VOLATILE COMPOUNDS IN SHERGOTTITE AND NAKHLITE METEORITES.
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INTRODUCTION. Since discovery of apparent carbonate carbon in Nakhla [1], significant evidence has accumulated for occurrence of volatile compounds in shergottites and nakhlites. Gooding and Muenow [2] showed that at least one shergottite (EET7/9001) contains substantial sulfur in a highly oxidized form and that the oxidation must have occurred on the shergottite parent planet. Burgess et al. [3] also found oxidized sulfur in ALHA77005, Shergotty, Nakhla, and Chassigny. Kerridge [4] reported carbon and deuterium of apparent pre-terrestrial origin in Shergotty and Lafayette (a nakhlite). In addition, discrete grains of salt minerals have been documented in EET7/9001 and Nakhla [5-7]. Here we present final results from our study of volatile compounds [8] in three shergottites, one nakhlite, and three eucrite control samples.

SAMPLES AND METHODS. Antarctic meteorites included Allan Hills specimens ALHA77005 (shergottite) and ALHA81001 (eucrite), as well as Elephant Moraine specimens EET7/9001 (shergottite; lithologies A, B, and C), and EET7/9004 (eucrite). A Pasamonte eucrite specimen (USNM-897) was obtained from the U. S. National Museum and a Naklda specimen (BMNH-1911,369) from the British Museum (Natural History). A Shergotty specimen (GSI-179) was sub-divided from material allocated by the Geological Survey of India to Duke [9]. Samples from the exterior (0-0.5-cm depth, including fusion crust) and interior (> 1-cm depth) of each meteorite were individually analyzed so that terrestrial weathering and contamination could be recognized and not misinterpreted as parent-body effects. Each sample (20-80 mg) was pyrolyzed by continuous heating to 1500 K at 5-6 K/min under 10^-7 torr vacuum in a Knudsen cell fitted with a high-purity aluminum oxide liner. Evolved gases were analyzed by a quadrupole mass spectrometer that was continuously scanned over m/e = 2-100 to simultaneously measure abundances of H20, CO2, CO, SO2, S2, H2S, HCl, Cl, and hydrocarbons.

RESULTS. WATER. For interior samples of shergottites, H2O-release profiles and total water concentrations (< 0.1%), are not obviously correlated with similar data for other volatile species. A typical H2O-release profile consists of a single broad peak at 350-700 K that suggests desorption of loosely bound water. Although Nakhla was previously suspected to contain hydrous phases [7,10,11], its measured water content was less than anticipated and its H2O-release profile was similar to those of shergottites. As discussed below for chlorine, however, it is possible that much of Nakhla's indigenous water reacted to form HCl during pyrolysis. Therefore, the actual water content of Nakhla may be significantly higher than the 0.007% measured as evolved H2O. Kerridge [4] reported a total of 13.3 ppm H in Shergotty and 58.3 ppm H in the nakhlite, Lafayette, but discarded as terrestrial contamination all gases extracted at < 723 K. Judging from our results for Nakhla, however, HCl evolution (and, by inference, water loss) begins at < 700 K in nakhlites. Therefore, it is possible that Kerridge underestimated the nakhlite water content. CARBON. Carbon-containing species included CO2, CO, and both saturated and unsaturated hydrocarbons (C1-C4 compounds). The contrast between exterior and interior samples is not always obvious although there are differences between shergottites/Nakhla, as a group, and eucrites. In addition, comparatively low hydrocarbon contents of Antarctic interior specimens distinguish them from non-Antarctic museum specimens. Evolution of CO2 from shergottite and Nakhla samples occurs as peaks centered near 750 K or 900 K, the same temperature range over which CaCO3 decrepitates under vacuum [12]. Because CaCO3 has been independently identified as discrete mineral grains in both EET7/9001 [5] and Nakhla [7], the evolved CO2 is attributable to CaCO3. Shergotty shows a surprising abundance of CO2 but, unlike other shergottite specimens, the CO2 evolution is closely correlated with the release of hydrocarbons. Therefore, it is likely that some or all of the "apparent carbonate" in Shergotty is associated with terrestrial organic contamination. Two different CO2 peaks in the Nakhla profile suggest two different CO2 carriers. Both of the Nakhla carriers must be inorganic, however, because the Nakhla sample in question is virtually free of hydrocarbons. For interior samples of shergottites and Nakhla, there exists an inverse correlation between CO2 and oxidized sulfur (correlation coefficient, r = -0.71 for 11 samples). Kerridge [4] reported a total of 69.3 ppm C in Shergotty and 112.0 ppm C in Lafayette. His procedure discarded as contamination all gases extracted at < 723 K and interpreted as pre-terrestrial the gases extracted above 1023 K (Lafayette) or 1073 K (Shergotty). Judging from our results for Shergotty and Nakhla, however, Kerridge's procedure might not have excluded some of the hydrocarbon contamination in Shergotty and might have sacrificed some of the carbonate in Nakhla. Our results indicate an intrinsic 210 ppm C in Nakhla.

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SULFUR. Most sulfur from interior samples of eucrites, and mildly weathered exterior samples of eucrites, evolves as $S_2$ with minor $H_2S$, consistent with a chemically reduced carrier such as pyrrhotite or troilite (FeS). In contrast, large fractions of the sulfur in shergottites and Nakhla evolves as $SO_2$. The fraction of sulfate in interior samples of eucrites is nearly zero and exhibits a maximum value of about 21% in a moderately weathered interior sample of EETA79004; the fraction of sulfate in shergottite and Nakhla interior samples is 17-100% (Fig. 1). The least oxidized shergottite material is bulk Lith-A from EETA79001 whereas the most oxidized material is Lith-C from the same meteorite; other shergottite and Nakhla samples fall between those limits. Evolution of $SO_2$ from interior samples of shergottites and Nakhla, which show little evidence of terrestrial weathering, is attributable to decrepitation of indigenous sulfates. Calcium sulfate of apparent pre-terrestrial origin has been independently documented for EETA79001 [5] and both Ca- and Mg-sulfate have been found in Nakhla [6,7]. Evolution of $SO_2$ from EETA79001/Lith-C is attributable partly to sulfates and partly to a more refractory component that probably occurs as sulfate ions dissolved in glass [2]. In some samples, such as Nakhla, $SO_2$ evolution at 800-1000 K is correlated with $CO_2$ evolution. In terms of total abundances, however, $CO_2$ and $SO_2$ are inversely correlated, as discussed for carbon results. CHLORINE. Most of the chlorine evolved from interior shergottite samples consists of nearly equal concentrations of Cl and HCl although a few samples favor the latter. Exterior samples of terrestrially weathered eucrites, which also contain hydrous weathering products, show HCl as the dominant chlorine species. Therefore, it is likely that HCl evolution from all samples is fostered by pyrolysis reactions involving hydrated phases. Nakhla represents a special case in which HCl and Cl profiles are staggered in temperature, possibly indicating two different chlorine carriers. In addition, evolution of Cl at 800-1150 K is strongly correlated with evolution of monatomic Na (and K). The correlated Cl/Na evolution is attributable to decrepitation of sodium chloride that has been previously identified in Nakhla [6]. Production of HCl can be tentatively attributed to decrepitation of hydrous "rust" that contains trace Cl [7]. For interior samples of shergottites and Nakhla, total chlorine (Cl + HCl) varies inversely with total sulfur but varies directly with the abundance of oxidized sulfur (Fig. 2). No such correlations exist for interior samples of eucrites.

CONCLUSIONS. Shergottites ALHA77005, EETA79001, and Shergotty, and the nakhlite Nakhla, all contain oxidized sulfur (sulfate) of pre-terrestrial origin; sulfur oxidation is most complete in EETA79001/Lith-C. Significant bulk carbonate was confirmed in Nakhla and trace carbonate was substantiated for EETA79001, all of which appears to be pre-terrestrial in origin. Chlorine covaries with oxidized sulfur, whereas carbonate and sulfate are inversely related. These volatile compounds were probably formed in a highly oxidizing, aqueous environment sometime in the late-stage histories of the rocks that are now represented as meteorites. They are consistent with the hypothesis that shergottite and nakhlite meteorites originated on Mars and that Mars has supported aqueous geochemistry during its history.