Martian magmas contained abundant chlorine, but little water

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ABSTRACT

The martian water cycle, a key for the habitability of Mars, largely relies on the abundance of water in martian magmas and their mantle sources, yet martian (SNC: shergottite, nakhlite, and chassigny classes) meteorites contain minimal water. However, some experimental studies have suggested that martian parental magmas contained as much as 2% H_2O. Here we integrate mineral-chemical, experimental, and cosmochemical constraints to show that martian magmas contained little water but abundant chlorine. Apatite and amphibole in martian meteorites are chlorine rich and water poor; this constrains the chlorine contents in their parental magmas to >0.3 wt% and water contents to <0.3 wt%. Our experimental work has shown that large amounts of water are not needed to explain the mineralogy of the martian meteorites because chlorine has effects similar to those of water on crystallization. Such chlorine-rich, water-poor martian magmas are consistent with Mars being chlorine rich (~2.5 ×) compared with the Earth. Furthermore, the bulk Cl composition of martian meteorites shows that they have not preferentially lost Cl by degassing of an H_2O-rich vapor. Together, these results show that chlorine, not water, was the dominant volatile species in martian basalts, and that these basalts contributed little H_2O to Mars' surface environment.

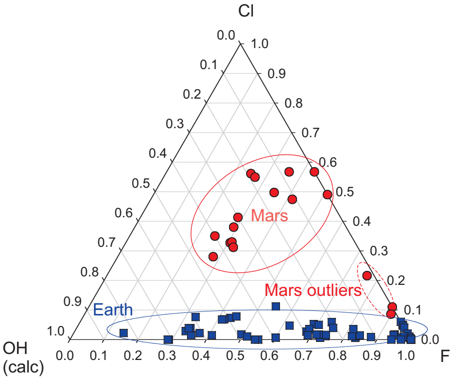

Figure 1. Proportions of Cl, F, and OH in apatite grains from martian meteorites (red circles) and in terrestrial basaltic rocks (blue squares). Martian apatite compositions that have significantly more F than average (Mars outliers) are shown by dashed red circle. OH is rarely analyzed directly, and so is calculated by difference in stoichiometry. References and rock types are listed in Table DR1 in the GSA Data Repository.[1] Terrestrial sample suites that have been altered or metasomatized are excluded.

INTRODUCTION

Despite the importance of water to martian geology and its potential in effecting the course of igneous petrogenesis, martian meteorites are nearly anhydrous: 5–15 ppm bound H_2 (released at temperature > 350 °C) (Leshin et al., 1996), equivalent to ~50–150 ppm H_2O. Comparable terrestrial basaltic rocks typically contain ~2000–20,000 ppm structural H_2O (Johnson et al., 1994). The low water contents of martian basalts imply a water-poor mantle with ~3–15 ppm H_2O (assuming that the basalt magmas are partial melts 10%–20%). However, some researchers argue that martian basaltic magmas contained as much H_2O as terrestrial magmas, and the basalts (now the martian meteorites) were nearly completely degassed on eruption and/or emplacement (Dann et al., 2001; Johnson et al., 1991; McSween et al., 2001; Nekvasil et al., 2007). By integrating mineral-chemical, experimental, and cosmochemical constraints, we show that magmas parental to the martian meteorites had chlorine instead of water as the dominant volatile species.

Known martian igneous rocks consist of the suite of martian meteorites (SNCs [shergottite, nakhlite, and chassigny classes] and ALH84001) (e.g., McSween, 1985; Treiman, 2005; Treiman et al., 2000) and compositions analyzed on the surface of Mars from the rover missions (e.g., Gellert et al., 2006; Squyres et al., 2007). Martian meteorites represent our only samples of the surface of Mars, and therefore are the focus of this study; compositional data from outcrops on the martian surface from orbiter and rover missions are used for comparison. The martian meteorites range from basaltic (shergottites) through cumulate (nakhlites, chassignites, and ALH84001) rocks. The martian meteorites are relatively young (1.3–0.17 Ga; e.g., Jones, 1986; Nyquist et al., 2001; Treiman, 2005), with the exception of ALH84001 (4.5 Ga; Nyquist et al., 2001), and there is some debate whether the shergottites represent typical martian basalts (e.g., Filiberto et al., 2006; McSween et al., 2009; Taylor et al., 2006). Therefore, we include data from the young martian meteorites, ALH84001, and compositions of surface basalts (thought to be ca. 3.65 Ga; Greeley et al., 2005) to show that all data sets are consistent (within the range of available data) with martian magmas having chlorine instead of water as the dominant volatile species.

MINERAL-CHEMICAL CONSTRAINTS

Apatite and amphibole monitor the volatile contents (OH, F, Cl) of their parental magmas. Therefore, their chemistry can be used to constrain the preeruptive volatile contents of their parental magmas (Mathez and Webster, 2005; Patiño Douce and Roden, 2006; Stormer and Carmichael, 1971; Westrich, 1982).

Apatite [$Ca_5(PO_4)_3(OH,F,Cl)$] readily accepts F, Cl, and OH from its parent magma (Mathez and Webster, 2005; Stormer and Carmichael, 1971; Zhu and Sverjensky, 1991), and can be used to compare the halogen contents of magmas from different planets. Apatites in martian meteorites contain little OH (McCubbin and Nekvasil, 2008; Patiño Douce and Roden, 2006) and have average Cl:F:OH (molar) ratios of ~5:3:2 (Fig. 1). In contrast, apatites from terrestrial basaltic rocks contain almost no Cl (<0.4 wt% Cl; e.g., Casey et al., 2007) and variable OH:F ratios (Fig. 1). There are, however, a few martian apatite compositions that have significantly more F than the average (e.g., McCubbin and Nekvasil, 2008); these outliers actually contain no water and are significantly enriched in Cl compared with apatite from terrestrial basalts. Therefore, all martian apatite compositions are consistent with their parental magmas being chlorine rich and water poor compared with terrestrial magmas.

Similarly, the volatile element compositions of amphiboles can be used to constrain the volatile contents of their parent magmas. Amphiboles accept volatiles from basaltic melt in the preference order OH > F > Cl (Enami et al., 1992; Sato et al., 2005; Zhu and Sverjensky, 1991). The amphiboles in martian meteorites are chlor-amphiboles, chlor-fluor-amphiboles, or oxy-amphiboles (Table 1) (Sautter et al., 2006; Watson et al., 1994), and not hydroxy-amphiboles, as would be expected in water-rich systems. The only direct water analyses of martian amphiboles are the Ti-rich kaersutites in the Chassigny (France) and Zagami

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[1] GSA Data Repository item 2009270, references and rock types used in Figures 1 and 3, is available online at www.geosociety.org/pubs/ft2009.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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TABLE 1. AMPHIBOLE AND PARENTAL LIQUID VOLATILE COMPOSITIONS

	Chassigny	NWA 2737	MIL 03346	Zagami
Amphibole compositions (wt%)				
Cl	0.1[a]	0.09[d]	6[e]	n.d.
F	0.5[a]	0.45[d]	0.25[e]	0.08[f]
H_2O	0.1–0.2[b] 0.4–0.7[c]	n.a.	0.1*	0.1–0.2[b]
Amphibole-melt partition coefficients				
D(Cl)	0.38[d,g]	0.38[d,g]	0.38[d,g]	0.38[d,g]
D(F)	1.65–4[d,g]	1.65–4[d,g]	1.65–4[d,g]	1.65–4[d,g]
D(H_2O)	>1–20[h]	>1–20[h]	>1–20[h]	>1–20[h]
Calculated parental liquid compositions (wt%)				
Cl	0.3	0.3	16	
F	0.1–0.3	0.1–0.3	0.06–0.2	0.02–0.05
H_2O	0.01–0.2 0.04–0.7		≪0.1	0.01–0.2

Note: n.a.—no analysis; n.d.—not detected.

*Maximum H_2O concentration based on stoichiometry.

[a] Floran et al. (1978); Johnson et al. (1991).
[b] Watson et al. (1994).
[c] McCubbin et al. (2009).
[d] Treiman et al. (2007).
[e] Sautter et al. (2006).
[f] Treiman et al. (1985).
[g] Gillis et al. (2003).
[h] Mysen et al. (1998)

(Nigeria) meteorites (e.g., Watson et al., 1994). The kaersutite in the Chassigny meteorite has a Cl:F (molar) ratio of ~1:10 (0.1 wt% Cl, 0.5 wt% F; Floran et al., 1978; Johnson et al., 1991) and two distinctly different analyses of water contents: 0.1–0.2 wt% H_2O (Watson et al., 1994) and 0.4–0.7 wt% H_2O (McCubbin et al., 2009). Kaersutite in NWA 2737 (northwest Africa) has a similar Cl:F (molar) ratio of ~1:10, and similar concentrations of chlorine and fluorine (0.09 wt% Cl, 0.45 wt% F; Beck et al., 2006; Treiman et al., 2007), suggesting similar water contents (Table 1). MIL 03346 (Miller Range, Antarctica) contains amphibole (chloro-potassic-hastingsite) with an even higher Cl:F (molar) ratio of ~18:1, and higher chlorine concentrations (6 wt% Cl, 0.25 wt% F) than the amphiboles in either the Chassigny meteorite or NWA 2737 (Sautter et al., 2006), suggesting even lower water contents (<0.1 wt% H_2O). Using a Cl amphibole-melt partition coefficient for terrestrial basalts of ~0.38 (Gillis et al., 2003) (consistent with the measured Cl kaersutite-melt partition coefficient of ~0.35 for NWA 2737; Treiman et al., 2007), one calculates magmatic chlorine concentrations of 0.3 wt% for the Chassigny meteorite, 0.3 wt% for NWA 2737, and 16 wt% for MIL 03346. This last value is far above the chlorine solubility limit in a basalt melt (~3 wt% Cl; Webster et al., 1999), and suggests that the partition coefficient is inappropriate for such a chlorine-rich system. Because amphibole preferentially accepts OH and F over Cl from a basaltic melt (Enami et al., 1992; Sato et al., 2005; Zhu and Sverjensky, 1991), these basalts must have had less F and H_2O than chlorine (F and $H_2O \ll 0.3$ wt%; Table 1). Thus, chlorine was the dominant volatile species in their parental basalts.

EXPERIMENTAL CONSTRAINTS

Results of some melting and crystallization experiments on martian meteorites suggest, based on mineral compositions and crystallization temperatures, that their parent magmas contained as much as 2 wt% H_2O (Dann et al., 2001; Johnson et al., 1991; McSween et al., 2001; Nekvasil et al., 2007). However, large quantities of magmatic water would not be required if some other component, such as chlorine, could induce the same combination of mineral compositions and temperatures.

Our recent experiments have shown that chlorine and water have similar effects on crystallization temperatures and mineral compositions (Filiberto and Treiman, 2009). Figure 2 shows the experimental near-liquidus mineral-melt phase relations for a martian basalt (Humphrey, Adirondack-class Gusev basalt) with no added volatiles (Filiberto et al., 2008), with added Cl (Filiberto and Treiman, 2009), and with added H_2O (Monders et al., 2007). In all sets of experiments, olivine is the liquidus phase at low pressure, and low-Ca pyroxene is on the liquidus at high pressure. Compared to the anhydrous phase relations, addition of 0.7 wt% Cl moves the olivine–low-Ca-pyroxene (pigeonite) liquidus point from 1385 °C and 12.5 kbar to 1305 °C and 8.5 kbar, greatly enlarging the stability field of low-Ca pyroxene. Addition of 0.8 wt% H_2O causes similar effects: the olivine–low-Ca-pyroxene (orthopyroxene) liquidus point moves from 1385 °C and 12.5 kbar to 1325 °C and 10.6 kbar. Thus, addition of comparable weight proportions of Cl and H_2O to the melt causes comparable depression of its liquidus. Chlorine and water, however, have different solubility mechanisms and effects on melt structure. Water attacks bridging oxygens in the melt structure, depolymerizing the melt, causing liquidus depression (e.g., Mysen and Cody, 2004; Zeng et al., 1999), but this has little (if any) effect on phase boundaries (e.g., Médard and Grove, 2008). However, chlorine complexes in the melt with network-modifying cations (Mg, Fe, Ca, Na), affecting cation and silica activity, viscosity, and melt polymerization (e.g., Stebbins and Du, 2002; Zimova and Webb, 2006), and thereby causes liquidus depression, increased pyroxene stability with respect to olivine, and multiple saturation point depression.

Addition of similar weight proportions of Cl and H_2O also has similar effects on the composition of crystallizing minerals. The first

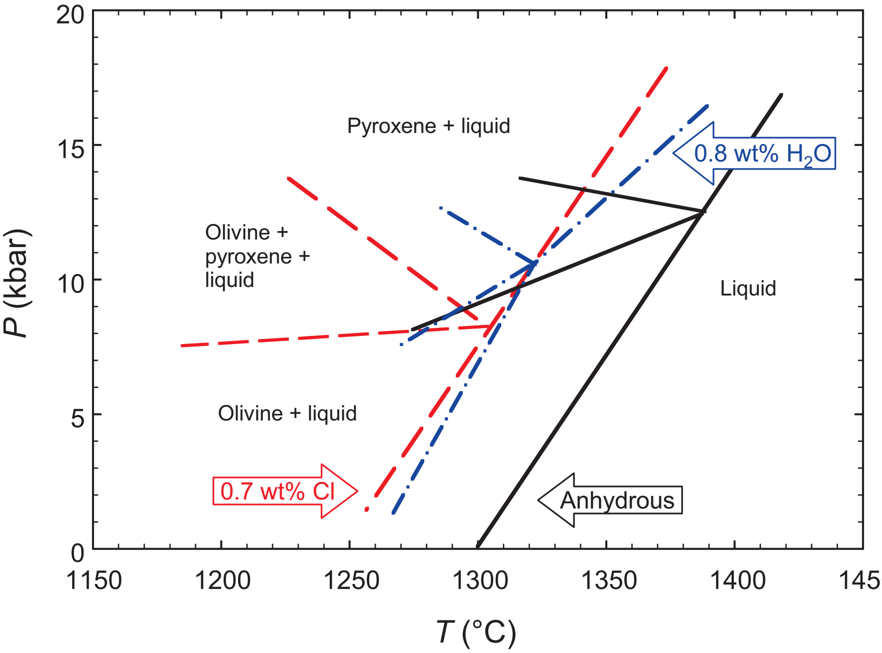

Figure 2. Experimentally determined phase relations for Humphrey bulk rock composition with 0.7 wt% Cl, 0.1 wt% H_2O (red dashed lines) (Filiberto and Treiman, 2009), compared with anhydrous (0 wt% Cl, 0.1 wt% H_2O) experimental work (black lines) (Filiberto et al., 2008), and hydrous (0 wt% Cl, 0.8 wt% H_2O) experimental work (blue dashed-dotted lines) (Monders et al., 2007). Since both "anhydrous" and chlorine-added experiments contain the same amount of dissolved water (<0.1 wt% H_2O), the shift in phase boundaries is caused solely by addition of chlorine (Filiberto and Treiman, 2009). Uncertainty for each piston-cylinder experiment is ±0.5 kbar and ±10 °C (e.g., Longhi, 2005; Médard et al., 2008); therefore, all shifts in phase boundaries are statistically significant and outside the error for piston-cylinder experiments.

pyroxenes to crystallize in both the Cl-added ($\mathrm{En}_{75}\mathrm{Wo}_{7}\mathrm{Fs}_{18}$; Filiberto and Treiman, 2009) and H_2O-added ($\mathrm{En}_{75}\mathrm{Wo}_{4}\mathrm{Fs}_{21}$; Monders et al.; 2007) experiments have higher Mg#s and lower Wo than in the anhydrous experiments ($\mathrm{En}_{68}\mathrm{Wo}_{9}\mathrm{Fs}_{23}$; Filiberto et al., 2008), while the olivine is slightly more magnesian (Fo_{75} for hydrous; Monders et al., 2007; Fo_{74} for chlorine added; Filiberto and Treiman 2009) than in the anhydrous experiments ($\mathrm{Fo}_{72;}$ Filiberto et al., 2008). Thus, chlorine-bearing martian basalts show the same mineral compositions and crystallization temperatures as water-bearing martian basalts. Therefore, mineral compositions previously interpreted as indicating water enrichment could equally well be explained by chlorine enrichment.

COSMOCHEMICAL CONSTRAINTS

The concept of chlorine-rich martian basalts is consistent with the composition of the martian mantle inferred from compositions of martian meteorites (e.g., Dreibus and Wänke, 1985). Nearly all martian meteorites have Cl/La ≈ 45 (Fig. 3). This Cl/La value is taken as that of the martian mantle (and the planet), because Cl and La are both comparably incompatible in the generation of basaltic magma. The martian Cl/La is ~2.5 times the average for terrestrial basalts and mantle rocks (Fig. 3), which is consistent with Mars' overall enrichment in cosmochemically volatile elements (like alkalies and other halogens) compared to the Earth (Dreibus and Wänke, 1987). Thus, one should expect martian basalts to be richer in chlorine than comparable terrestrial basalts.

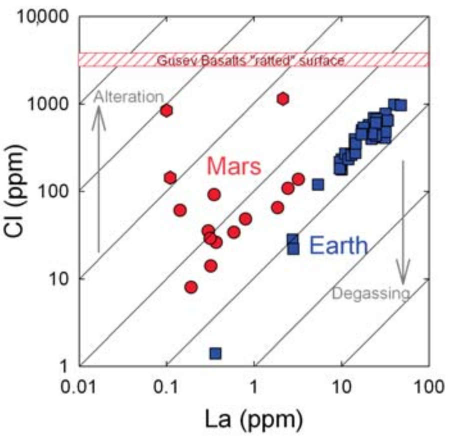

Figure 3. Average bulk chlorine and lanthanum abundances in martian basaltic meteorites (red circles), martian meteorites affected by weathering (red hexagons), and in terrestrial basaltic rock (blue squares). Terrestrial samples chosen for this study represent magmas that are relatively undegassed (Simons et al., 2002). Unaltered martian meteorites average Cl/La ~45, or 0.015× Cl chondritic. Terrestrial lavas average Cl/La ~20, or 0.007× Cl. Also plotted for comparison is Cl concentration of Gusev Adirondack Class basalts (red box) after removal of all surface coatings and alteration products by rock abrasion tool ("ratted" surface); La was not measured in these rocks (Gellert et al., 2006). Data sources and rock types are given in Table DR2. Arrows (gray) show effects of alteration and degassing on basaltic magmas.

Chlorine-rich martian magmas are also consistent with evidence from spacecraft data. Results from the gamma ray spectrometer (GRS) on the Mars Odyssey orbiter show that the martian surface is Cl-rich, with abundances from 0.2 to 1 wt% (Keller et al., 2006). The highest Cl abundances are in the Tharsis volcanic province (to ~0.8 wt %) and are attributed to volcanic exhalations (Keller et al., 2006). On the surface, Mars Exploration Rover (MER) *Spirit* analyses show that Gusev crater basalts (Adirondack class) average ~0.2 wt% Cl (Gellert et al., 2006) (interior, after removal of surface dust and weathering rind), far above that in typical Earth basalts (Fig. 3). However, the Gusev basalts contain vesicles, which imply that they degassed during eruption and that their preeruptive Cl contents were higher. However, the martian meteorites do not show physical evidence for degassing (e.g., vesicles) or chemical evidence for degassing. If the meteorites had extensively degassed a water-rich vapor, as has been proposed to explain the formation of a dry basalt from a water-rich parental magma (Dann et al., 2001; Johnson et al., 1991; McSween et al., 2001; Nekvasil et al., 2007), the Cl/La ratios of the basalts should vary depending on the extent of degassing (Dreibus and Wänke, 1987; Treiman, 2003). However, no martian meteorites have Cl/La ratios <45 (Fig. 3), suggesting that this is the magmatic Cl/La ratio for martian basalts and that none have degassed. Therefore, the martian meteorites could not have degassed a water-chlorine–rich vapor phase, and so could not have crystallized from water-rich parental magmas.

CONCLUSIONS

The compositions of martian apatites and amphiboles, and bulk compositions of martian meteorites, show that their parental magmas contained abundant chlorine but little water. Experimental results on melting and crystallization of martian basalts, interpreted in terms of water-rich magmas (Dann et al., 2001; Johnson et al., 1991; McSween et al., 2001; Nekvasil et al., 2007), are equally well explained by chlorine-rich magmas. Martian basalts contain significantly more chlorine than comparable terrestrial basalts, an observation consistent with Mars' overall enrichment in volatile elements. Together, these results suggest that parental magmas of martian meteorites contained chlorine, and not water, as their dominant volatile species. If so, eruption of these chlorine-rich, water-poor basalts contributed to the acidity of the martian surface environment, but added little of the water so necessary for life as we know it.

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