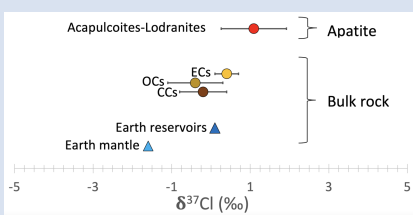# Chondritic chlorine isotope composition of acapulcoites and lodranites

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Abstract

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Bulk rock chondrites and Earth's reservoirs share a common chlorine isotopic value, while more differentiated bodies such as the Moon or Vesta record significant chlorine isotopic fractionation in their Ca phosphates. As such, an important but scarcely studied parameter is the effect of melting and differentiation processes on chlorine concentration and isotopic composition of a planetesimal. Here we report chlorine abundances and isotopic compositions for apatite in a range of primitive achondrites, acapulcoites and lodranites. These meteorites originated from a parent body that experienced some partial melting, allowing an assessment of chlorine behaviour during the early stages of planetary evolution in the inner Solar System. Overall, while bulk rock estimates of F and Cl abundances are indicative of degassing during the early stages of partial melting, no chlorine isotopic fractionation is recorded in apatite. Consequently, acapulcoites and lodranites retain their chondritic precursor isotopic signature for chlorine.

Introduction

Volatile elements are tracers of degassing processes experienced by planetesimals during thermal metamorphism and differentiation in the inner Solar System. Chlorine (Cl) is by far the most abundant halogen in chondritic meteorites, in the range of hundreds of $\mu g.g^{-1}$ in bulk samples (Brearley and Jones, 2018; Lodders and Fegley, 2023), while being depleted in the Earth compared to chondritic abundances (Dreibus et al., 1979). Bulk chlorine isotope analyses of terrestrial mantle-derived samples originating from various mid-ocean ridges, together with sediments sampling the crust, have led to the observation that no isotopic fractionation of Cl occurred during the differentiation, and subsequent volatile loss, of the Earth (Sharp et al., 2013). In addition, the bulk chlorine isotopic composition of enstatite (ECs), ordinary (OCs) and carbonaceous (CCs) chondrites are similar to those of terrestrial reservoirs, which also led to the inference of a chondritic origin for Earth's chlorine and the initial suggestion of a homogenous chlorine isotopic composition of the nebula, with a $\delta^{37}Cl$ value estimated at -0.3 ± 0.3 ‰ (Sharp et al., 2013).

However, measurements of low chlorine isotopic compositions in a variety of extraterrestrial samples have challenged the chondritic/terrestrial $\delta^{37}Cl$ value to be representative of the nebula's primitive chlorine isotopic composition. Indeed, significant chlorine isotopic variations between the terrestrial mantle and surface reservoirs have been highlighted, the former recording a lower $\delta^{37}Cl$ value, from -1.6 to -4.0 ‰ (Bonifacie et al.,

2008; Layne et al., 2009). The hypothesis of a low chlorine isotopic composition for the nebula has been further strengthened with even lower $\delta^{37}Cl$ values measured in a variety of meteoritic samples, down to -6 ‰ for Mars (Shearer et al., 2018), -3.8 ‰ for eucrites (Sarafian et al., 2017; Barrett et al., 2019), -4.7 ‰ for the OC Parnallee (Sarafian et al., 2017), and even down to -7.2 ‰ for iron meteorites (Gargano and Sharp, 2019). As a result, it is now suggested that the primitive chlorine composition of the nebula should have been close to -7.2 ‰; the higher $\delta^{37}Cl$ values observed denoting a later incorporation of ^{37}Cl-enriched HCl hydrates into accreting material in the case of chondrites or degassing processes for larger bodies (e.g., Sharp et al., 2013), although interaction with an HCl-rich ice impactor could have happened on differentiated bodies (Tartèse et al., 2019).

The main carriers of chlorine in most meteorites are the three minerals belonging to the apatite group, namely hydroxyapatite, chlorapatite and fluorapatite, represented by the general formula $Ca_5(PO_4)_3(F,Cl,OH)$ (McCubbin and Jones, 2015). Therefore, in the following the term apatite will be used to refer generically to the three mineral species. Apatite is a volatile bearing Ca phosphate widespread in extraterrestrial samples and, as such, is key to investigate the distribution of volatile reservoir(s), in particular hydrogen and chlorine, in the inner Solar System, and to infer characteristics of the parent body from which they derive such as volatile depletion or differentiation (McCubbin et al., 2023). In situ chlorine isotopic measurements in apatite by secondary ion mass spectrometry (SIMS) tend to show some

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significant variability compared to bulk analyses performed either by gas source isotope ratio mass spectrometry (IRMS) or thermal ionisation mass spectrometry (TIMS) (Sharp *et al.,* 2013; Gargano *et al.,* 2020). The large ranges of $\delta^{37}Cl$ values measured in lunar and eucrite apatite have been interpreted as isotopic fractionation following metal chloride degassing during planetary differentiation (*e.g.*, Sharp *et al.,* 2010; Barnes *et al.,* 2019; Barrett *et al.,* 2019) and, in the case of the Moon, may not be representative of bulk rock $\delta^{37}Cl$ value (Gargano *et al.,* 2020). As such, effects of differentiation processes on chlorine isotopic composition in apatite, such as metamorphism and partial melting, have yet to be investigated.

In order to comprehend the processes responsible for $\delta^{37}Cl$ fractionation in apatite and the potential effect of parent body processes, in particular partial melting, we have investigated the chlorine abundances and isotopic compositions in phosphates from primitive achondrites, acapulcoites and lodranites. Acapulcoites and lodranites derive from a single partially differentiated parent body, and the chemical composition of its chondritic precursor lies between ordinary and enstatite chondrites (*e.g.*, Keil and McCoy, 2018; and references therein). These samples are strategic targets as they recorded a range in the degrees of planetary differentiation, from 1 % of partial melting for the less metamorphosed acapulcoites, some of which still retain relict chondrules, up to 20 % partial melting for the most differentiated lodranites, which have suffered from melt loss as evidenced by the depletion in plagioclases (*e.g.*, McCoy *et al.,* 1997). As such, these samples enable investigation of the role of thermal metamorphism and partial melting on the $\delta^{37}Cl$ composition of apatite in meteorites.

Materials and Methods

Three acapulcoites (Acapulco, NWA 10074, Dhofar 125) and two lodranites (Lodran and NWA 11970), covering 1 % to 20 % partial melting were investigated (see Supplementary Information for details on samples). Whole mount X-ray maps of Ca, Fe, Mg, P and Si were collected on the Cameca SX-100 electron probe microanalyser (EPMA) at the University of Arizona to identify phosphates in thin sections of Acapulco, NWA 10074, Dhofar 125, NWA 11970 and Lodran (Fig. S-1). Chemical characterisation of phosphates was carried out with a JEOL Superprobe JXA-8230 EPMA at the Department of Earth Sciences, University of Firenze. Chlorine concentration and isotopic measurements were performed with the secondary ion mass spectrometer NanoSIMS 50L at the Open University, UK. Secondary negative ions $^1H^{16}O^-$, $^{18}O^-$, $^{35}Cl^-$, $^{37}Cl^-$ and $^{40}Ca^{19}F^-$ were measured on 12 phosphates from Acapulco, NWA 10074 and Dhofar 125 (n = 21) using a Cs^+ primary beam of ~10 pA rastered over a 5 μm × 5 μm surface area. These same negative ions were imaged by scanning mode over a 10 μm × 10 μm surface area for three phosphates in Lodran and Dhofar 125 (n = 4) <50 μm, too small for spot analyses. Unfortunately, no suitable phosphates for NanoSIMS analyses were found in NWA 11970, mainly due to the presence of cracks. Further details on the analytical protocol can be found in Supplementary Information (Figs. S-2 to S-4).

Phosphates in Acapulcoites-Lodranites: Petrographic Context and Volatile Abundances. Phosphates in acapulcoites and lodranites occur either as interstitial grains of fluorapatite or chloroapatite associated with Fe-Ni metal (Zipfel *et al.,* 1995) or in large veins (McCoy *et al.,* 1996). In fact, similarly to ordinary and carbonaceous chondrites, halogens have been concentrated in apatite almost entirely as a result of secondary processes such as thermal metamorphism (Brearley and Jones, 2018). Phosphorus contained within Fe-Ni metal diffuses out of the metal to form secondary phosphates as metamorphic grade increases (Jones *et al.,*

2014). Interestingly, apatite in OCs also contain low H_2O contents, <100 μg.g^{-1} (Jones *et al.,* 2014), similar to the estimation of water abundances in acapulcoites, *i.e.* <50 μg.g^{-1} (Stephant *et al.,* 2023). Jones *et al.* (2014) suggested that in OCs the apatite record the latest stages of fluid compositions, which were halogen-rich and water-poor. These fluids could have been derived by degassing of melts, produced either by partial melting in the interior of the ordinary chondrite parent bodies, or as a result of impact melting. As such, acapulcoites and lodranites could have recorded these fluids in their apatite, in a similar manner to the OCs.

In apatite, F–, Cl–, and hydroxyl (OH–) anions occupy the X crystallographic site. Cl contents measured by EPMA and by NanoSIMS present a good match, with NanoSIMS Cl abundances ranging from 0.48 ± 0.02 to 6.53 ± 0.33 wt. % (Fig. S-5). As such, we assume that X = F + Cl + OH and recalculated F abundance based on Cl and OH abundances, since F could be overestimated (Davidson *et al.,* 2020; McCubbin *et al.,* 2021; Supplementary Information). The acapulcoite-lodranite apatite compositions cover the entire chloro-fluor-apatite compositional range (Fig. 1). Apatite in acapulcoites Acapulco and NWA 10074 are all F-rich, with F contents >2.80 wt. %, that display mostly subhedral shapes associated with Fe-Ni metallic phases (Fig. S-2). Acapulco apatite are generally several hundreds of μm in size and have an average Cl content of 1.6 ± 0.12 wt. % (n = 5; 2 s.d.), while NWA 10074 apatite sizes range around 100 μm and are Cl-poor with Cl contents below 0.52 ± 0.01 wt. %. Dhofar 125 contains Cl-rich apatite with varying F concentrations (2.89–5.57 wt. % Cl; 0.69–2.26 wt. % F; Fig. 1) and have smaller grain sizes than in the other two acapulcoites (50−100 μm). In the two lodranites Lodran and NWA 11970, apatite is rarer and much smaller (20 to 50 μm). NWA 11970 phosphates are heavily fractured and thus are unsuitable for NanoSIMS analyses. Lodran apatite are all F-poor chlorapatite (6.17 ± 0.05 wt. % Cl) while NWA 11970 is mostly comprised of volatile-free merrillites, with only one F-rich chlorapatite found in the thin section (2.49 wt. % F; 2.44 wt. % Cl). The F-rich composition of most acapulcoite and lodranite apatite analysed here is similar to some OCs affected by impact melting (McCubbin *et al.,* 2023), as well as igneous apatite found in eucrites (*e.g.*, McCubbin *et al.,* 2021) and in lunar mare basalts (*e.g.*, Boyce *et al.,* 2014). Chondritic apatite are typically enriched in Cl (Brearley and Jones, 2018), similar to Lodran and Dhofar 125, suggesting latest stages of fluid compositions. However, it is important to note here that OC apatite also contain another unknown component, other than OH, missing in acapulcoite-lodranites (Jones *et al.,* 2014).

During degassing of its parental melt, apatite should evolve towards fluorapatite composition due to the relative volatility of the X site components: H > Cl > F (McCubbin *et al.,* 2021). Here, Lodran which underwent higher degrees of partial melting (*i.e.* 20 %; Supplementary Information), contain chlorapatite, which would tend to argue against preferential degassing of Cl towards F. However, considering that higher abundances of H and Cl should be released with increasing partial melt, the abundances of H, Cl and F in apatite cannot directly hint to the potential volatile loss experienced by these primitive achondrites during partial melting. As such, we need to estimate the bulk abundances of volatiles in acapulcoites and lodranites in order to gain insight into the behaviour of Cl, F and H abundances during early planetary differentiation.

Bulk Rock Abundances of F, Cl and H_2O in Acapulcoites and Lodranites. Using the method detailed in McCubbin *et al.* (2021), we estimated the bulk F abundance for each meteorite studied here, as well as their Cl and H_2O bulk abundances (Supplementary Information; Table S-2). Acapulcoites bulk F
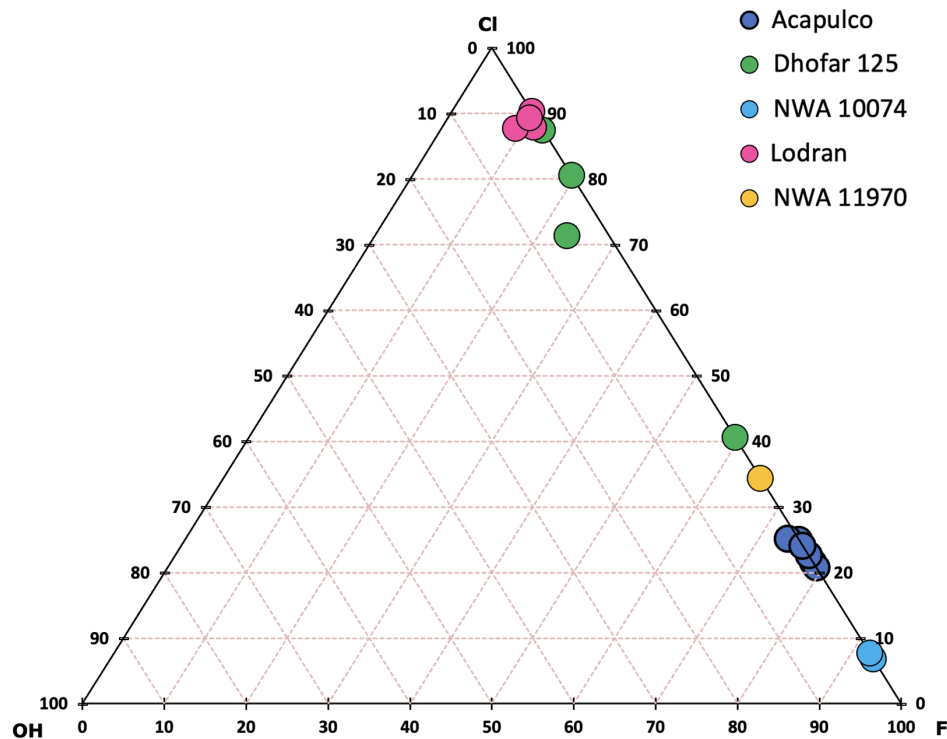
Figure 1 Ternary plot of apatite X-site occupancy (wt. %) from acapulcoites and lodranites. Procedures adopted for estimating OH and F contents are explained in the main text and Supplementary Information (McCubbin et al., 2021).

abundances range from 42–392 $\mu g.g^{-1}$ while Lodran bulk F content is estimated to be 2 $\mu g.g^{-1}$. Bulk Cl concentrations have been estimated by McCoy et al. (1997) for Acapulco (i.e. 250 $\mu g.g^{-1}$) and Lodran (i.e. 10 $\mu g.g^{-1}$), as well as from Garrison et al. (2000) for Acapulco (i.e. 204 ± 26 $\mu g.g^{-1}$) with other acapulcoites ranging from 131 ± 38 to 268 ± 28 $\mu g.g^{-1}$ and lodranites from 35 ± 110 to 78 ± 69 $\mu g.g^{-1}$. Due to large uncertainties on the Cl, F and OH partition coefficient between melt and apatite (McCubbin et al., 2015), we estimate lower limits for Cl bulk abundances. These estimations are in the range of literature data, ranging from 47 to 501 $\mu g.g^{-1}$ Cl for acapulcoites and from 59 to 73 $\mu g.g^{-1}$ Cl for lodranites. Depending on modal abundance estimations (see Supplementary Information), Dhofar 125 may have a higher bulk Cl content due to terrestrial alteration, highlighted by its higher degree of weathering (W1/W2) and the contiguous terrestrial alteration products to phosphates (Fig. S-2). Indeed, an increase of 0.4 vol. % of Ca phosphates in modal abundance estimation results in a bulk Cl abundance estimation almost three times higher. This highlights the necessity of precise Ca phosphate modal abundance in order to determine accurate bulk Cl abundances. Acapulcoites contain more F and Cl than lodranites, suggesting that lodranites experienced degassing or volatile-rich melt loss as a result of partial melting increase. Regarding the H_2O content, estimations based on apatite (i.e. 3–30 $\mu g.g^{-1}$ H_2O) are consistent with those made on nominally anhydrous minerals via NanoSIMS analyses (i.e. 3–19 $\mu g.g^{-1}$ H_2O; Stephant et al., 2023). Overall, the bulk F and Cl contents of acapulcoite-lodranites are consistent with OC compositions (i.e. F = 8–300 $\mu g.g^{-1}$; Cl = 7–270 $\mu g.g^{-1}$; Lodders and Fegley, 2023 and references therein), while ECs are even more enriched in Cl (up to 1000 $\mu g.g^{-1}$; Brearley and Jones, 2018). As such, it would seem that the acapulcoites-lodranites still record the F and Cl contents of their chondritic precursor, which were very similar in composition to OC and EC parent bodies. Comparing the estimated volatile bulk content between

acapulcoites and Lodran, it appears that Cl and F have either degassed or been carried away together with melt loss in lodranites, both mechanisms happening during early degree of partial melting (<20 %).

Effect of Partial Melting on the Chlorine Isotopic Composition of Phosphates. The $\delta^{37}Cl$ values in acapulcoite-lodranite apatite range from –0.91 ± 1.27 to +2.81 ± 0.71 ‰ (2 s.d.) (Table 1), with no apparent correlation with their large variation in Cl content (Fig. 2). Moreover, no correlation between $\delta^{37}Cl$ measured in a specific sample and its degree of partial melting is observed. As such, chlorine isotopic compositions of apatite in acapulcoites and lodranites do not record notable (<2 ‰) fractionation during partial melting. As a result, the average $\delta^{37}Cl$ value for acapulcoite-lodranites of +1.1 ± 0.8 ‰ (n = 25; 1 s.d.) is considered to be representative of the acapulcoite-lodranite parent body (ALPB), the chemical composition of which lies in between H ordinary chondrites and EL enstatite chondrites (e.g., Keil and McCoy, 2018).

Sharp et al. (2013) measured bulk $\delta^{37}Cl$ values in OCs, ECs, and CCs, with values of –0.4 ± 0.7 ‰, +0.4 ± 0.3 ‰ and –0.2 ± 0.6 ‰, respectively (Fig. 2), arguing for no variation in chlorine isotopic composition amongst the different groups of chondrites. Taken at face value, the slightly heavier $\delta^{37}Cl$ of ALPB derived from apatite (+1.1 ± 0.8 ‰) could argue for some preferential loss of ^{35}Cl induced during metamorphism or degassing associated with partial melting. In fact, Sharp et al. (2013) also argued that the ~1 ‰ increase observed in bulk $\delta^{37}Cl$ between EH3 and EL6 chondrites could be interpreted as a sign of isotopic fractionation due to degassing. Moreover, the higher and more variable $\delta^{37}Cl$ values in apatite compared to bulk rock, also observed in lunar basalts, have been suggested to result from local degassing affecting apatite-forming melts and/or fluids (Gargano et al., 2020). As a result, Gargano et al. (2020) suggested that lunar apatite are not representative of $\delta^{37}Cl$ bulk

Table 1 Cl content and δ³⁷Cl values and associated 2σ errors for apatite in Acapulco, NWA 10074, Dhofar 125 and Lodran measured by NanoSIMS (see Supplementary Information for details on the NanoSIMS protocol). Cl content estimated by EPMA is also given for comparison.

Sample	Phosphate number	Cl (wt. %)	2σ	δ³⁷Cl (‰)	2σ	Cl by EPMA (wt. %)
Acapulco	Ph1_2	1.61	0.08	0.96	0.81	1.49
	Ph1_3	1.64	0.08	1.42	0.81	1.49
	Ph2_2	1.64	0.08	0.46	0.82	1.44
	Ph2_3	1.62	0.08	0.06	0.83	1.44
	Ph3_2	1.74	0.09	1.33	1.27	1.72
	Ph3_3	1.71	0.09	−0.92	1.27	1.72
	Ph4_2	1.73	0.09	0.02	0.76	1.56
	Ph4_3	2.11	0.11	1.13	0.71	1.56
	Ph5_2	1.47	0.07	0.72	0.80	1.73
	Ph5_3	1.70	0.09	1.53	0.77	1.65
NWA 10074	Ph1-2	0.48	0.02	1.87	1.21	0.50
	Ph1_3	0.49	0.02	1.96	1.21	0.50
	Ph2_2	0.53	0.03	0.17	1.16	0.50
	Ph2_3	0.51	0.03	1.70	1.17	0.50
	Ph4_2	0.55	0.03	0.57	1.16	0.55
	Ph4_3	0.55	0.03	1.37	1.16	0.55
Dhofar 125	Ph1_1*	5.51	0.29	0.75	0.52	5.65
	Ph1_2*	6.05	0.32	1.57	0.51	5.65
	Ph2_2*	6.42	0.34	0.22	0.51	6.04
	Ph3_2	1.92	0.10	1.96	0.83	2.89
	Ph4_1	6.53	0.33	2.23	0.66	5.03
	Ph5_4	3.97	0.20	2.81	0.71	0.46
	Ph5_5	2.01	0.10	1.61	0.82	0.46
	Ph5_6	2.36	0.12	0.64	0.78	0.46
Lodran	Ph2*	5.44	0.28	1.02	0.51	6.18

* Identifies measurements made by image mode rather than spot mode.
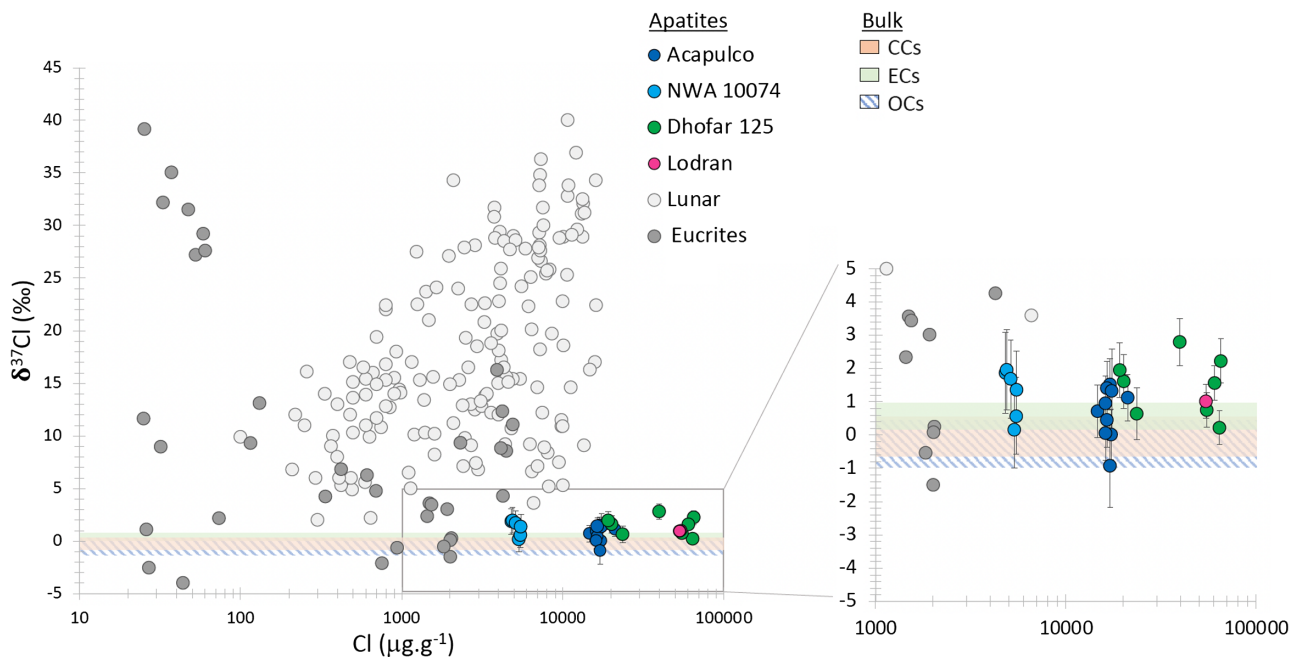
Figure 2 Chlorine abundance (µg.g⁻¹) and isotopic composition (δ³⁷Cl in ‰) of acapulcoite and lodranite apatite, lunar apatite (Barnes *et al.,* 2019 and references therein), eucrite apatite (Barrett *et al.,* 2019 and references therein). The orange band, green band and dashed blue band represent the average for bulk carbonaceous chondrites (CCs), enstatite chondrites (ECs) and ordinary chondrites (OCs), respectively (Sharp *et al.,* 2013).

rock. Nevertheless, our estimation of the ALPB $\delta^{37}Cl$ value is within uncertainties of $\delta^{37}Cl$ of bulk chondrites. Therefore, contrary to differentiated bodies, apatite in chondrites and primitive achondrites can be considered representative of their bulk Cl isotopic composition.

Conclusion

Overall, while some Cl and F degassing might have occurred at 20 % partial melting, as highlighted by the lower F and Cl contents estimated in Lodran, no significant chlorine isotopic fractionation among acapulcoite-lodranite apatite during early stages (*i.e.* lower degrees) of partial melting occurred. Therefore, acapulcoites and lodranites retain their chondritic $\delta^{37}Cl$ value, similar to that of most bulk chondrites. A similar conclusion has been reached regarding the hydrogen isotopic composition of acapulcoites and lodranites, where the δD value matches the inferred hydrogen isotopic composition of water in OCs (*cf.* Jin *et al.*, 2021; Stephant *et al.*, 2023). As such, it seems that acapulcoites and lodranites have retained most of their initial volatile chondritic isotopic compositions.

Acknowledgments

Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2406.

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