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Revisiting the Eoarchean Akilia quartz-pyroxene rock with potassium isotopes: Implications for early-ocean sedimentation

Zheng-Yu Long[a,b,1], Frédéric Moynier[a,1], Wei Dai[a], Mark van Zuilen[c,d], Jun Deng[b], and Kun-Feng Qiu[b,1]

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The Eoarchean quartz-pyroxene rock from Akilia Island in Greenland has been proposed as one of Earth's oldest banded iron formations (BIF) and a potential host for the earliest biosignatures. However, the origin of its protolith, whether it metamorphosed from an igneous or sedimentary precursor, remains debated. Here, we revisit this long-standing Akilia controversy using potassium isotope analyses, comparing Akilia samples with BIFs and black shales spanning the Eoarchean to Mesoproterozoic. Our results demonstrate that BIFs and black shales show systematic potassium isotope variations correlated with their potassium contents. Potassium-poor BIF layers display heavier isotopic compositions close to seawater values, whereas clay-rich layers exhibit elevated potassium contents and lighter isotopic signatures. The Akilia quartz-pyroxene rock was initially characterized by low potassium concentrations and heavy potassium isotopic compositions consistent with chemical sediments deposited from ancient seawater. It was subsequently modified by metasomatic fluids derived from nearby metamorphosed igneous rocks. These findings support a sedimentary origin for the Akilia quartz-pyroxene rock. Furthermore, our study provides an isotopic framework for interpreting ancient oceanic environments and offers insights into the potassium cycling and habitability of early Earth.

potassium isotopes | banded iron formation | Akilia quartz-pyroxene rock | continental weathering | early earth

Significance

The Eoarchean (>3.65 Ga) quartz-pyroxene rock from Akilia, Greenland, represents a key fragment of Earth's earliest rock record. By integrating novel potassium isotope data with petrological observations, our study identifies a chemical sedimentary signature overprinted by metasomatic processes. These findings strengthen the evidence for sedimentary environments in Earth's nascent crust and shed light on Archean continental weathering and potassium fluxes to the early oceans. Our work thus underscores the potential of potassium isotopes as a powerful tool for deciphering ocean chemistry and nutrient cycling during the planet's formative stages.

Understanding the origin of the Eoarchean (>3.65 Ga) banded quartz-pyroxene (qtz-px) rock from Akilia is of fundamental importance. It bears on how soon oceanic sedimentary processes began and where the earliest biosignatures might be sought. This rock, found on Akilia Island in Greenland, was initially interpreted as a ferruginous quartzite containing ^{13}C-depleted graphite, a geochemical signature associated with biogenic activity (1). If true, it would represent evidence of oceanic sedimentation and possibly life over 3.83 billion years ago (2–4). This would make Akilia one of the oldest water-laid sedimentary deposits on Earth, contemporaneous with the Isua supracrustal belt, which hosts some of the oldest documented banded iron formation (BIF) (5, 6). However, the Akilia rock has undergone significant metamorphism and intense deformation, obscuring its original characteristics (7). This has led to a vigorous debate about its protolith: Does the Akilia rock truly represent chemical sediment deposited in an early ocean (akin to BIFs) (1, 3, 4, 6, 8–10), or is it a metamorphosed ultramafic igneous rock altered by fluid activity (5, 7, 11–14)?

BIFs are chemical sediments consisting of iron-rich layers interbedded with silica (chert), spanning from the Archean to Proterozoic (15, 16). Most unaltered BIFs have very low K content, as primary iron oxides, carbonates, and silica incorporate negligible K (15). The low abundance of K in many BIFs affirms that they are characteristically "clean" chemical sediments (17, 18), including the oldest BIFs at ~3.8 to 3.7 Ga in Isua (10). When present, K often correlates with Al (clays) or Ti (heavy minerals) and, to a lesser extent, Zr and Hf, indicating detrital contamination (19). This is because continents grew and emerged through the Late Archean–Paleoproterozoic, and occasional detrital inputs are involved in some BIFs (20). Moreover, during early diagenesis, Fe(III)-oxyhydroxide gels and cherts may react with interstitial clays or seawater ions. If pore fluids supply K (e.g., from volcanic ash or overlying sediments), stilpnomelane [$K(Fe,Mg)_8(Si,Al)_{12}(O,OH)_{27}$] forms authigenically (15, 21). Furthermore, if metamorphic fluids introduce K from adjacent strata, even originally K-free BIFs can develop K-bearing silicates (22). Therefore, the occurrence and variability of minor amounts of K subtly record early sedimentary and later alteration processes.

Potassium (K) stable isotopes (expressed as δ^{41}K) have emerged as a powerful tracer for distinguishing low-temperature sedimentary processes from high-temperature igneous ones. In magmatic systems, K isotope fractionation is minimal (23–25). In contrast, during chemical weathering of silicate rocks, the lighter ^{39}K is preferentially incorporated into

Author affiliations: [a]Université Paris Cité, Institut de Physique du Globe de Paris, CNRS, Paris 75005, France; [b]State Key Laboratory of Geological Processes and Mineral Resources, Frontiers Science Center for Deep-time Digital Earth, School of Earth Sciences and Resources, China University of Geosciences, Beijing 100083, China; [c]CNRS-UMR (Unité Mixte de Recherche) 6538 Laboratoire Geo-Ocean, European Institute for Marine Studies, Université de Bretagne Occidentale, Plouzané 29280, France; and [d]Naturalis Biodiversity Center, Leiden 2333CR, The Netherlands

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[1]To whom correspondence may be addressed. Email: long@ipgp.fr, moynier@ipgp.fr, or kunfengqiu@qq.com.

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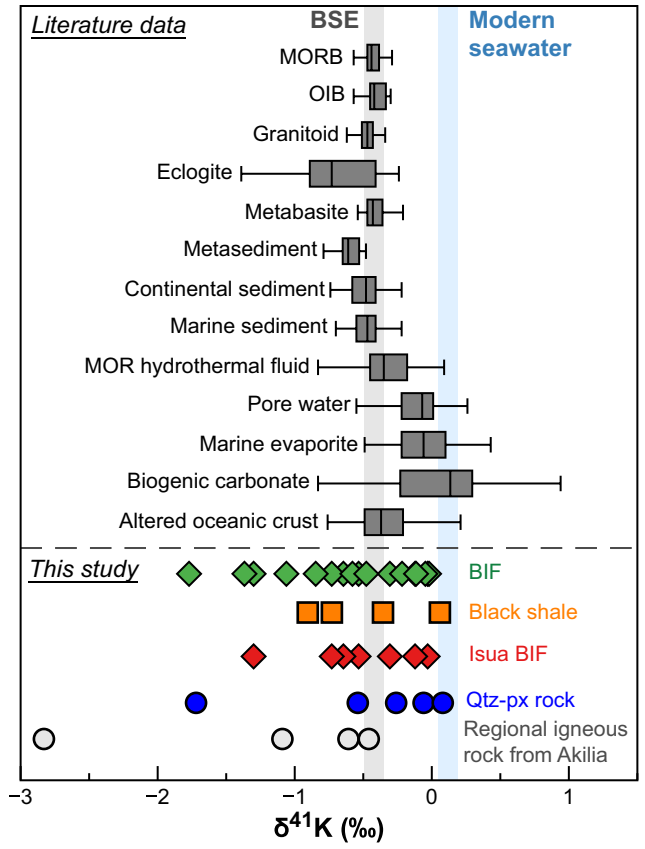

Fig. 1. Potassium isotopic compositions of the investigated samples, compared with previously reported data. The BSE range is adapted from ref. 25, while modern seawater is sourced from ref. 28. Data sources for reported materials are detailed in *SI Appendix*, Table S2.

secondary minerals (e.g., clays), enriching dissolved loads in ^{41}K (26, 27). Modern seawater is isotopically heavy (0.12 ± 0.07 ‰) (28) relative to igneous rocks and is, in fact, the heaviest K reservoir on Earth. Marine carbonates and evaporites generally have elevated $\delta^{41}K$ values due to the incorporation of seawater K, including that in porewater and mid-oceanic ridge fluids (Fig. 1), whereas igneous rocks derived from the mantle (−0.42 ± 0.13 ‰) (23, 25, 29) or continental crust (−0.44 ± 0.05 ‰) (30) retain lighter compositions (approximately −0.5 to −0.3‰; Fig. 1). Here, we apply this principle to the Akilia qtz-px rock. By measuring $\delta^{41}K$ in the Akilia samples and comparing them to known BIFs and black shales, we provide an independent assessment of the competing hypotheses for its origin.

Results

The analytical results of element concentrations and isotope compositions are listed in *SI Appendix*, Table S1. Bulk-rock analyses of $\delta^{41}K$ for the Akilia qtz-px rocks (+0.08 to −1.72 ‰), Archean–Proterozoic (3.8 to 1.5 Ga) BIFs (−0.02 to −1.78 ‰), and black shales (+0.06 to −0.91 ‰) span from near-seawater values to substantially lighter compositions. In contrast, representative samples (granitoid gneiss, ultramafic rock, and gabbroic rock) from Akilia with documented igneous protoliths range from BSE-like values to notably lighter values (−2.83 to −0.46 ‰). Notably, the well-preserved fine-grained Akilia qtz-px sample (0.08 ± 0.08 ‰) and the fine-grained Isua BIF sample (−0.03 ± 0.06 ‰) both exhibit relatively high $\delta^{41}K$ values. K concentrations also vary widely, from 111 to 4222 μg/g in the Akilia qtz-px rocks, 31.7 to 29,609 μg/g in BIFs, and 12,556 to 23,356 μg/g in the black shales. In the BIF samples (including Isua), variations in K concentrations positively coincide with their Al, Ti, Zr, and Hf contents (*SI Appendix*, Fig. S4).

Discussion

Early-Oceanic K Cycling and Sedimentary K Isotope Signatures. The continental crust in the Early Archean was either sparse or largely submerged; thus, seafloor hydrothermal fluids likely dominated oceanic K cycling during this period. From the Late Archean into the Paleoproterozoic, as continents progressively emerged (31), riverine dissolved loads became the primary source of K to seawater (28). Once K-bearing biogenic silica was deposited onto the seafloor, authigenic minerals (e.g., clays) could form, preferentially incorporating lighter ^{39}K (32). For instance, authigenic glauconite, which precipitates directly from seawater, typically exhibits lower $\delta^{41}K$ (e.g., −0.3 ‰) (33). In addition, detrital (terrigenous) inputs also influenced the K cycling and the isotopic compositions of oceanic sediments. Secondary clays formed during continental weathering (e.g., illite, vermiculite, and smectite) preferentially retain lighter ^{39}K (e.g., $\delta^{41}K$ down to −0.74 ‰) (33–36). When transported via rivers into marine basins, these detrital materials carry this distinct low $\delta^{41}K$ signature. Thus, the notably low $\delta^{41}K$ values (down to −1.3 ‰) observed in marine sediments can partly be explained as inherited from continental weathering residues (37). In fact, both detrital input and diagenetic clay formation can coexist and reinforce the isotopically light K trend in sediments. An inverse [K]–$\delta^{41}K$ relationship was observed in BIFs and, to a lesser extent, black shales (Fig. 2). This represents a consequence of mixing between two endmembers, i.e., heavy seawater-derived K and light clay-host K. BIFs and black shales represent two sedimentary modes and differ in how they gain K.

Black shales are clastic, fine-grained sediments deposited from muds rich in continental debris, mainly consisting of detrital clay minerals along with silt-size quartz and organic matter (38). These clay minerals were formed on land and transported to the basin, bringing with them the K that was stripped of heavy isotopes during continental weathering (26, 33, 37). As a result, in black shales with high K contents, $\delta^{41}K$ values are notably lower (Fig. 2), reflecting the dominance of ^{39}K-rich detrital aluminosilicates. Meanwhile, dissolved K^+ in pore fluids is efficiently removed via adsorption and authigenic clay formation (39, 40). Profiles of deep-sea sediments reveal that K is progressively extracted from pore fluids into solid phases (39); diagenetic clays thus can inherit the heavier K isotopes from the K-rich fluid. In addition, in Precambrian black shales, burial metamorphism (at temperatures of 100 to 200 °C) drives the transformation of smectitic clays to illite (38), during which formed clays might drive $\delta^{41}K$ to more negative values (33–35).

By comparison, BIFs are Precambrian chemical sediments with minimal terrigenous (detrital) input (17, 18). K in BIFs is mostly from seawater or pore fluids, perhaps adsorbed onto ferric oxides/hydroxides. Any addition of K is from minor clay or mica minerals introduced during or after deposition (15, 21), as supported by the positive correlation of K contents with detrital proxies such as Al, Ti, Zr, and Hf contents (*SI Appendix*, Fig. S4). Given that the oceanic K reservoir was isotopically heavy, even trace levels of K in chemically precipitated sediments like BIFs may display elevated $\delta^{41}K$ values, especially as mineral surface adsorption preferentially captures heavier ^{41}K (32). Thus, pure BIFs are K-depleted and exhibit high $\delta^{41}K$ (Fig. 2), indicating a primary contribution of seawater K adsorbed by Fe oxides/hydroxides. In contrast, any minor diagenetic K additions (e.g., ferroan clays or micas formed at late stages) likely alter their original $\delta^{41}K$ values. As a result, the

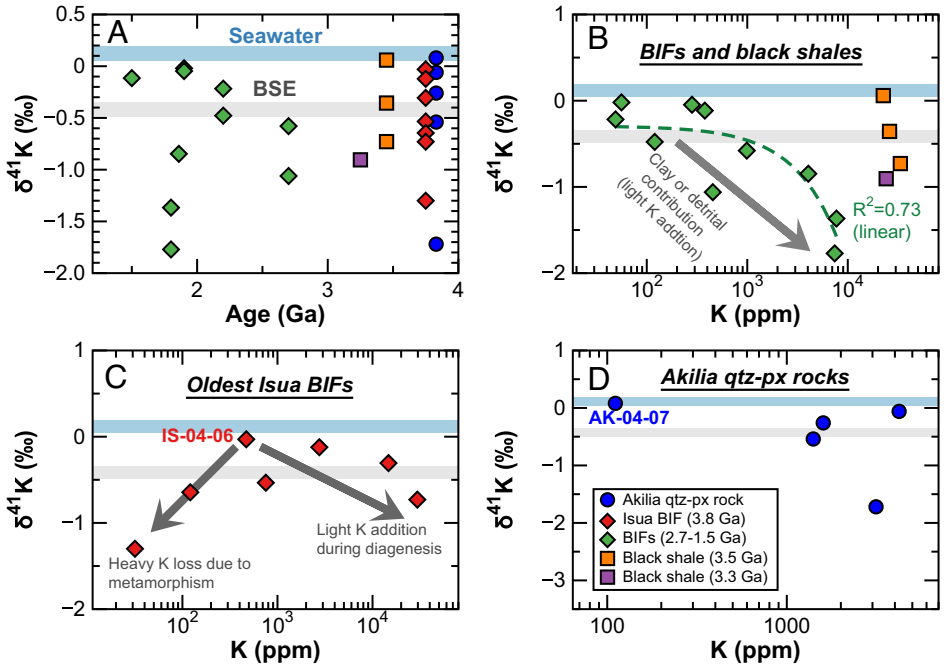

Fig. 2. Plots of (*A*) δ^{41}K versus Age (Ga) and (*B-D*) δ^{41}K versus K contents. The BSE range is adapted from ref. 25, while modern seawater is sourced from ref. 28.

more K-bearing clay a sample contains, the lower its δ^{41}K (Fig. 2*B*) (26, 33, 37).

In summary, the observed inverse [K]–δ^{41}K trends reflect the result of global K cycling from continental weathering and clay formation to marine deposition. This makes δ^{41}K a promising proxy for reconstructing variations in ocean chemistry.

Identifying the Processes Controlling K Isotopic Variation in Isua BIF. The ~3.8 to 3.7 Ga BIFs of the Isua supracrustal belt in Greenland offer rare insights into Earth's earliest sedimentary. Although the ancient formations have undergone regional amphibolite-facies metamorphism (41), they preserve significant primary geochemical characteristics (10). For instance, bulk δ^{56}Fe values in Isua BIFs are heavily positive (+0.3 to +0.5‰) relative to igneous rocks (6, 10). Such heavy Fe isotopic compositions are explained by partial oxidation of Fe(II) (of hydrothermal sedimentary origin) during deposition, producing Fe(III)-(oxyhydr)oxides that are enriched in ^{56}Fe. These heavy Fe isotopic compositions, along with sedimentary rare-earth element patterns, survive in the metamorphosed BIF (6, 10), suggesting the metamorphic overprint did not fully homogenize or reset the original geochemical signatures. Among the Isua samples, the well-preserved Isua BIF sample (IS-04-06) exhibits the highest δ^{41}K values. Using this as a reference point, the δ^{41}K values exhibit two distinct trends (Fig. 2*C*), implying two processes influenced the K budget of these rocks, i.e., incorporation of clay minerals and isotopic fractionation during metamorphism.

In some Isua samples, δ^{41}K values decrease with increasing K content, similar to the observations in other BIFs (Fig. 2*B*). Isua BIFs have very low initial concentrations of K and Al (most rocks have <0.01 wt% K_2O and <1 wt% Al_2O_3) (10). Any enrichment in K is therefore a clear signal of an externally derived component during deposition or early diagenesis. Detrital input (continent-derived) or authigenic clay deposition (seafloor hydrothermally derived) could explain why more K-rich samples have decreased δ^{41}K values. In contrast, other Isua samples show both K content and δ^{41}K decline. During amphibolite-facies metamorphism (~500 to 600 °C) at Isua, clay or micas (if any) would undergo dehydration reactions, potentially releasing K into fluids. When K-bearing minerals break down, the lighter ^{39}K tends to be retained in the solid residue, while the fluid is enriched in ^{41}K (42). Thus, the Isua BIF layers would have lost an amount of isotopically heavy K into metamorphic fluids, lowering their δ^{41}K values and K contents. Likewise, the decrease of fluid-mobile Ba and Rb contents in the samples is observed (*SI Appendix*, Fig. S5). In particular, the most K-depleted rock (IF-G; also most Ba- and Rb-depleted) shows the lowest δ^{41}K, indicating that K was reset. Metamorphism also significantly modified the initial Nd and Hf isotopic composition of this sample (43).

Despite high-grade metamorphism, the negative trend of δ^{41}K and K concentration (indicative of clay involvement during diagenesis) is observable (Fig. 2*C*). In fact, not all metamorphism fractionates K isotopes. For example, blueschist- to eclogite-facies metasediments from the Western Alps exhibit δ^{41}K values akin to their unmetamorphosed equivalents (44), indicating that sedimentary δ^{41}K signatures persist despite metamorphism. Any metamorphic redistribution of K thus appears to have been on the scale of individual bands or microscale fluid escape, rather than large-scale infiltration that would have equilibrated K isotopes across the whole rock.

Sedimentary Protolith with Metamorphic Overprint for Akilia qtz-px Rock. Although the Akilia qtz-px rock has been interpreted as either highly metamorphosed BIFs (1, 3, 4, 6, 8–10) or ultramafic igneous (5, 7, 11–14), it is well documented that its protolith underwent pervasive alteration by silica- and iron-rich fluids during high-grade metamorphism. This fluid–rock interaction, occurring under amphibolite- to granulite-facies conditions, drastically modified the rock chemistry by introducing external material from surrounding rocks (45). Previous studies have shown that, in contrast to the relatively light Fe isotopic compositions of mid-ocean ridge hydrothermal fluids, the heavier Fe isotopic signatures identified in qtz-px rocks were inherited from partially oxidized seawater (similar to Isua BIFs), resulting in elevated δ^{56}Fe values (6, 10). Meanwhile, decreases in δ^{56}Fe in certain portions of this rock were interpreted as evidence for the introduction of

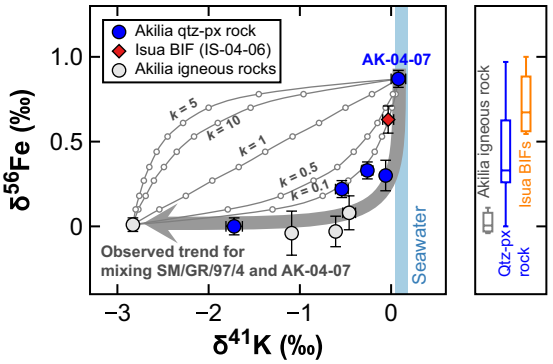

Fig. 3. A plot of δ^{56}Fe versus δ^{41}K. The reported δ^{56}Fe ranges for igneous rocks, qtz-px rocks from Akilia, and BIFs from Isua are also shown (6, 10). Binary mixing between samples SM/GR/97/4 (end-member *1*) and AK-04-07 (end-member *2*) is modeled. Dots indicate 10% increments of each end-member in the mixture. The slope of the binary mixing hyperbolae varies with the *k* value (defined as $k = [K/Fe]_1/[K/Fe]_2$, where K and Fe refer to the element content of end-members). Modeling uses *k* values of 0.1, 0.5, 1, 5, and 10. The modern seawater is sourced from ref. 28.

isotopically lighter Fe, mobilized from surrounding lithologies via fluids (6, 10). However, this interpretation has recently been challenged by discoveries of several igneous surrounding rocks that also exhibit elevated δ^{56}Fe (45), indicating that seawater oxidation may not be the sole cause of the heavy Fe isotopic composition. Here, we employ a combined K–Fe isotopic approach to reassess this debate.

Akilia rocks of documented igneous protolith exhibit δ^{41}K values from –2.83 to –0.46 ‰ (Fig. 1). This signature likely reflects metamorphic processes or the superposition of multiple events (likely involving kinetic processes). In any case, the metamorphosed igneous rocks adjacent to the qtz-px rock are viable sources of lighter ^{39}K. Because earlier high-grade metamorphism had likely driven off heavy K isotopes in igneous surrounding rocks, K-bearing fluids derived from the breakdown of micas or K-feldspar in these rocks would have been enriched in ^{39}K (i.e., low δ^{41}K). Once such fluids infiltrated the qtz-px rock, they introduced isotopically lighter K. Similar to fine-grained Isua BIF, the "pristine" Akilia qtz-px rock (AK-04-07) preserves marine sedimentary signatures marked by a high δ^{41}K value (Fig. 2). It is noteworthy that δ^{41}K and δ^{56}Fe positively covary in the qtz-py rock (Fig. 3), suggesting simultaneous resetting by a common process. Such a coupled isotopic trend is unlikely from sedimentary processes alone, given the distinct geochemical behaviors of K and Fe during sediment deposition and diagenesis (see *SI Appendix* for details). Additionally, closed-system dehydration or internal diffusion under closed-system conditions cannot generate these isotopic trends (*SI Appendix*). Because the original qtz-px rock was nearly devoid of K, any introduction of K had to come from an external source. A plausible scenario is that the qtz-px rock was initially the chemical sediment of seawater/hydrothermal origin with heavier K and Fe isotopes. It was subsequently modified by extensive metasomatic fluid interaction involving adjacent metamorphosed ultramafic rocks (the source of both light Fe and K isotopes), producing intermediate isotopic compositions (Fig. 3). Correlations between δ^{41}K and element ratios (e.g., Mg/K, Mn/K, Ca/K; Fig. 4) further support significant fluid–rock interaction (10).

Heavy K Isotopic Composition in Ancient Seawater. Since modern rivers have an average flux-weighted δ^{41}K of –0.38 ‰ (46), the river input cannot explain substantially elevated δ^{41}K in seawater (0.12 ± 0.07 ‰) (28). The "excess" heavy K was considered as the result of the preferential removal of ^{39}K by K-bearing solids (e.g., clays, zeolites, altered basalts) (33–36). Our data support that continental weathering and associated K input have significantly influenced marine environments since the Archean. This influence may even extend back into early Archean, at least locally, as evidenced by the low δ^{41}K values indicative of continental input recorded in the oldest known black shales and BIFs (Fig. 1). Exposed Archean crust, especially K-bearing feldspar in granitoid (TTG) terrains, served as a source of K. Volcanic weathering (volcanic islands or large igneous provinces) also contributed. High atmospheric CO_2 levels and potentially warm climates during the early Archean have been proposed as factors enhancing rock dissolution (47, 48). These processes supplied K to the oceans (albeit lower in absolute amount than modern fluxes). Two major sinks (burial in clays and alteration of oceanic crust) both remove K from seawater and fractionate its isotopes. In the Archean, when oceans were likely enriched in dissolved silica (due to minimal biogenic silica removal), the formation of authigenic aluminosilicates may have been even more pronounced, as high silica and Fe^{2+} concentrations would promote abundant clay deposition. Much of the dissolved Si has been sequestered into clays since ~3.5 Ga (49). By analogy, this process would drive residual seawater toward higher δ^{41}K due to the preferential incorporation of ^{39}K into clays (26, 32). Furthermore, the Archean seafloor was tectonically active with high heat flow. Recent studies of Paleoarchean mafic rocks (3.5 to 3.4 Ga greenstones) show δ^{41}K down to –0.69 ‰ (50), which is the result of the incorporation of lighter ^{39}K during low-T seafloor hydration (35, 50). The complementary heavier ^{41}K would have remained in the seawater. Given seafloor spreading and perhaps more extensive ridge systems in the Archean, this mechanism could play a key role in Archean seawater chemistry.

In summary, key sinks such as seafloor alteration and clay mineral formation preferentially remove lighter isotopes, thereby buffering seawater to a relatively heavy δ^{41}K. The Eoarchean banded rocks (Isua and Akilia) provide proxies for δ^{41}K in Archean seawater (or mid-ocean ridge hydrothermal fluids). Their relatively heavy δ^{41}K values, similar to those of <2.5 Ga BIFs and modern seawater (Fig. 2*A*), indicate that Archean oceans were likely governed by similar K cycling processes as today. Although the exact value may have fluctuated with changes in flux balance (e.g., during supercontinent emergence or periods of enhanced weathering), the heavy K isotopic composition of seawater appears to have remained relatively stable across geological timescales.

Nutrient Supply and Early Biospheric Implications. K is essential for cellular processes. Early life could have originated in environments with elevated K^+/Na^+ ratios such as evaporative ponds or terrestrial hydrothermal systems, rather than the Na-dominated Archean ocean (51). Nevertheless, as continental weathering intensified, rivers likely provided a continuous flux of K to the oceans (52). Our data support the presence of effective continental weathering and material transfer into the ocean during the early Archean. Previous studies of 2.7 Ga BIFs demonstrated that much of their iron originated from continental rocks and was delivered through microbial cycling on continental shelves (53). Similarly, other nutrients including K likely became integrated into early microbial ecosystems in nearshore settings. Recent investigations of 2.1 Ga black shales in the Francevillian Basin (Gabon) found that microbes concentrated seawater-derived K^+ within their cells and biofilms; upon microbial death and early diagenesis, this biologically sequestered K was released into pore waters, creating localized supersaturation (40). This process effectively transferred seawater K into authigenic illite,

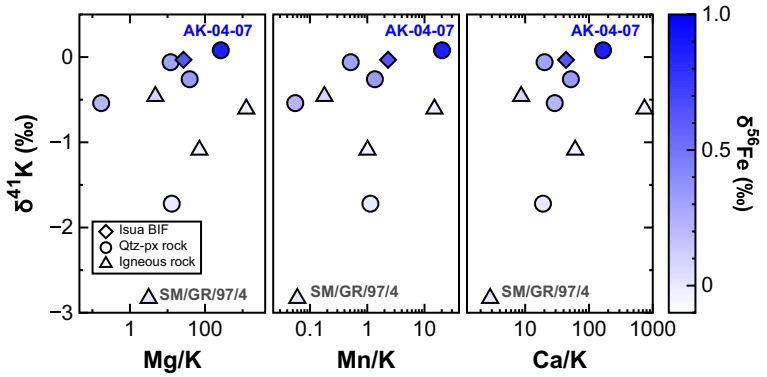

Fig. 4. Relationship between $\delta^{41}K$ values and major element ratios (Mg/K, Mn/K, Ca/K) for investigated samples. The color ramp on the right indicates $\delta^{56}Fe$ values, with varying color intensity corresponding to $\delta^{56}Fe$ in each sample. Element content and Fe isotopic data are sourced from (6, 10).

substantially increasing bulk-rock K_2O concentrations. Such biotic diagenesis suggests that Precambrian microbes could readily trap seawater K in sediments, likely incorporating lighter K isotopes into K-rich clay minerals. This mechanism explains why some black shales possess substantially more K than can be accounted for by detrital inputs alone while also implying an early large sink of ^{39}K, consistent with our isotope observations.

Samples and Methods. We analyzed a total of 30 samples, including Akilia qtz-px rocks (n = 5) and regional igneous rocks (n = 4), BIFs (n = 17), and black shales (n = 4). The BIFs were collected from major global deposits spanning from 3.8 Ga to 1.5 Ga. The 3.45 Ga black shales (oldest) are from the Pilbara Craton and the 3.25 Ga black shale is from the Kaapvaal Craton. Further sample formation is provided in the *SI Appendix*. At Akilia, the qtz-px rock occurs as a ~5 m-wide band surrounded by mafic amphibolite gneisses and ultramafic rocks (*SI Appendix*, Fig. S1). We collected five samples from the center and contact zone of the qtz-px rock. It is noted that the Akilia qtz-px rock sample AK-04-07 and the Isua BIF sample IS-04-06 were collected from the fine-grained part previously identified as least affected by adjacent igneous rocks (6, 10). For qtz-px rock, the primary mineral assemblage comprises quartz, grunerite, amphibole, clinopyroxene, orthopyroxene, and magnetite (3, 10). Among these, amphibole is the primary host of K (*SI Appendix*, Fig. S2).

Potassium isotopic measurements were conducted at the Institut de Physique du Globe de Paris (IPGP). Approximately 15 mg of powdered sample was weighed and dissolved in a 3:1 mixture of concentrated HF-HNO_3 acid for 48 h at 120 °C. After evaporating to dryness at 90 °C, samples were redissolved in a 3:1 mixture of concentrated HCl-HNO_3 acid for another 48 h at 120 °C to eliminate residual fluorides. Subsequently, solutions were again dried and refluxed with concentrated HNO_3. Finally, the prepared sample solutions were loaded onto Savillex PFA microcolumns filled with precleaned AG50W-X12 resin (Bio-Rad, 200 to 400 mesh). More details on sample preparation, chemical purification, and analytical procedures were described in the literature (54). Potassium isotopic fractionation is expressed relative to the NIST SRM 3141a standard using the $\delta^{41}K$ notation: $\delta^{41}K$ (‰) = $[(^{41}K/^{39}K_{sample}/\ ^{41}K/^{39}K)_{standard} - 1] \times 1{,}000$. Terrestrial standards AGV-2, BHVO-2, and BCR-2 yield $\delta^{41}K$ values of–0.46 ± 0.05 ‰ (2SD; n = 5), –0.43 ± 0.04 ‰ (2SD; n = 5), and –0.45 ± 0.06 ‰ (2SD; n = 6), respectively, consistent with published results (54). The analytical results are presented in *SI Appendix*, Table S1.

Data, Materials, and Software Availability. All study data are included in the *SI Appendix*.

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