Magmatic degassing as the primary source of salt in Archean oceans

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Abstract

The salinification of Earth's early oceans impacted both the climate and the evolution of life. However, available halogen data of Archean seawater samples are at apparent odds with a conventionally assumed mantle origin of sea salt, highlighting a critical lack of mechanistic understanding of how the Archean oceans became salty. Here, we present new triple halogen (Cl-Br-I) data from high temperature (~750–840 °C) fluid inclusions from a gabbroic intrusion in Iceland. Our data show that the magmatic degassing fractionates the halogens and generates fluids with high Br/Cl and I/Cl that match with Archean seawater. These observations suggest that seawater salinity in the Archean oceans was primarily regulated via fluids exsolved from cooling intrusions during genesis of early crust, and that the mantle had a modern-like halogen composition by 3.5 Ga. Our findings highlight the strong connection between the chemistry of the mantle, magmatic fluids and the oceans, with implications for understanding the chemical environments that guided the evolution of early marine organisms.

Introduction

Oceans of the Archean eon (4.0–2.5 Ga) harboured Earth's earliest known organisms (Knoll et al., 2016). In order to understand the conditions that were conducive to early life, considerable effort has been dedicated to deciphering the chemical makeup of Archean seawater (e.g., Walker, 1983). The origin and abundance of sea salt are of particular interest due to its impact on the climate (Olson et al., 2022) and the solubility of oxygen (Knauth, 1998), and its close link to the evolution of Earth's mantle and crust (Schilling et al., 1978; Sharp and Draper, 2013). In the modern oceans, the dominant influx of solutes stems from riverine transport of continental weathering products, whereas Archean solute fluxes were sourced mainly by magmatism and magmatic-hydrothermal activity (e.g., Kamber and Webb, 2001). Accordingly, ocean salinity in the Archean and Paleoproterozoic has been assumed to be mantle-derived (Schilling et al., 1978; Marty et al., 2018). However, available samples of Archean seawater show higher Br/Cl and I/Cl relative to both the modern and the primitive mantle, challenging this explanation (Fig. 1). The mismatch could be explained by a shift of mantle halogen composition towards lower Br/Cl and I/Cl over time, fractionation of mantle-derived halogens during magmatic processes, or a hitherto unknown Br-I sink in Archean oceans. Testing of these hypotheses is challenging because the halogen composition of the Archean mantle is difficult to constrain (Kendrick, 2024) and the extent of halogen fractionation between melts and magmatic fluids—which distribute mantle halogens to the oceans at submarine hydrothermal systems—remains uncertain (Kendrick et al., 2018).

In addition, magmatic temperature fluids (> 600 °C) have never been sampled *in situ* and fluid inclusion (FI) studies targeting fossil magmatic-hydrothermal high-T fluids have largely focussed on felsic, continental and/or convergent margin settings (e.g., Audétat and Zhang, 2019). Hence, compositional data of magmatic fluids from mafic-dominated crustal environments is limited, with no combined single-FI Cl-Br-I analyses available. This hampers our ability to estimate mantle-derived solute fluxes into the oceans, especially in the Archean, when magmatism was dominantly mafic and submarine (Cawood and Hawkesworth, 2019).

To address this knowledge gap, we report the first triple halogen (Cl-Br-I) data for individual magmatic-temperature FIs in oceanic mafic-dominated crust. The data were collected from the fossil Vesturhorn volcanic complex in East Iceland, which offers an opportunity to investigate the effect of the magmatic fluid formation process on halogens at a modern setting resembling Archean crust.

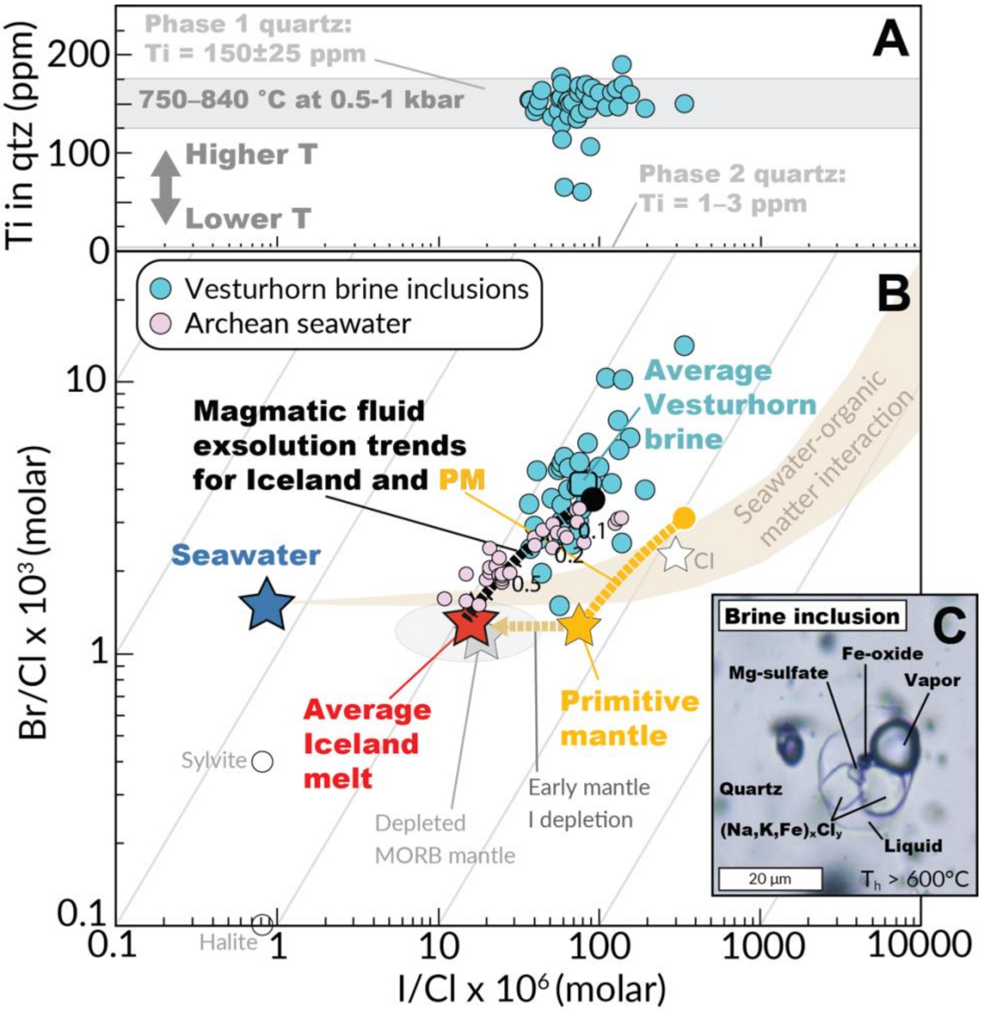

Figure 1. Halogen systematics of magmatic brines. (A) Brine inclusion host quartz Ti contents. Phase 1 quartz has high Ti contents of 150±25 ppm (1σ), indicating formation temperatures of 750–840 °C (Fig. S2). Low Ti contents (<3 ppm) in Phase 2 quartz indicate a lower-T hydrothermal origin. (B) The halogen compositions of Vesturhorn FIs are distinct from Icelandic melts (Waters, 2021) but overlap with Archean seawater (Channer et al., 1997; Burgess et al., 2020). Fluid exsolution trends are modelled assuming closed-system equilibrium fluid-melt partitioning (Tables S3, S4) from melt compositions representing an Iceland average basalt and the primitive mantle. Plotted are compositions of mid-ocean ridge basalts (Kenrick et al., 2017) and their source mantle (Kendrick, 2024), primitive mantle (Kendrick et al., 2017) and CI chondrites (Lodders and Fegley, 2023). The light brown field envelopes modern marine pore water compositions reflecting binary mixing of seawater and high-I/Cl organic matter (Kendrick et al., 2018; Fusswinkel et al., 2022). (C) A typical Vesturhorn brine inclusion with multiple daughter phases.

Samples and results

Here, we report halogen (Cl-Br-I) and trace element laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) data from quartz-hosted FIs. Full methodological details and datasets are provided in the Supplemental Material[11]. The FIs occur in a miarolitic cavity within an exposed gabbroic intrusion belonging to the 3.7–4.3 Ma Vesturhorn volcanic complex (Martin et al., 2011) in East Iceland. Iceland is arguably the best-available modern analogue of Early Earth crustal formation processes due to its high magma production rates and a thickened, oceanic plateau-type crust formed by a hot mantle plume (Reimink et al., 2014). The simple geological context of Vesturhorn minimizes fluid contributions from secondary, post-magmatic events, or external fluid sources (e.g., seawater or sediments) which complicate interpretation of FI data at continental, submarine, or ancient settings.

The investigated miarolitic cavities occur within an aplitic vein that grades into a coarse-grained gabbroic host rock (Fig. S1A). The cavities host two generations of quartz that are distinguished by their optical and cathodoluminescence (CL) colours, inclusion assemblages and Ti contents. The earliest-formed quartz (Phase 1) is represented by optically clear quartz cores with purple-to-violet CL colours, and high Ti contents (150±25 ppm; 1σ). Phase 1 quartz is overgrown by prismatic crystals of later-formed green quartz with actinolite inclusions (Phase 2) that is CL inactive and Ti-poor (1-2 ppm; Fig. 1A, Fig. S1B–D). In this work, we focus on the most common FI assemblage consisting of co-occurring vapor-rich (70–90 % vapor, 10–30 % liquid at 20 °C) and brine FIs (liquid-vapor-multiple solid: Figs. 1C, S1E–F). This assemblage forms inclusion trails that exclusively occur in the interior, actinolite-free Phase 1 quartz and terminate prior to the Phase 2 quartz overgrowth zones (Fig. S1B), and are hence interpreted to be coeval with Phase 1 quartz. High homogenisation temperatures of the fluids (> 600° C) and high Ti concentrations of the host quartz suggests that the fluids were trapped at high temperatures (750–840 °C; Figs. 1A, S2), compatible with a magmatic or near-magmatic formation environment. We interpret the FIs to represent magmatic fluids formed during late-stage crystallization of the host gabbro.

The brine inclusions contain a (Na,K,Fe)Cl-rich aqueous fluid with a salinity of approximately 50 wt.% $NaCl_{eq}$ (Fig. S3). Compared to most magmatic fluids associated with silicic plutons, the brines have high K-Fe contents relative to Na (Fig. S3A) and have distinctly high Fe/Mn and Ba/Mn (Fig. S4A–B). The major cation composition of the fluids was likely buffered by the host rock due to rapid fluid-rock equilibration at low water/rock ratios and high temperatures (Dolejš and Wagner, 2008). The brines are rich in Cu-Zn-Mo-Ag, similar to high-temperature FIs observed in deep parts of the active Reykjanes hydrothermal system in Iceland (Bali et al., 2020) and continental granitic systems (Fig. S3B).

Discussion

Fractionation of halogens during magma degassing

Our new measurements provide the first empirical Cl-Br-I estimates for high-T magmatic fluids in oceanic environments and offer an opportunity to test melt-fluid partitioning behavior of halogens in natural samples. Experimental studies show that Cl, Br and I are strongly hydrophilic and partition from a melt into an exsolved aqueous magmatic fluid phase, with $D_{fluid–melt}$ increasing from Cl to Br to I (Miranda et al., 2025). Hence, fluid exsolution from intrusions is expected to fractionate the halogens, generating magmatic fluids with high Br/Cl, I/Cl and I/Br relative to the source melt. The Cl-Br-I ratios measured in the brine FIs are assumed to closely represent that of the bulk magmatic fluid, since halogens strongly partition into the brine phase during vapor-liquid separation, and

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[1] Supplemental Material (detailed method descriptions, Tables S1–S5, and Figs. S1–S10) for this preprint can be accessed at github.com/eemu.

because Cl-Br-I are not fractionated during vapor-liquid phase separation nor reaction with basaltic host rocks (Kendrick, 2018).

We modelled the I/Cl and Br/Cl composition of magmatic fluids exsolving from Icelandic magmas, using a range of $D_{fluid\text{–}melt}$ calculated after Miranda et al. (2025; Figs. 1B, S8; Tables S3, S4). Magmatic fluids develop higher Br/Cl (up to ~4 × 10^{-3}) and I/Cl (up to ~90 x 10^{-6}) relative to an average Icelandic undegassed melt composition (Br/Cl = 1.4 × 10^{3}, I/Cl = 15 × 10^{-6}; Waters, 2021). The magmatic fluid exsolution trend occupies an almost unpopulated field among published halogen data from geological environments (Fig. 1B). The Vesturhorn brines overlap with this trend, with average molar Br/Cl and I/Cl of 4.1 × 10^{-3} and 73 × 10^{-6}, respectively. This halogen signature, together with high formation temperatures (> 750°C), strongly support that the FIs trap a primary magmatic fluid prior to mixing with external fluids. Because mid-ocean ridge basalts and Icelandic melts have a near-identical halogen composition (Fig. 1B), the Vesturhorn FIs provide an accurate proxy of the halogen signature of magmatic fluids produced by modern ocean floor magmatism. Of particular note is that the elevated I/Cl of the Vesturhorn brines contrasts sharply with the low I/Cl of modern seawater (0.9 × 10^{-6}), precluding seawater contribution, whereas their high Br/Cl is distinct from the seawater-organic matter mixing trend (Figs. 1B, S10). Hence, Cl-Br-I systematics offer a uniquely suited geochemical tool for distinguishing between magmatic, marine organic matter and seawater-derived fluid sources in geological environments in general.

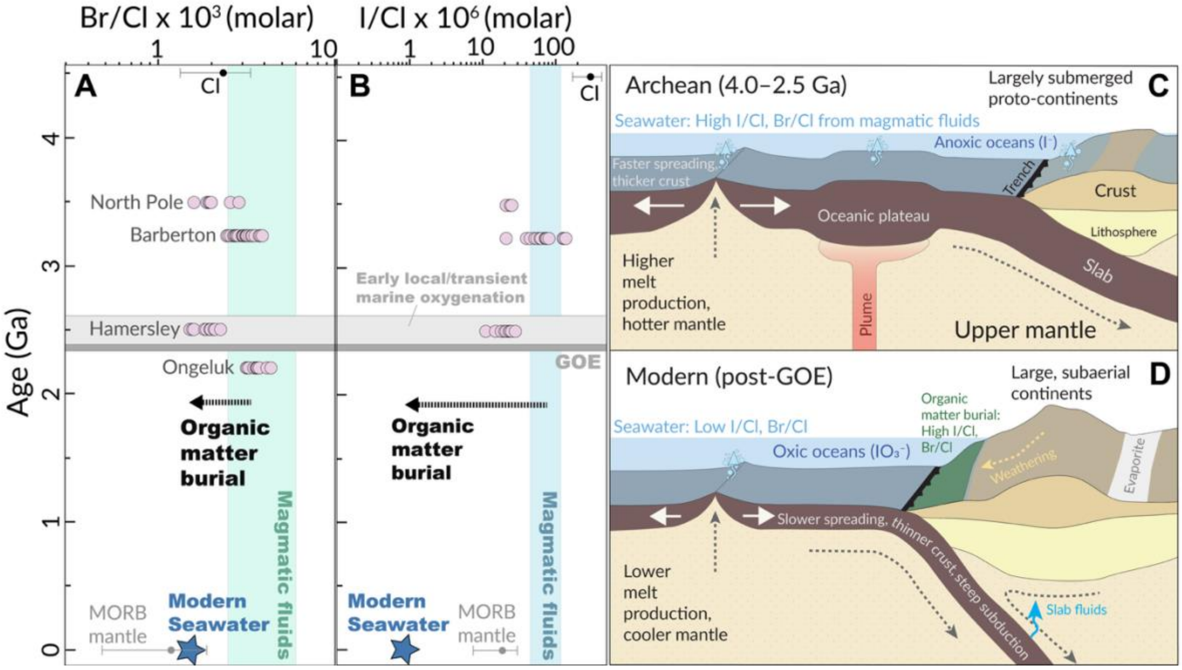

Figure 2. The (A) Br/Cl and (B) I/Cl evolution of Earth's oceans. The two cartoons illustrate the regime shift from (C) an Archean anoxic ocean world, where magmatism is the primary halogen supplier to the oceans, to (D) the modern state, where the oceans' halogen budget is regulated by continental weathering, subduction and organic matter burial. The grey fields indicate the 2.4 Ga Great Oxygenation Event and possible earlier transient/localized oxygenation events (Liang et al., 2025). Published Archean seawater data are plotted from North Pole, Australia (Foriel et al., 2004; Burgess et al., 2020), Barberton, South Africa (Channer et al., 1997), Hamersley Group, Australia (Burgess et al., 2020) and the Ongeluk Formation, South Africa (Gutzmer et al., 2003). The MORB mantle and CI chondrite compositions are from Kendrick (2024) and Lodders and Fegley (2023), respectively.

Halogen composition of Earth's early oceans and mantle

A key observation from our new halogen data is that the Vesturhorn Br/Cl and I/Cl signatures overlap with the halogen signature of Archean seawater (Fig. 1B). This similarity provides compelling independent evidence that the origin of salt in Archean seas was magmatic and sourced from fluids formed during submarine magmatism, for reasons outlined below. The composition of Archean seawater is known indirectly from compositions of sediments that precipitated from seawater (Veizer et al., 1982; Kamber and Webb, 2001) and from FIs thought to preserve Archean seawater or seawater-origin hydrothermal fluids (Channer et al., 1997; Burgess et al., 2020). Ocean salinity appears to have been close to its modern value of 3.5 wt.% NaCl since at least 3.0 Ga (Marty et al., 2018). However, Archean seawater (or hydrothermal fluids of seawater-origin) had higher Br/Cl ($1.5–4.3 \times 10^{-3}$) and I/Cl ($21–140 \times 10^{-6}$) relative to modern seawater (Br/Cl $=1.5 \times 10^{-3}$; I/Cl $= 0.87 \times 10^{-6}$; Figs. 1B,2; Burgess et al., 2020). This change in halogen composition reflects a wider biogeochemical regime shift in the Earth's oceans following the Great Oxygenation Event (GOE) at ~2.4 Ga, marking the large-scale oxygenation of the oceans and diversification of marine organisms (e.g., Holland, 2002), with major consequences for oceanic halogen budgets (Fig. 2). Post-GOE oceans are depleted in I and Br relative to Archean seawater, as marine algae fixate the oxidized iodine species, IO_3^-, and to a lesser degree, Br, in seawater (Hardisty et al., 2014). The lack of biological halogen sinks in pre-GOE oceans (Fusswinkel et al., 2022) means that the observed Archean seawater signatures reflect an inorganic high Br/Cl-I/Cl halogen source.

Continental weathering was limited during the Archean, as the continental masses were submerged by oceans that covered up to 98% of global surface area (Flament et al., 2008). Accordingly, volcanism forming the mafic oceanic crust, oceanic plateaus, as well as the proto-cratons occurred below sea level (Cawood and Hawkesworth, 2019). The low continental freeboard combined with higher rates of magmatism (Herzberg et al., 2010) caused a ~50 times higher magmatic-hydrothermal/continental weathering solute input ratio into the oceans relative to the present (Fig. 2C; Kamber and Webb, 2001). Similarly, the halogen budget in Archean oceans has been assumed to be mantle-buffered (Schilling et al., 1978; Burgess et al., 2020), although the Br/Cl and I/Cl of Archean seawater do not match with either the modern upper mantle nor estimated primitive mantle compositions (Fig. 1B).

Our data demonstrate that this mismatch could be caused by fractionation of the halogens during exsolution of magmatic fluids from a mantle-derived source melt. All published Archean seawater halogen compositions fall close to the modelled magmatic fluid exsolution trend of Icelandic magmas, representative of the modern mantle, whereas no data overlap with the primitive mantle fluid exsolution trend (Fig. 1B). We propose that this overlap reflects an Archean mantle that had a similar halogen composition to the modern, i.e., depleted in I relative to Cl compared to the primitive mantle. Our findings are compatible with the halogen mantle evolution model of Kendrick et al. (2020) suggesting that Earth's mantle may have reached modern mantle-like I/Cl values by 3.5 Ga—the age of the oldest Archean seawater samples (Fig. 2)—due to early subduction of serpentinized lithosphere. Alternatively, mantle with a subchondritic I/Cl could be produced by preferential partitioning of I to the core during core-mantle differentiation (Jackson et al., 2018) or collisional erosion of a Hadean halogen-rich surface reservoir during Earth's accretion (Sharp and Draper, 2013).

Regardless of the mechanism that caused early I depletion of the mantle, our observations suggest that the halogen input to the Archean seas was dominantly magmatic and channeled via magmatic fluids emanating from cooling intrusions in the Archean submarine crust (Fig. 2C). Magmatic fluid fluxes from intrusions to the oceans could be substantial, as intrusive magmatism contributes ~4–10 times more mass to the oceanic crust than extrusive volcanism (White et al., 2006). A simple mass balance model shows that the flux of magmatic fluids to the Archean oceans may have been sufficient to salinate the oceans to 0.5–1x modern seawater salinity within < 1.5 Ga (Fig. S9). Notably, magmatic fluids with similar halogen signatures may have formed both in Archaean mafic-dominated submarine crust as well as in submerged plateau-type proto-cratons.

This work demonstrates that the halogen composition of Earth's early oceans provide independent constraints on the timing and mechanisms of the contemporaneous evolution of Earth's mantle and crust. This insight should spark efforts to improve our currently patchy temporal resolution of Archean and Proterozoic ocean halogen compositions (Fig. 2A–B). The implied high volumes of magmatic fluid inputs to Archean oceans has implications for understanding the chemical environments near Archean submarine hydrothermal vents. Our data show that aqueous environments with high K/Na and high Zn and Mn concentrations—essential nutrients for protocells—are not exclusive to inland geothermal systems (Mulkidjanian et al., 2012) but could also form near submarine vents with high magmatic fluid influx.

Conclusions

This study provides a new baseline composition for magmatic fluids produced during the genesis of oceanic crust. The results have implications for the chemistry of hydrothermal vents on the Archean seafloor, where magmatic fluid fluxes may have been higher than in modern systems. The data confirm experimental evidence that Cl-Br-I fractionate during fluid exsolution from a silicate melt, resulting in a characteristic high Br/Cl and I/Cl magmatic fluid halogen signature that overlaps with Archean seawater. Together, these observations reconcile the mismatch between mantle and seawater halogen signatures, providing compelling evidence that the salinity in the Archean oceans had a dominantly magmatic origin and was sourced from a mantle that had a similar halogen composition to the modern by 3.5 Ga.

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