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Resolving the chlorine isotope composition of Earth's depleted mantle

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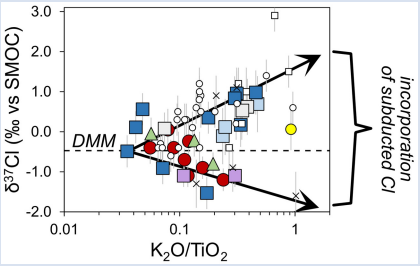
Abstract



The chlorine isotope ratio (δ^{37}Cl value) of Earth's mantle has implications for volatile exchange between Earth's surface and mantle, as well as for the source of volatile delivery to Earth. However, there is disagreement about this value, with estimates ranging from −3 ‰ to +0.9 ‰. To resolve this, we examine the δ^{37}Cl values of mid-ocean ridge basalt (MORB) glasses from several ridge segments. We find that the δ^{37}Cl value of the depleted MORB-source mantle (DMM) is ~ −0.5 ‰, and deviation from that value results from incorporation of subducted material in the DMM. MORB samples that have shallowly assimilated Cl extend towards δ^{37}Cl values of −0.6 ‰ to −1.5 ‰, suggesting assimilation of hydrothermal brines will not result in seawater-like δ^{37}Cl values (0 ‰). The calculated Bulk Silicate Earth δ^{37}Cl value is −0.04 ± 0.13 ‰, statistically indistinguishable from chondrites. This similarity suggests that >91–99 % of Earth's Cl was inherited from chondrites with little contribution from ingassing of the solar nebula.

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Introduction

The chlorine isotope ratio (δ^{37}Cl value) of Earth's mantle has important implications for both the extent of volatile exchange between Earth's surface and mantle and the source of Earth's volatiles during early accretion. The δ^{37}Cl values of Earth's major surface reservoirs (*e.g.*, hydrosphere, evaporites, sediments) have ranges that are largely agreed upon (*e.g.*, Barnes and Sharp, 2017), but the δ^{37}Cl value of Earth's mantle is more contentious. Previous studies have examined the δ^{37}Cl values of mid-ocean ridge basalt (MORB) glasses because Cl isotopes do not significantly fractionate at high temperatures (*e.g.*, Balan *et al.*, 2019) during partial melting or fractional crystallisation, so MORB samples should preserve the δ^{37}Cl values of their mantle source if there is minimal shallow assimilation. Studies of MORB glasses and other mantle-derived samples have come to contrasting conclusions about the δ^{37}Cl values of the depleted MORB-source mantle (DMM), with recent estimates ranging from −3 ‰ to +0.9 ‰ (Sharp *et al.*, 2007, 2013; Bonifacie *et al.*, 2008; Layne *et al.*, 2009; Pinti *et al.*, 2020). These DMM estimates span a large portion of the natural δ^{37}Cl range of predominant Cl hosts on Earth's surface (−2 ‰ to +2 ‰; Barnes and Sharp, 2017). Each estimate has differing implications for the cosmochemical source of Earth's volatile elements and the extent of volatile exchange between Earth's mantle and surface. It is therefore imperative to better constrain the δ^{37}Cl composition of Earth's mantle. To do that, we analysed the δ^{37}Cl values of MORB glasses spanning a range of trace element and radiogenic isotope compositions. We combine our new data with previously published MORB glass δ^{37}Cl values to examine the effects of shallow assimilation and mantle heterogeneity and more tightly constrain the δ^{37}Cl value of Earth's mantle.

Samples and Geologic Context

Samples analysed in this study are MORB glasses from the Mid-Atlantic Ridge (MAR); the East Pacific Rise (EPR); the Galapagos Spreading Centre (GSC); the Southwest Indian Ridge (SWIR); and the Central Indian Ridge (CIR) (Fig. S-3a). These localities range from ultrafast to ultraslow spreading centres. Samples were dredged from depths of 1305–5150 metres below sea level. Selected samples have MgO contents of 5.6–10.4 wt. % and have trace element (*e.g.*, K_2O/TiO_2, Nb/Zr, Th/La) and radiogenic isotope (*e.g.*, $^{143}Nd/^{144}Nd$) compositions (Fig. S-3b) that include D-MORB, N-MORB, and E-MORB. Samples also span a wide range of Cl/K (0.010–0.819; Fig. S-3b). Details of sample locations and compositions are provided in Table S-3. Measurements of δ^{37}Cl values of pristine MORB glasses were conducted *via* isotope ratio mass spectrometry at the University of Texas at Austin and *via* secondary ionisation mass spectrometry at the University of Lausanne, Switzerland. For details of preparation procedures and analytical methods see the Supplementary Information S-1 and S-2.

Results

Chlorine isotope ratios of MORB glasses measured in this study range from −1.5 ‰ to +1.0 ‰, slightly larger than the range of

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previous studies (−1.9 ‰ to +0.4 ‰; Sharp *et al.*, 2007; Bonifacie *et al.*, 2008). After integrating MORB $\delta^{37}Cl$ data from previous studies filtered for potential analytical artifacts (see Supplementary Information S-3), $\delta^{37}Cl$ values do not correlate with indices of seawater/brine assimilation such as Cl/K when all data are considered together (Fig. 1). The most depleted sample from this study (RC2806 1D-1; $^{143}Nd/^{144}Nd = 0.513234$, $^{87}Sr/^{86}Sr = 0.702125$; Le Voyer *et al.*, 2015) has a $\delta^{37}Cl$ value of −0.5 ‰. For all localities, samples with higher K_2O/TiO_2 and lower $^{143}Nd/^{144}Nd$ extend to both positive and negative $\delta^{37}Cl$ values (Fig. 2). See Table S-3 for all collected and compiled $\delta^{37}Cl$ values and other geochemical data used in this study.

Discussion

Effects of seawater/brine assimilation. Shallow assimilation of seawater or brine has been shown to significantly increase the Cl content of MORB glasses (*e.g.*, Michael and Schilling, 1989). Previous studies have used Cl/K as a filter for seawater and high

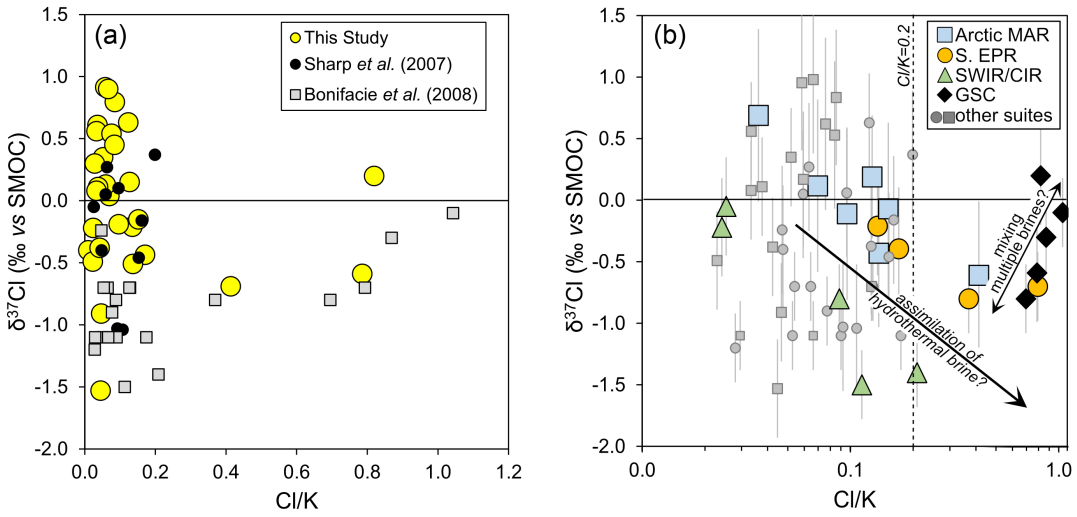

Figure 1 Cl/K plotted against $\delta^{37}Cl$ values of MORB glasses from this study and literature. Data are grouped by **(a)** the study that measured them and **(b)** by locality. **(a)** There is no clear consistent correlation between Cl/K and $\delta^{37}Cl$ values. **(b)** Only suites that contain one or more samples with Cl/K > 0.2 are denoted by coloured symbols. All other suites in which all measured samples have Cl/K < 0.2 are coloured in grey. Note that x-axis (Cl/K) in **(b)** is log scale to more easily discern trends between Cl/K and $\delta^{37}Cl$ values. Error bars are 2 s.d. References for the Cl and K concentrations are given in Table S-3.

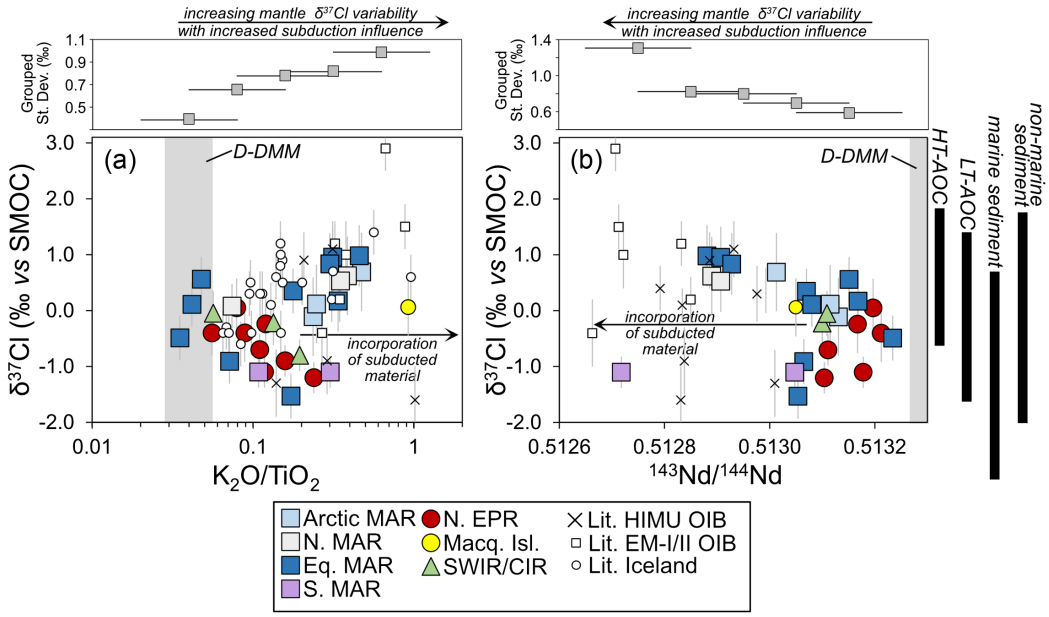

Figure 2 Chlorine isotope ratios of MORB samples from this study and MORB and OIB samples from literature filtered for Cl contamination plotted against **(a)** K_2O/TiO_2 and **(b)** $^{143}Nd/^{144}Nd$. Error bars are 2 s.d. Large coloured symbols are MORB samples from this study, Sharp *et al.*, (2007) and Bonifacie *et al.* (2008). Literature EM-I/II and HIMU OIB data are from John *et al.* (2010). Literature Iceland data are from Halldórsson *et al.* (2016). Grey vertical bars are the most depleted end member of DMM (D-DMM) K_2O/TiO_2 and $^{143}Nd/^{144}Nd$ values based Workman and Hart (2005) and Shimizu *et al.* (2016). The black bars to the right show the range of $\delta^{37}Cl$ values in subducted materials (HT-AOC = high temperature altered oceanic crust, LT-AOC = low temperature altered oceanic crust; from Barnes and Sharp, 2017 and references therein). Grouped St. Dev. = grouped standard deviations, which shows the standard deviation in $\delta^{37}Cl$ values of a subset of MORB and OIB samples for a range of K_2O/TiO_2 (left) or $^{143}Nd/^{144}Nd$ (right) shown by the horizontal black lines. References for K_2O/TiO_2 and $^{143}Nd/^{144}Nd$ are given in Table S-3.

salinity brine assimilation (*e.g.*, Michael and Cornell, 1998; Le Roux *et al.*, 2006). Although Michael and Cornell (1998) proposed that any sample with Cl/K > 0.08 has been affected by brine assimilation, subsequent studies have shown higher Cl/K values up to 0.09–0.14 can be primary mantle signatures (Shimizu *et al.*, 2016; Kendrick *et al.*, 2017). Samples with Cl/K in this range may contain primary or assimilated Cl. In this study, we filter samples based on Cl/K thresholds from Shimizu *et al.* (2016) (see Supplementary Information S-3). Filtered samples may contain assimilated Cl and so are not considered in the discussion of mantle δ^{37}Cl values.

Bonifacie *et al.* (2008) interpreted correlations between MORB glass δ^{37}Cl values and both Cl concentration and Cl/K as the result of seawater/brine assimilation driving MORB towards seawater-like δ^{37}Cl values (~0 ‰) at higher Cl/K. However, when data from this study and filtered literature (see Supplementary Information S-3) are considered together, there is no clear correlation between δ^{37}Cl and Cl/K (Fig. 1a). When samples are considered by locality, samples with unambiguous signs of brine assimilation (Cl/K > 0.2) tend to have lower δ^{37}Cl values than other samples from the same locality, extending to δ^{37}Cl values lower than −0.6 ‰ to −1.5 ‰ (Fig. 1b). This indicates that shallow assimilation does not drive MORB glasses to seawater-like δ^{37}Cl values of 0.0 ‰, as suggested by Bonifacie *et al.* (2008), but rather towards δ^{37}Cl values lower than seawater. An exception to this is the GSC suite because the samples with the highest Cl/K do extend towards seawater-like δ^{37}Cl values (Fig. 1b). However, all GSC samples measured in this study and by Bonifacie *et al.* (2008) have Cl/K of 0.70–1.04, and so have likely all assimilated significant Cl. The generally negative δ^{37}Cl value of assimilated components indicates that assimilation of seawater-derived Cl from a high salinity brine or hydrothermal fluid will not necessarily result in seawater-like δ^{37}Cl values in MORB, as has been previously suggested.

Variations in MORB δ^{37}Cl values related to mantle heterogeneity. Previous studies of MORB δ^{37}Cl values assumed that the DMM source of MORB was largely a single reservoir (Sharp *et al.*, 2007) or that it was divided into an E-MORB and N-MORB source (Bonifacie *et al.*, 2008). However, the mantle is not composed of just one or two reservoirs, but a large range of chemically heterogenous materials including at least three end members (EM-I, EM-II, HIMU) in addition to DMM (*e.g.*, Stracke, 2012). These end members likely result from incorporation of subducted oceanic crust and sediment into the mantle source of basalts, resulting in higher K_2O/TiO_2 (*e.g.*, Jackson and Dasgupta, 2008) and lower $^{143}Nd/^{144}Nd$ (*e.g.*, White and Hofmann, 1982). Although these components are more abundant in ocean island basalts (OIBs), they can also be introduced at mid-ocean ridges either from interaction with a mantle plume (*e.g.*, Le Voyer *et al.*, 2015) or from distributed components in the upper mantle (*e.g.*, Castillo *et al.*, 1998). Some of the disagreement on the DMM δ^{37}Cl values may arise from incorporation of these subduction-derived enriched components in some of the previously analysed samples. The possibility of MORB Cl isotope heterogeneity was raised by both Sharp *et al.* (2007) and Bonifacie *et al.* (2008) but was never explored.

When MORB data filtered for shallow assimilation from this study and previous studies are combined, the most depleted samples (lowest K_2O/TiO_2; highest $^{143}Nd/^{144}Nd$) from several localities have δ^{37}Cl values of −0.2 ‰ to −0.5 ‰, and other MORB samples diverge towards lower and higher δ^{37}Cl values at greater degrees of enrichment (higher K_2O/TiO_2, lower $^{143}Nd/^{144}Nd$; Fig. 2). Previously published OIB data (John *et al.*, 2010; Halldórsson *et al.*, 2016) also follow these general trends (Fig. 2). We calculated the standard deviation in δ^{37}Cl values of subsets of MORB and OIB samples grouped by increasing K_2O/TiO_2 and $^{143}Nd/^{144}Nd$ ranges (see Fig. 2a,b). The standard deviation of the sample subsets increases with increasing subduction influence (Fig. 2a,b), consistent with addition of subducted altered oceanic crust and sediment with heterogenous δ^{37}Cl values (−3.0 to +3.0 ‰; Barnes and Sharp, 2017 and references therein; Fig. 2) into the DMM. The δ^{37}Cl values higher or lower than DMM are not uniquely associated with specific mantle components. Samples with HIMU/FOZO influence (Fig. S-3b) extend to both higher and lower δ^{37}Cl values (Fig. S-4a), as do samples influenced by EM-I/EM-II components (Figs. S-3b, S-4b). Although the addition of subducted crust and sediment in the upper mantle influences MORB δ^{37}Cl values, there is currently no clear differentiation between the δ^{37}Cl values of different subducted materials.

To quantify the relationship between changes in δ^{37}Cl values and incorporation of subducted components, we calculate the absolute deviation from the DMM end member δ^{37}Cl value for each sample. Here the DMM δ^{37}Cl value selected is −0.5 ‰ based on the value of the most depleted sample (RC2806 1D-1). We then compare these absolute deviations to the $D^{Pb/Nd/Sr}$ values for each sample (method modified from Jackson *et al.*, 2020; Supplementary Information S-4). The $D^{Pb/Nd/Sr}$ value describes the distance in three dimensional radiogenic isotope space that a sample is from the DMM end member. As the samples extend farther from the DMM component in radiogenic isotope space (higher $D^{Pb/Nd/Sr}$), the δ^{37}Cl values also deviate more strongly from the DMM value (Fig. 3). This indicates much of the observed variability in the MORB δ^{37}Cl values results from

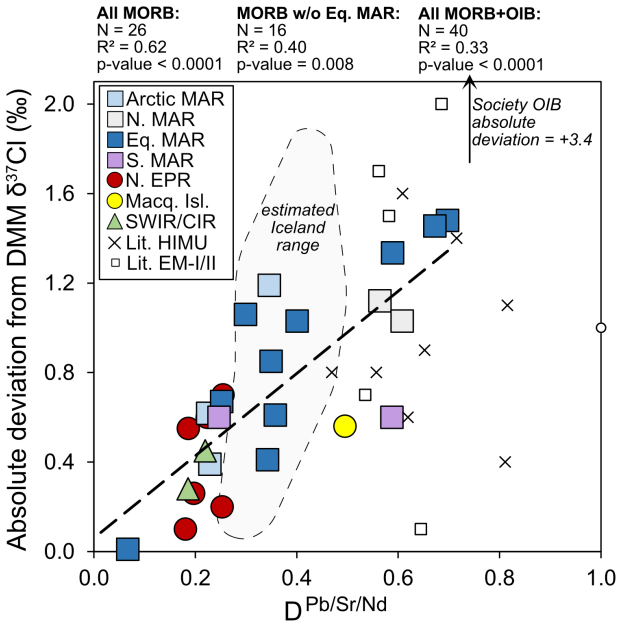

Figure 3 Absolute deviation from the DMM δ^{37}Cl value (−0.5 ‰; see main text) plotted against $D^{Pb/Sr/Nd}$ for MORB glasses filtered for seawater/brine assimilation. Large coloured symbols are MORB samples from this study, Sharp *et al.* (2007), and Bonifacie *et al.* (2008). The dashed black line is a linear regression through all MORB samples. The R^2 value and non-directional p-value labelled "All MORB" include all MORB samples, whereas "MORB w/o Eq. MAR" excludes the Equatorial MAR from the linear regression, and "All MORB + OIB" includes all MORB and previously published OIB data. Literature OIB data from John *et al.* (2010). "Estimated Iceland range" indicates that the $D^{Pb/Sr/Nd}$ values are not from measured $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ of these glasses but based on Icelandic basalts from the GEOROC database. One OIB sample from Society Islands extends beyond the range of this plot (absolute deviation from DMM = +3.4 ‰). References for radiogenic isotopes are given in Table S-3.

incorporation of subduction-derived components with heterogeneous δ^{37}Cl values into an ambient DMM reservoir with a δ^{37}Cl value of ~ −0.5 ‰. Although this trend is largely defined by the HIMU-influenced Equatorial MAR samples (Fig. 3), the observed correlation is still statistically significant if the Equatorial MAR samples are not considered (non-directional p-value = 0.008). When including OIB data from John *et al.* (2010), the correlation remains significant (non-directional p-value < 0.0001). Therefore, after accounting for shallow assimilation, mantle heterogeneity, and potential analytical artifacts in previous studies (see Supplementary Information S-3), contradictory estimates of the DMM δ^{37}Cl value from studies in the past two decades can largely be reconciled.

Estimate of the Bulk Silicate Earth δ^{37}Cl value and similarity to chondrites. Previous studies have suggested multiple sources of volatile elements to Earth, including nebular ingassing, comet addition, chondrite addition, or a mixture of these three (*e.g.*, Sharp, 2017). The δ^{37}Cl value of the Bulk Silicate Earth (BSE) can shed light on the source of Earth's volatile elements. We calculated the BSE δ^{37}Cl value by adding all surface material into the processed mantle with their respective Cl concentrations and δ^{37}Cl values. The methods and parameters for this procedure are given in the Supplementary Information S-5. Because seawater and evaporites both have near-zero δ^{37}Cl value and account for a combined 44 ± 6 % of the BSE Cl (Table S-2), whereas the processed mantle contains only 9 ± 2 % (Table S-2), the BSE δ^{37}Cl value must be close to 0 ‰. Indeed, we calculated that the BSE δ^{37}Cl value is −0.04 ± 0.13 ‰ (2 s.e.), statistically indistinguishable from bulk δ^{37}Cl values of all chondrites considered together (−0.12 ± 0.26 ‰; 2 s.e.), or carbonaceous (−0.2 ± 0.4 ‰; 2 s.e.), ordinary (−0.4 ±0. 6‰; 2 s.e.), or enstatite (+0.2 ± 0.4 ‰; 2 s.e.) chondrites considered individually (Sharp *et al.*, 2013). In contrast, the BSE δ^{37}Cl value is dissimilar to δ^{37}Cl estimates of the solar nebula (−7 ‰; Gargano and Sharp, 2019).

Previous authors have noted the general similarity between the δ^{37}Cl values of Earth's various reservoirs and chondrites and interpreted that as indicative of a chondritic source of Cl on Earth (Sharp *et al.*, 2013). However, later studies suggested the δ^{37}Cl value of the proto-Earth was originally similar to the solar nebula (−7 ‰; Gargano and Sharp, 2019) and obtained its current δ^{37}Cl value through kinetic fractionation associated with volatile loss during degassing or impact erosion (Sharp *et al.*, 2016; Gargano and Sharp, 2019). Other studies suggest the δ^{37}Cl values of chondrites are derived through equilibrium isotope fractionation of nebular gases at low temperature (Sharp *et al.*, 2013, 2016). These studies imply that the δ^{37}Cl values of Earth and chondrites are not genetically related and any similarity between the two is coincidental. The total range of δ^{37}Cl values on differentiated bodies that have undergone extensive Cl loss spans >46 ‰ (Sarafian *et al.*, 2017; Gargano and Sharp., 2019; Barrett *et al.*, 2019). Given this large range, it appears unlikely that varying degrees of planetesimal degassing, impact erosion, and/or subsequent ingassing would result in nearly identical δ^{37}Cl values in the BSE and in distinct classes of chondrites. We do not argue that such a mechanism is impossible, but rather that a more parsimonious model is that Earth's chondrite-like average δ^{37}Cl value reflects Cl derivation from a chondrite source.

The chondrite-like BSE δ^{37}Cl value requires that little to no Cl was added to Earth from ingassing of the solar nebula. Previous studies have suggested ingassing of the solar nebula into a terrestrial magma ocean as a potential major source of volatiles to the early Earth (*e.g.*, Sharp 2017; Olson and Sharp, 2019). Such incorporation would also impact Cl, which existed as HCl gas in the proto-solar nebula (Fegley *et al.*, 2020). Given the chondritic δ^{37}Cl value of Earth, a maximum of 1–9 % of Earth's Cl can be derived from ingassing of the solar nebula. Chlorine abundance in the early solar nebula was 10–1000 times lower than C, N, Ne, and Ar (Lodders *et al.*, 2009), but HCl is 100–100,000 times more soluble than CH_4, CO_2, N_2, Ne, and Ar in silicate melts at high temperature/low pressure conditions (Fegley *et al.*, 2020 and references therein). Therefore, ingassing of these volatiles will be similar to or lower than for Cl, so Earth's low nebular Cl fraction requires relatively little ingassing of these other volatiles as well. This is consistent with the estimate made by Marty (2012) that ≤10 % of Earth's volatile elements are derived from nebular ingassing based on C, N, H, and noble gas isotope compositions. Therefore, the estimates of Earth's δ^{37}Cl value as well as other volatile element isotope ratios are consistent with minimal nebular ingassing contribution to Earth's volatile content.

Conclusions

This study examines δ^{37}Cl values of MORB glasses to better constrain the δ^{37}Cl value of Earth's mantle and the source of volatiles on Earth. The unmodified DMM has a δ^{37}Cl value ≈ −0.5 ‰, and deviations from that value are either due to incorporation of subducted material with heterogeneous δ^{37}Cl values or assimilation of seawater/brine. In the context of a heterogeneous mantle, the previous disparate mantle δ^{37}Cl estimates are not contradictory, but rather sample different subduction-related components in the upper mantle. Shallow assimilation of hydrothermal brines results in MORB δ^{37}Cl values that extend lower than −0.6 ‰ to −1.5 ‰ and does not necessarily result in seawater-like δ^{37}Cl values. We calculate that the BSE has a δ^{37}Cl value ≈ −0.04 ± 0.13 ‰, consistent with a chondritic source of Cl on Earth with a maximum of 1–9 % contribution from early ingassing of the solar nebula.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2526.

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