of cosmic dust (here defined as grains of size <3 mm).

Cosmic dust comprises mineral grain aggregates produced by collisions between asteroids<sup>8</sup> and the sublimation and disintegration of comets<sup>9</sup> (Fig. 1a). Such particles produced further from the Sun can then drift inwards due to Poynting–Robertson drag and be accreted by Earth. Cosmic dust contains bioessential elements (for terrestrial life), for example, P, S, N and C, at concentrations well above that of

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**Fig. 1 | Delivery dynamics, atmospheric entry and terrestrial sedimentation of cosmic dust.** **a–c**, Schematic illustrations of factors considered in this study related to the formation of terrestrial sediments rich in cosmic dust. **a**, Dynamical sources of cosmic dust grains. Comets (undifferentiated) scattered inwards from the outer Solar System disintegrate to produce dust particles. Asteroids predominantly generate dust through collisions. **b**, Atmospheric entry of cosmic dust involves partial melting ( $f_m$ ) and partial vaporization ( $f_v$ ), both of which

influence the final mass of cosmic dust per unit mass of total sediment in a given sedimentary environment ( $f_{\text{dust}}$ ,  $\text{kg kg}^{-1}$ ). **c**, The relative abundance of cosmic dust within terrestrial sediments is set by the local sedimentation rates of cosmic dust versus terrestrial (endogenous) sediment, with dust proportion, therefore, being maximized in areas of low endogenous sediment production due to the action of local sedimentary concentration mechanisms.

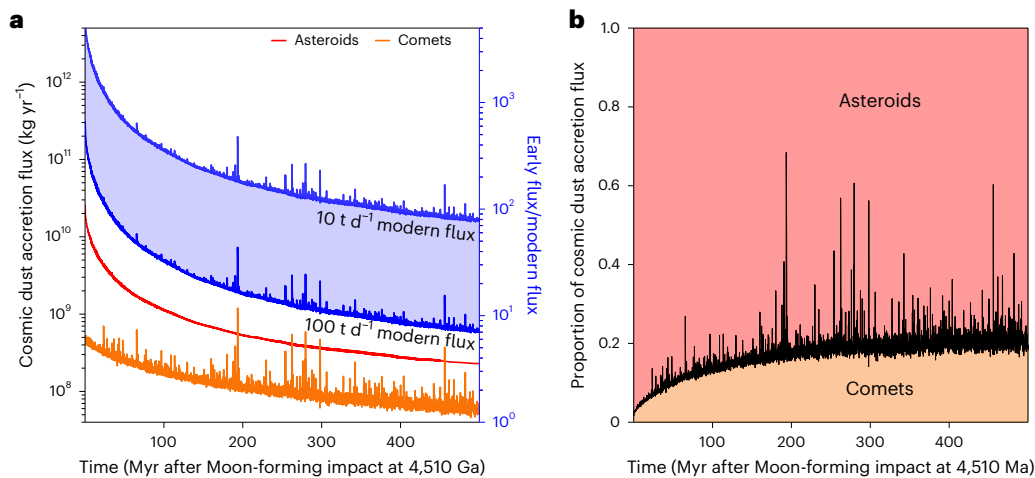
Earth's crust<sup>10–12</sup>. Many cosmic dust grains represent nearly pristine samples of their parent body objects, which appear to span comets (volatile-rich outer Solar System materials) and asteroids (a mixture of early-formed differentiated objects and relatively late-formed and small-to-moderate-sized undifferentiated objects, of both outer and inner Solar System origin)<sup>11,13</sup>.

In contrast to larger objects, the flux of cosmic dust to Earth is essentially constant on yearly timescales. Moreover, some fraction of cosmic dust grains pass relatively gently through the Earth's atmosphere, thereby retaining a greater fraction of primitive CHNS than do large (for example, bolide-type) impactors<sup>14–17</sup>. The continuous and mostly non-destructive accretion of cosmic dust across planetary surfaces suggests an advantage over discrete and violent episodes of bioessential element delivery by larger impactors, potentially greatly improving the chances of the successful entry of extraterrestrial matter into prebiotic chemistry.

Cosmic dust is accreted across the Earth's entire surface area, such that it is initially dilute in terms of mass per unit area relative to discrete

delivery events by larger objects<sup>18</sup>. On this basis, the prebiotic importance of cosmic dust has previously been questioned<sup>18,19</sup>. However, there are many planetary processes that can concentrate fine-grained materials from across large surface areas to form concentrated deposits, for example, aeolian, fluvial and glaciogenic sorting mechanisms, which produce dunes, beaches and moraines, respectively. Indeed, these mechanisms operate today and locally concentrate cosmic dust by up to 1,000-fold relative to the global baseline; on the basis of which, cosmic dust has been proposed to be relevant to the origin of life<sup>20–25</sup>. Despite these possible links, there are no existing quantitative models of the supply to and surface cycling of cosmic dust on early Earth.

Here, we use astrophysical simulations and geological models to quantify both the flux and compositions of cosmic dust accreted to the surface of early Earth (Fig. 1a). We combine these results with geological models of subaerial concentration in a range of environments (Fig. 1b,c). Using our results, we assess the possibility that cosmic dust deposits on early Earth fertilized prebiotic chemistry.



**Fig. 2 | Accretion flux of interplanetary dust particles to early Earth. a**, Total flux and individual fluxes of different cosmic dust populations, divided by parent body type. **b**, Proportions of the total accretion flux of cosmic dust represented

by each population over time. Boundaries between the proportion of each dust source are demarcated with solid black lines.

## Results

### Estimating early dust accretion fluxes

We simulate cosmic dust accretion to Earth during the first 500 million years after the Moon-forming impact at 4.51 Ga (ref. 26). Numerical models (Methods) were used to simulate the generation, interplanetary transport and terrestrial accretion flux of dust produced by two compositionally distinct parent body populations: Jupiter-family comets (JFCs) and asteroids (Fig. 1). Asteroidal dust is considered as the sum of dust generated by main belt asteroids and unstable planetesimals located in planet-forming regions (Methods).

Comparing the results of our numerical model to estimates of the present-day accretion flux of cosmic dust to Earth, we find that the total early accretion fluxes of cosmic dust would have been of the order of 100 to 10,000-fold higher than observed today, depending on the value chosen to represent the modern flux (Figs. 2 and 3)<sup>27</sup>. These elevated dust fluxes arise mainly from (1) a more massive main asteroid belt leading to higher rates of collisional grinding<sup>28</sup>, (2) dynamical perturbation of the asteroid belt and injection of cometary objects by giant planet migration<sup>9</sup> and (3) the collisional erosion of the population of rocky objects left over from planet formation, which were not yet depleted.

Our simulations suggest that early cosmic dust fluxes would have been dominated by fragments of the dynamically unstable asteroid population and comets, with a relatively minor contribution from the main belt (Supplementary Fig. 1). Short-lived spikes in the proportion of cometary material in the cosmic dust accretion flux occur as a result of the scattering of large or long-lived comets, with these spikes lasting for of the order of one to several million years. The timing of these cometary dust spikes has no special relevance in the models, simply reflecting a particular realization of a semi-random history. Critically, this means that intervals of high-flux comet-dominated dust supply could have occurred at any point during the first 500 Myr of Earth history.

Grain-size frequency distributions for the mean mass accretion flux of all three cosmic dust sources are bimodal between 1 and 3,000  $\mu\text{m}$ , peaking at around 1–10 and 100–500  $\mu\text{m}$  (Supplementary Fig. 2). These distributions are ultimately very much like those arriving at Earth's atmosphere and observed in cosmic dust deposits today<sup>10,13,29</sup>, with a somewhat larger contribution by particles larger than 500  $\mu\text{m}$ . Overall, our model predicts an early cosmic dust flux that differs mainly in having a higher total mass per unit time and distinct compositional make-up compared with that observed today. However, like today<sup>30</sup>, and with great relevance for prebiotic chemistry,

volatile-rich dust grains—including >65% cometary material at some points—are expected to have dominated the cosmic dust flux to early Earth (Fig. 2b).

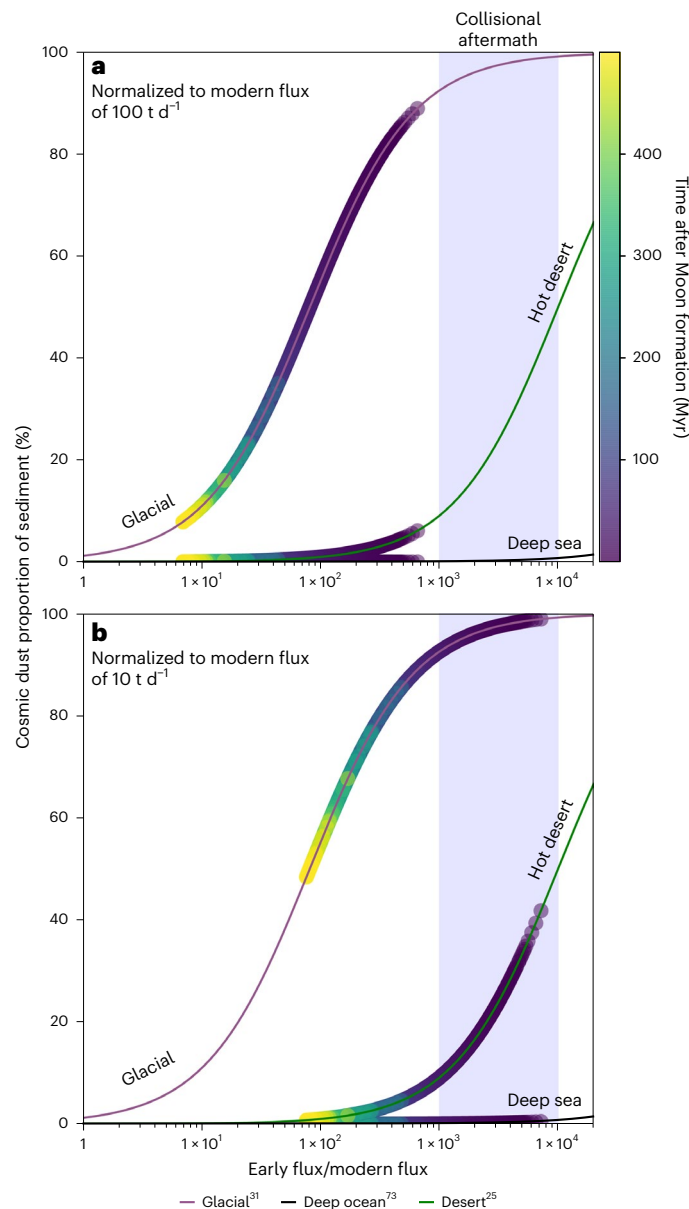
### Occurrence and composition of cosmic dust deposits

The final concentration of cosmic dust in a sedimentary deposit will depend on dust input rate, endogenous sediment supply and local dust concentration mechanisms. Using our estimates of dust accretion fluxes to prebiotic Earth, we construct a model to predict the proportion of dust within coeval unconsolidated sediments (Methods and Fig. 1c). We consider a range of end-member sedimentary environments: glacier surfaces, hot deserts and deep-sea sediments. The proportion of cosmic dust by mass, the relative proportions of types of cosmic dust, and their chemical compositions and degrees of alteration have been measured for all these environments on Earth today, allowing us to ground truth our calculations (Supplementary Table 1). We use the resulting empirically derived scaling factors to estimate both the total cosmic dust concentration by mass and the relative abundance of cosmic dust types within sediments on early Earth as a function of early dust flux relative to modern values (Fig. 3).

Many aspects of early Earth remain uncertain. To obtain meaningful results, we assume that all early Earth environmental parameters are equivalent to present-day values, except the rate of cosmic dust accretion. This minimizes sources of uncertainty that would otherwise be difficult or impossible to constrain, allowing us to test the prebiotic relevance of dust supply in one well-constrained early Earth scenario.

We further assume that the concentration mechanisms operating in these environments will operate with the same efficiency at all accreted mass fluxes of dust considered in our work. That is, sedimentary environments are far from being dust saturated. We focus on environments where high cosmic dust concentrations are achieved without a high degree of fractionation with respect to different categories of dust. This allows us to assume that differences in the compositional make-up of the accreted cosmic dust translate directly into the resulting estimated compositions of putative early Earth cosmic dust deposits<sup>13,31</sup>. Furthermore, although the presence of vegetation in modern-day environments strongly affects sedimentation mechanisms and although early Earth would have lacked such interferences, the environments modelled in our study are essentially absent of any vegetation, such that our all-else-being-equal approach is applicable.

To interpret our results, we must normalize our values for the early Earth accretion flux of cosmic dust to a reference value for the modern



**Fig. 3 | Predicted proportion by mass of cosmic dust within sediments, which varies with the ratio of the early to modern accreted dust flux.**

**a, b**, Cosmic dust proportion with a conservatively high reference value for modern dust accretion (**a**) and an optimistically low reference value (**b**). The predicted proportion of dust within terrestrial sediments varies as a function of local environmental processes and conditions that preferentially concentrate dust relative to terrestrial sediment<sup>25,31,73</sup>. Dust can be expected to represent a higher concentration of overall sediment in environments with low generation rates of local endogenous sediment and where dust concentration mechanisms are active, for example, sediment traps on glaciers, versus for example, deep sea sediments, where endogenous sedimentation rates are high and concentration mechanisms are ineffective (Fig. 1c). Individual data points, coloured according to their associated simulation time step, indicate the average sediment compositions predicted by our simulations. The shaded regions indicate the broad range of possible elevated dust fluxes that may transiently occur following individual collisions of large parent bodies, which would translate into extremely cosmic-dust-rich sedimentary compositions in the context of our model.

top-of-atmosphere dust accretion flux. We compare our results to a range of conservative values for the top-of-modern-atmosphere dust accretion flux: a high value of  $100 \text{ t d}^{-1}$  and a low value of  $10 \text{ t d}^{-1}$  (ref. 27). Given that our model for predicting the proportion of cosmic dust in

early sediments is pegged to present-day systems, using lower values to represent the modern dust flux renders our predicted early dust flux relatively higher and, hence, predicts higher cosmic dust proportions in early sedimentary systems (Fig. 3).

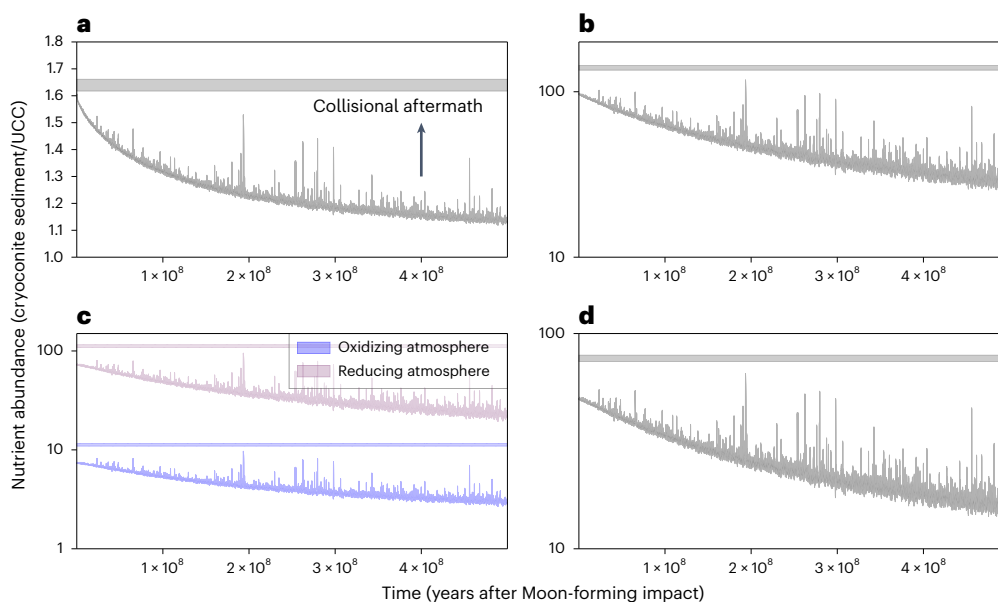
We find that cosmic dust represents a minor component of deep sea sediments, even at the highest dust accretion rates estimated by our model (Fig. 3). Moreover, cosmic dust may have represented >50% of sediments in desert and glacial settings given the same cosmic dust fluxes. The highest concentrations (>80%) would have occurred within glacial ablation zones in cryoconite-type sediments (Fig. 3), just as these sediments contain the highest reported cosmic dust concentrations today<sup>20</sup>. Cryoconite forms on Earth today in small (metre to centimetre) meltwater pools on the surface of retreating ice sheets, with dust components non-destructively introduced by wind-driven transport<sup>32</sup>. Compositionally, cosmic-dust-rich analogues of these sedimentary environments would represent a unique and compelling environment for fostering prebiotic chemistry.

We can gain an insight into this plausible prebiotic utility by exploring the probable chemical compositions of putative cosmic sediments. This is achieved by combing estimates of the contributions of cometary versus asteroidal dust with estimates of their respective average C, N, P and S concentrations (Supplementary Table 1). Although other arid environments are plausible settings for concentrated deposits of cosmic dust, for example, hot deserts (Fig. 3), we focus here on glaciogenic cosmic sediments, the environment we expect to be richest in cosmic dust for any given dust accretion flux (Figs. 3 and 4). Moreover, glacial environments are unusual in being generally arid yet always having the capacity to generate liquid water, unlike hot arid environments. Our results show that the continuous accretion of cosmic dust and its concentration in glaciogenic settings could have generated sediments enriched in bioessential elements relative to the average upper continental crust<sup>33</sup> by up to 100-fold.

These enrichments peak for S, N and C during episodes of enhanced cometary delivery. The highest-amplitude comet-delivery spikes that our model predicts result in around twofold higher concentrations of N and C in the cryoconite sediments than achieved during background cosmic dust accretion. The highest possible enrichments may be obtained in the time frame immediately following the collisional break-up of parent body objects. These episodes of enhanced delivery (million-year timescales) may outpace background cosmic dust fluxes by around 1 order of magnitude<sup>34</sup> (Figs. 3 and 4), yielding equivalently enriched sediment compositions. Given the rarity of parent body break-up events over the last 4 billion years, ground truth constraints on the impact of post-break-up dust fluxes on the concomitant proportions by mass of cosmic dust in terrestrial sediments are hard to establish. However, our estimate of peak fluxes of around  $\times 10,000$  that of modern fluxes and lasting of the order of millions of years is supported by observations of cosmic dust, micrometeorite and fossil meteorite abundance in Ordovician sediments<sup>34,35</sup>, which record the aftermath of the L chondrite parent body disruption event at  $474 \pm 22 \text{ Ma}$  (ref. 36).

Our results highlight that cosmic-dust-rich deposits would have formed most readily in environments of low endogenous sediment production on early Earth over much of its early history, with exceptionally high cosmic dust concentrations developing in the aftermath of individual parent body break-up events. In particular, we find that cosmic dust proportions would have been high in glacial settings, specifically, cryoconite fields within glacial ablation zones (Figs. 3 and 4). Here, cosmic sediments (>50% cosmic dust by mass fraction) would have formed, enriched with respect to average crustal rocks both in terms of the fraction of bioessential elements in reduced form and the overall concentration of those elements. This outcome is robust, given that early sources of cometary material were chemically comparable to the comets sampled by recent missions<sup>37</sup> and unmelted carbonaceous micrometeorites<sup>13,38</sup> (Supplementary Table 1).





**Fig. 4 | Predicted chemical composition and utility for prebiotic chemistry of dust-rich sediments on early Earth.** a–d. Results are plotted for the absolute concentration of each element relative to the observed average concentration of emergent upper continental crust (UCC) on Earth, today<sup>33</sup>: phosphorus (a),

sulfur (b), nitrogen (c) and carbon (d). The results were obtained using a modern-day dust flux of  $10 \text{ t d}^{-1}$  (ref. 27). In c, both oxidizing and reducing atmosphere scenarios are shown, due to the strong consequences for the survival of N-bearing organic molecules during entry under reducing conditions.

## Discussion

In principle, cosmic-dust-rich cryoconite is a compelling scenario for prebiotic chemistry (Fig. 5). However, there are also caveats that should be highlighted. A possible challenge for the prebiotic relevance of our scenario is that many ice sheet surfaces undergoing melting are semipermeable and connected to a wider supraglacial drainage system<sup>39</sup> (Fig. 5). Cryoconite most often forms in the ablation zone of glaciers, where ice dynamics or the continual development of the supraglacial drainage system typically preclude permanent or even multi-annual cryoconite holes. As such, prebiotically interesting species leached from dust-rich deposits in cryoconite holes may drain along with meltwater to the level of the water table (Fig. 5a) and become diluted in the process<sup>39</sup>.

In contrast, the dry valleys of Antarctica shelter cryoconite holes, which form with an ice surface lid and have cold surrounding ice limiting lateral water transport. These holes result from the particular and unique conditions of the dry valleys, namely extreme aridity and cold, with surface melting possible for only brief periods in summer. Solar radiation heats low-albedo inclusions within the ice, which then melt the ice around them in a local greenhouse effect, despite the icy lid<sup>32</sup> (Fig. 5b). Here, biogeochemical recycling results in the waters of cryoconite holes becoming 100-fold enriched in bioavailable forms of limiting nutrients relative to surrounding ecosystems<sup>32</sup>. Abundant life occurs in such systems, mostly supported by nutrients leached from the sedimentary dust deposits at the base of the cryoconite holes<sup>40</sup>. In a prebiotic Earth scenario, we may, therefore, expect similar cosmic-sediment-filled environments to form on the surface of impermeable ice lids that are abiotically enriched in prebiotic feedstock.

Nonetheless, even dry-valley-type cryoconite deposits are subject to transport through ice dynamics of up to  $20 \text{ m yr}^{-1}$  (ref. 41), precluding long-term formations. However, the regular destabilization of cryoconite sediments directly supplies meltwater and sedimentary material to endorheic proglacial lakes (Fig. 5c). Indeed, ecosystems in these closed proglacial lakes are known to depend on fertilization by cryoconite-derived nutrients<sup>32,42</sup>. Glacier margins, therefore, provide settings capable of both locally concentrating cosmic dust and initiating closed-system aqueous prebiotic chemistry with the products from the dissolution and leaching of cosmic dust.

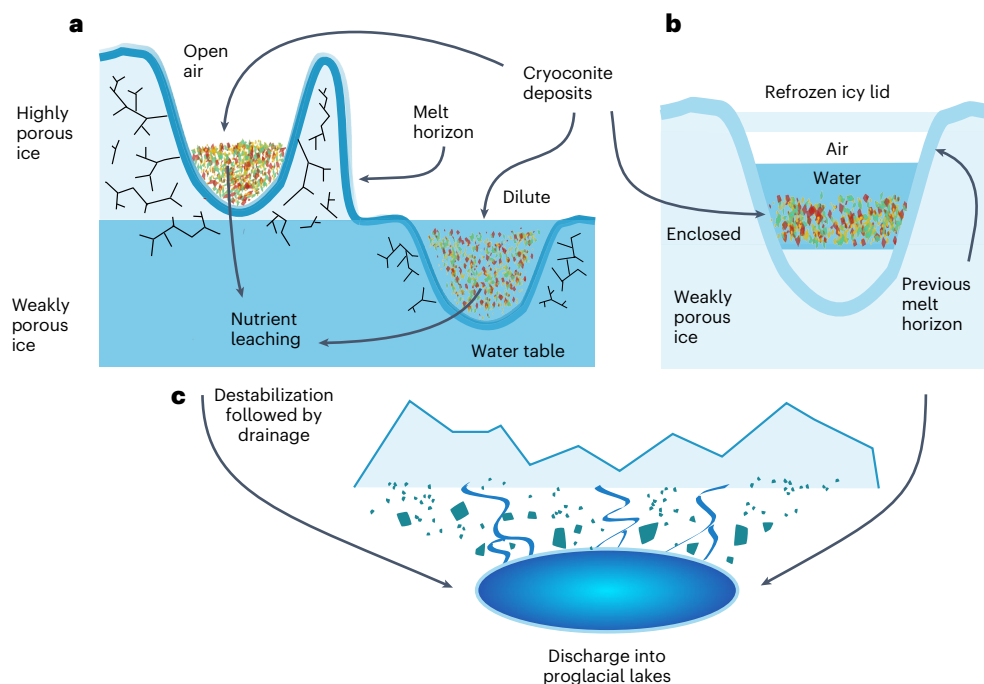
Both cosmic-dust-rich cryoconite sediments and endorheic proglacial lakes would appear to have many attractive properties for both initiating and sustaining prebiotic chemistry. These environments may be networks of icy prebiotic chemical ‘reactors’, replete with the known advantages of ice-hosted prebiotic chemistry: freeze–thaw wet–dry cycles (annual–daily)<sup>43</sup>, low water–rock ratios, potential for UV irradiation as well as UV shielding<sup>3,44</sup>, and the ability to exchange over weekly-to-yearly timescales with other cosmic-dust-filled reactors<sup>20</sup>, analogous to the stream intersection models envisaged by Sutherland<sup>45</sup> and Rimmer<sup>46</sup>.

The geochemistry of cosmic-dust-rich sediments provides unique advantages over and above glacial settings hosting sediments of solely terrestrial geochemical character. In particular, identifying soluble, early Earth sources of P and S is a long-running challenge<sup>47,48</sup>. Prebiotic chemistry directly initiating after exposure of cosmic dust deposits to liquid water in, for example, cryoconite holes or proglacial lakes would have had access to reactive P- and S-rich materials of a very fine grain size, namely:

- P (up to 1,500 ppm) – schreibersite ( $\text{Fe}_{3\text{P}}$ ), apatite ( $\text{Ca}_5\text{PO}_4[\text{OH}, \text{Cl}, \text{F}]$ ), merrillite ( $\text{Ca}_9\text{NaMg}(\text{PO}_4)_7$ ) (ref. 49);
- S (up to 5 wt%) – troilite ( $\text{FeS}$ ), pyrrhotite ( $\text{Fe}_{1-x}\text{S}$ ), pentlandite ( $\text{Fe}, \text{Ni}$ )<sub>9</sub>S<sub>8</sub>, pyrite ( $\text{FeS}_2$ ), chalcopyrite ( $\text{Cu}, \text{Fe}$ )S<sub>2</sub>, sulfonic acids<sup>50,51</sup>.

These materials are analogous to the powders used to accelerate the dissolution of chemical reagents in a laboratory setting and often have a grain size of tens to hundreds of micrometres, like the size distribution of cosmic dust particles<sup>13,31</sup>. Such fine particles aid dissolution and, hence, help to sustain a high throughput of key species for prebiotic chemistry. Indeed, glacial meltwater discharge zones are known today to yield among the highest known fluvial fluxes of reactive and particulate phosphate on Earth<sup>52</sup>.

Some nontrivial fraction of the P content of early cryoconite sediments would have been speciated as phosphide (perhaps around 50%)<sup>49</sup>. The action of UV light combined with dissolved  $\text{H}_2\text{S}$  and  $\text{HS}^-$  species has been shown to be capable of oxidizing species released during phosphide corrosion in water, forming phosphate<sup>53</sup>, a key constituent in and



**Fig. 5 | Schematic illustration of sedimentary deposition of cryoconite rich in cosmic dust. a**, Open-air deposits may exist above the local water table, whereas dilute deposits lie in it. However, from a prebiotic chemistry perspective, both will suffer from nutrient leaching into a wider volume of diffuse water in the variably porous ice sheet. **b**, This is not the case for icy-lid cryoconite deposits, which are encased on all sides by weakly porous ice to cold impermeable ice.

**c**, Regardless of their setting, cryoconite deposits are inherently unstable and most will be destabilized and drained within multi-annual timescales. Drained meltwater and cryoconite sediments will be transported in part to proglacial endorheic lakes, where longer-term stockpiling of dust-derived species may occur.

a catalyst for prebiotic chemistry<sup>54</sup>. Sulfide and its derivative oxidized species (for example, bisulfite) have further been shown to assist the prebiotic syntheses of nucleic acids<sup>3</sup> and many components of central carbon metabolism<sup>55</sup>. Given the apparent scarcity of high-throughput sources of fine-grained sulfide, phosphate and phosphide on Earth<sup>47,48</sup>, we suggest that cosmic dust sedimentation in glacial systems provides a highly relevant geological feedstock for prebiotic chemistry.

Our results show that Antarctic-like ice sheets on prebiotic Earth would have plausibly given rise to aerially extensive regions of cryoconite-hosted cosmic-dust-rich deposits and derivative proglacial lakes, with many attractive properties for prebiotic chemistry. Our results, therefore, highlight a particular environmental context for dust-fed scenarios for prebiotic chemistry, linking the relevance of the scenario to the climate of early Earth. This provides a helpful line of reasoning by which our scenario can be falsified on planetary climatic grounds alone. At present, it is unclear whether or not the glacial environments needed to forge concentrated cosmic dust deposits would have been common on early Earth. Glaciers are known to have formed up to 2.5 billion years ago on Earth<sup>56</sup>. Recent models also suggest a cold early Earth<sup>57</sup>, coincident with the time frame explored in our study and which could support our scenario.

With regard to C and N, we suggest that cosmic dust may also have been a stockpiling agent for prebiotic chemistry, rather than a direct participant. Alongside the arrival of single large impactors<sup>58</sup>, cosmic dust sedimentary cycling may be a plausible mechanism by which C- and N-rich cometary matter can become involved in prebiotic chemistry. Although the soluble component of cosmic dust will be liberated rapidly during any interaction with liquid water, much of the C and N content of cosmic dust arrives in the form of mostly insoluble and inert species. These include N-bearing kerogen (largely poly-HCN), N-heterocycles, amino acids, amines, amides, purines, polar species, aromatics, and hydroxy, dicarboxylic and carboxylic acids<sup>59</sup>.

Instead of suggesting their irrelevance to prebiotic chemistry, the inert character of many of these species strongly implies their capacity to accumulate over time. In settings with low rates of terrestrial sediment deposition (glacier surfaces, for example), cosmic dust could have built up sedimentary blankets with around 0.2 wt% N and 5 wt% C (Fig. 4c). Transport fractionation of organics relative to silicate phases could have generated still greater C and N enrichment. A pure nitrogenous kerogen sedimentary end member would have around 3 wt% N and 70 wt% C (ref. 60). Indeed, extreme organic C enrichments are observed in cryoconite sediments with increasing distance inland across the ablation zone of the Greenland ice sheet, at least in part due to transport fractionation of organic materials<sup>61</sup>. A key caveat is that such sediment compositions are possible only by assuming that the atmospheric entry of dust through a reducing anoxic atmosphere promotes minimal loss of N, in contrast to the substantial (90–99%) loss observed during entry through Earth's present-day oxidizing atmosphere (Fig. 4c)<sup>62</sup>.

Stockpiling mechanisms are commonly invoked in proposed scenarios for cyanosulfidic prebiotic chemistry<sup>45</sup>. The thermal processing of inert CN-rich and relatively H-poor reducing sediments, for example, by meteorite impacts<sup>45</sup> or melting into magma chambers<sup>63</sup>, could have supplied prebiotically relevant concentrations of HCN to overlying aqueous environments<sup>64</sup>. Such a mechanism of continuous HCN supply could be made possible by an intermediate stage of sedimentary sorting and stockpiling of dust-derived inert CN-bearing organics. Due to cosmic dust sedimentation and a heating pathway—closely analogous to the volcanic remobilization of sedimentary organic C on Earth today<sup>64</sup>—the requisite starting ingredients for cyanosulfidic prebiotic chemistry could plausibly have been delivered to, concentrated in and liberated into clement environments for the origin of life.

We have discussed different geological mechanisms that could separately supply P + S and C + N from cosmic dust to prebiotic chemistry.

However, although these mechanisms operate across different spatial scales and timescales, they are not mutually exclusive. Thermally processed deposits of exogenous organic matter may have degassed HCN into lake systems fed by glacial meltwater rich in P and S sourced from locally dissolving cosmic dust. Taken together with recent findings from geology, astronomy and prebiotic chemistry, our results provide support for the fertilization of prebiotic chemistry by cosmic dust on early Earth. Furthermore, cosmic dust is potentially a widespread and flexible planetary fertilizer, being accreted in quantities that may be assessed by observation<sup>9,65</sup> to potentially habitable exoplanets.

## Methods

### Model parameters for cosmic dust deposition

Cosmic dust grains in pristine modern sedimentary systems appear to derive mainly from undifferentiated carbonaceous chondrite or cometary precursor bodies<sup>66,67</sup>. Cosmic dust that appears to originate from differentiated asteroids represents less than 5% by mass of all cosmic dust in most deposits<sup>68</sup>. The compositional disparity between modern accretion fluxes of cosmic dust and predicted early fluxes must be considered in interpreting the relevance of our results for understanding the supply of bioessential elements to early Earth's surface. Bridging this gap requires us to make assumptions, both about the parent bodies of cosmic dust falling to Earth today and how those precursor objects are represented in our simulations.

We assume that all cometary material is well represented by the compositions of unmelted micrometeorites and apparently cogenetic carbonaceous chondrite meteorites<sup>68</sup>. Dust from asteroids is conservatively assumed to sample differentiated precursor bodies. Cosmic dust grains from differentiated material is considered to represent an equal mixture of crust and core material and to be well represented by the compositions of silicate and iron-type grains arriving on Earth, today<sup>69</sup>. These assumptions inform the values in Supplementary Table 1.

Particle diameter exerts strong control on the survival of cosmic dust during atmospheric entry (Supplementary Fig. 2). The majority of particles with sizes <0.1 mm survive to the Earth's surface with minimal heating<sup>29</sup>, and empirical studies demonstrate that they retain a relevant proportion of their temperature-sensitive organic matter<sup>15</sup>. We estimated the proportion of early accreted material that survives unaltered versus that which melts and that which vaporizes using the results of Love and Brownlee<sup>29</sup> (Supplementary Fig. 2). Again, we take an all-else-being-equal approach to justify this approach, in which the atmosphere of early Earth is considered to be of similar mass to today.

Our findings are robust to uncertainties regarding the assumed composition of the terrestrial sedimentary component. Although we plot all results relative to and assuming a modern-day average upper continental crust for terrestrial sediment within the modelled early dust deposits, different choices would result in higher reduced fractions or nutrient-enrichment factors. This is because estimated early crustal compositions that differ from modern average upper continental crust are instead more like Earth's mantle<sup>33</sup> with lower concentrations of P, S, C and N, resulting in their stronger relative enrichment in early cosmic dust-rich sedimentary deposits. We have not modelled the leaching of bioessential elements from dust during sedimentary transport. This assumption is valid for the arid conditions and aeolian transport mechanisms operating in the desert and glacial settings that favour the formation of cosmic dust deposits.

Although stochastic and relatively short-lived on geological timescales (several million years), intervals of comet-dominated dust supply occur with high probability in any given simulation run. These intervals are also sufficiently long-lived to allow complete cycling of cosmic dust through all the surface environments that we consider<sup>13,23</sup>. Therefore, although the background supply of dust provides notable enrichments, our results highlight that stochastic large cometary break-up events are crucial for producing dust-rich sediments with the highest concentrations of bioessential elements.

### Estimating cosmic dust abundance within early sediments

We take an all-else-being-equal approach to assess the potential for the formation of cosmic sediment on early Earth. Critically, we assume that the deposition flux of dust in a given environment directly scales with the flux accreted to Earth. That is, processes acting to concentrate dust operate with equal efficiency at higher dust accretion rates. This assumption is defensible because maximum sediment loading is expected to be extremely low in the low sedimentation environments that we consider, even given much higher early cosmic dust accretion fluxes.

Given these assumptions, we can set up a linear relationship between the cosmic dust accretion flux to early Earth relative to the at-atmosphere flux estimated today ( $\epsilon_x$ ) and the ratio of cosmic dust mass per unit mass of sediment in a given sedimentary system ( $\delta$ , kg kg<sup>-1</sup>). As cosmic dust fluxes increase, total sediment within the system of interest will increase. To track this, we must include as a constant the original mass of terrestrial sediment mass per unit mass of sediment ( $1 - \delta$ ) in the sedimentary system. The fraction of cosmic dust in the putative early sedimentary system ( $f_{\text{dust}}$ ) is then given by

$$f_{\text{dust}} = \frac{\delta_{\text{modern}} \epsilon_x}{\delta_{\text{modern}} \epsilon_x + (1 - \delta_{\text{modern}})} \quad (1)$$

The principal parent body sources are considered to be asteroids and comets, so that:

$$f_{\text{dust}} = \frac{\sum_{x=\text{cometary, asteroidal}} F_x \delta_{\text{modern}} \epsilon_x}{\sum_{x=\text{cometary, asteroidal}} F_x \delta_{\text{modern}} \epsilon_x + (1 - \delta_{\text{modern}})} \quad (2)$$

We can calculate the concentration of a given element in an early sedimentary deposit ( $\gamma$ ) as a mixture of the chemical compositions ( $\beta$ ) (Supplementary Table 1)<sup>13,15,25,33,38,62,70-73</sup> of endogenous sediment (E) and cosmic dust as follows:

$$\gamma = \frac{\sum_{x=\text{cometary, asteroidal}} F_x \delta_{\text{modern}} \epsilon_x \beta_x + (1 - \delta_{\text{modern}}) \beta_E}{\sum_{x=\text{cometary, asteroidal}} F_x \delta_{\text{modern}} \epsilon_x + (1 - \delta_{\text{modern}})} \quad (3)$$

The proportion of the early dust sedimentation flux made up by each cosmic dust class is also partially determined by the effects of atmospheric entry. Volatiles may be preferentially lost during atmospheric entry heating of dust grains<sup>14,15,38</sup>. We use empirical observations of the volatile chemistry of cosmic dust particles to inform our calculations, which show that S, C and P are largely conserved during entry, even if they may be to some extent chemically reorganized<sup>74</sup>. In contrast, a large fraction of nitrogen is lost from cosmic dust particles during atmospheric entry, largely due to degradation by oxidation<sup>62</sup>. Entry into an anoxic and possibly reducing early atmosphere, relevant for early Earth, may have been far less efficient for destabilizing and removing nitrogen from dust grains, an end-member scenario which we consider here.

As cosmic dust vaporization during atmospheric entry is heavily dependent on particle size<sup>29</sup>, we examined whether the size frequency distribution of early dust accretion fluxes predicted by our model differs to that observed today. We use the results of Love and Brownlee<sup>29</sup> to estimate the proportion of all cosmic dust that is vaporized versus melted or survived unaltered during atmospheric entry today. To facilitate a like-for-like comparison between our results and those of Love and Brownlee<sup>29</sup>, we generate continuous distributions of accreted mass as a function of grain size from histograms of mean mass accreted per grain size bin. We interpolate between the centre point of each bin. These continuous distributions are then used to predict the fraction of early cosmic dust grains that vaporize ( $f_v$ ) during entry for each source population (Supplementary Fig. 2).

Overall, the size distributions of material arriving at the top of the atmosphere and arriving at the surface after entry are essentially like



those observed today<sup>13</sup>. However, the exact ablation behaviour may differ for each population based on mineralogical and entry velocity differences<sup>10,16</sup>. To largely eliminate this uncertainty, we proceed with our approach of anchoring to empirical observations. We compare the flux estimate of cometary versus asteroidal dust arriving at Earth's surface to top-of-atmosphere flux estimates to obtain an empirical value for  $f_v$ . Similarly, we use empirical observations of unmelted to melted dust particles to estimate the fraction that experiences melting during entry ( $f_m$ ) and compositional analyses to estimate the fraction of volatiles lost during entry from both unmelted and melted particles ( $f_{\text{loss}}$ ). This approach then yields:

$$\gamma = \frac{\sum_{x=\text{cometary, asteroidal}} (1-f_v)(\theta_x \beta_x f_m f_{\text{loss}} + \theta_x \beta_x (1-f_m)) + (1-\delta_{\text{modern}})\beta_E}{\sum_{x=\text{cometary, asteroidal}} (1-f_v)(\theta_x f_m f_{\text{loss}} + \theta_x (1-f_m)) + (1-\delta_{\text{modern}})} \quad (4)$$

where  $\theta_x$  is equivalent to  $F_x \delta_{\text{modern}} \epsilon_x$ .

Uncertainty regarding the measured masses of cosmic dust per kilogram of sediment is obtained from literature reports, which were conducted at different times, in different places and using different methods to both obtain sediment and to filter cosmic dust from the material. Ultimately, the uncertainty for the proportion of cosmic dust in the sediment is trivial compared to that introduced by assumptions made during the construction of the dust flux model. For example, rounding up reported the  $2\sigma$  uncertainty on cosmic sediment fraction to the nearest interval, uncertainties of around 10% characterize the majority of measurements. Details of the values of all parameters used in our model for cosmic sediment composition can be found in Supplementary Table 1.

### Numerical model of collisional dust generation and transport to Earth

We simulated the distributions of dust produced by two compositionally distinct parent body populations: JFCs and asteroids. Asteroids were treated as the sum of two dynamically distinct populations: relatively stable main belt objects and rapidly depleted unstable planetesimals left over from planet formation. For each population, the overlap of the dust grain orbits with Earth was used to calculate the resulting fluxes of cosmic dust accreted onto Earth.

Intrinsic to this model are assumptions about the history of the Solar System and its formation. For the start time of our models, we use the Moon-forming impact. Models in the literature suggest that this occurred early,  $\sim 50$ – $100$  Myr after the formation of calcium-aluminium-rich inclusions<sup>75–77</sup>. We assume that the Moon-forming impact occurred at 50 Myr, which is time zero of our simulations. We also assume that the giant planet instability occurred early<sup>78,79</sup>. Simulations were run for 500 Myr for each source population.

We modelled the distributions of dust produced by each source population separately using a kinetic model<sup>80</sup> that follows the evolution of a population of particles in terms of their orbital elements and sizes. The numbers of particles in bins of particle size, their pericentre and their eccentricity evolve due to several forces. Destructive collisions between particles remove particles that collide and produce smaller fragments, Poynting–Robertson drag causes particles to lose angular momentum and drift towards the Sun and radiation pressure acts radially outwards on small particles.

**Comets.** The dust produced by JFCs was simulated using the model of ref. 81, which was originally created to model the production of the present zodiacal cloud through the spontaneous fragmentation of comets. We updated the model to allow comets to be scattered at a variable rate. In the early Solar System, comets should be scattered inwards at a much higher rate than today. The rate of comet scattering was extrapolated from  $N$ -body simulations that followed the distribution of JFCs throughout the 4.5 Gyr history of the Solar System<sup>82</sup>.

The input rate of comets was found to be tens to hundreds of times higher in the first 500 Myr of the Solar System than the present rate and declines with time (Supplementary Fig. 3). This cometary model gives the distribution of dust produced by comets with time, which was then input into the kinetic model to find how the dust evolves after it is released from the comets.

**Asteroids.** For the asteroid belt, the kinetic model was initialized with bodies with the size distribution<sup>83</sup> of the current belt, going from diameters of 1 to 1,000 km. The initial orbits of asteroids were assumed to be those of the present main belt (JPL Small-Body Database, [https://ssd.jpl.nasa.gov/tools/sbdb\\_query.html](https://ssd.jpl.nasa.gov/tools/sbdb_query.html)). The initial mass of the asteroid belt and its subsequent evolution is poorly constrained. Although the collisional evolution of asteroids will slowly deplete the overall mass of the belt, it is probable that the dynamical evolution of planets dominated the depletion of the asteroids (for example, ref. 84). Based on ref. 85, we assume that following the giant planet instability, the asteroid belt had a mass four times higher than its present mass ( $\sim 5 \times 10^{-4} M_{\oplus}$ ; refs. 86,87) and that it lost half of its mass over the subsequent 100 Myr by depletion of unstable resonances. We, therefore, initialized the model with a mass of  $2 \times 10^{-3} M_{\oplus}$  of asteroids. The asteroids then evolve in the kinetic model by collisional evolution, producing dust, which further evolves due to collisions and radiation forces.

**Rapidly depleted asteroids.** Similarly, the collisional evolution of asteroids left over from terrestrial planet formation was traced with the kinetic model. The size distribution was assumed to be the same as that of the current asteroid belt<sup>83</sup>. The orbits of these asteroids are taken from  $N$ -body simulations<sup>88</sup>, which were run to find the bombardment history of early Earth by rapidly depleted objects. The total initial mass of rapidly depleted asteroids was chosen using an iterative process to match the observation that  $0.005 M_{\oplus}$  (ref. 89) of highly siderophile elements should have been accreted to Earth during the Late Veneer from rapidly depleted asteroids. The number of rapidly depleted asteroids present in the inner Solar System decreases roughly exponentially with time due to a combination of ejection from the system by Jupiter, accretion onto Earth and collisional evolution. The  $N$ -body simulations give the dynamical depletion of bodies with time, whereas our kinetic model gives the collisional depletion. By calculating the mass of asteroids accreted onto Earth when taking into account both the dynamical and collisional depletion, we found that an initial mass of  $0.065 M_{\oplus}$  of asteroids left over after terrestrial planet formation led to  $0.005 M_{\oplus}$  of asteroids being accreted by Earth. We initialized this mass of asteroids in the kinetic model on the orbits from the  $N$ -body simulations. The model then accounted for the collisional depletion of asteroids, which produced smaller bodies and thus dust, and the dynamical depletion of asteroids, which removed bodies from the simulation according to the evolution found by the  $N$ -body results. The model, therefore, gives the distribution of dust produced by the collisional evolution of rapidly depleted asteroids left over from planet formation.

Having found the distribution of dust in the inner Solar System over time using the kinetic model, we then found the accretion rates of dust from each source population onto the early Earth as a function of time. The rate of each orbit overlapping with the Earth was found with the method of ref. 90, which was multiplied by the population of dust on each orbit to give the cosmic dust flux to Earth with time from each source.

### Data availability

All relevant data needed to evaluate the findings of the manuscript are included in the figures or in Supplementary Data 1.

### Code availability

Details of specific codes used to perform simulations are available upon request.



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## Author contributions

C.R.W. conceived the project and performed the meta-data curation and analysis and geological modelling. J.K.R. wrote the numerical model and performed all the astrophysical simulations. O.S. and M.W. supervised the project. All authors contributed to the writing and editing of the paper's text and figures.

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## Competing interests

The authors declare no competing interests.

## Additional information

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