OBSERVATION OF THERMAL ALTERATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN ORDINARY CHONDRITES VIA RAMAN SPECTROSCOPY. J. M. Young¹, T. D. Glotch¹, and M. Yesiltas¹ ¹Stony Brook University – Department of Geosciences – 255 Earth and Space Sciences, Stony Brook, NY 11790 (jordan.young@stonybrook.edu)²

Introduction: The study of polycyclic aromatic hydrocarbon (PAH) materials in meteorites, specifically from a spectroscopic standpoint, has received a great deal of attention [1, 2, 4], primarily due to the effects of temperature on the spectral properties of PAHs. The main Raman spectral features of PAHs are the disordered and graphitic (D and G) carbon bands, which occur at ~1350 cm⁻¹ and ~1575 cm⁻¹ respectively. The exact positions of these bands, as well as other spectral parameters such as the band full width half maxima (FWHM), are sensitive to structural and compositional changes associated with the thermal metamorphism of their host chondrites. Thus it stands to reason that portions of a chondrite's thermal history can be gleaned from observing these spectral features [3, 5].

The study of the thermal history of chondritic meteorite via Raman spectroscopy in the recent past has mainly focused on the spectral characteristics of PAH contents in various carbonaceous chondrites, specifically those in the petrologic type range of 3.0-3.9 [3,5,6,7]. This is due to the relatively large eponymous carbon contents of those meteorites. Unequilibrated ordinary chondrites (UOCs) have received particular attention. These UOCs only include the ordinary chondrites of petrologic type 3 that have experienced low thermal metamorphism. However, there has been little study of the equilibrated variety of ordinary chondrites (petrologic type 4-7).

Due to being the majority of collected falls [8], and measurable PAH contents, ordinary chondrites present an opportunity to conduct a comprehensive study of the thermal alteration of meteorites as recorded by the spectral properties of their PAH contents. As such, the purpose of this study is to observe the spectral properties of PAH materials in ordinary chondrites via Raman spectroscopy. Through these observations, we should be able to better understand the link between spectral parameters, petrologic type, and peak metamorphic temperature.

Methods: This study was conducted using a WiTEC alpha300R confocal Raman imaging, coupled with a 512 nm NdYAG green laser. A 50x (NA .80) objective was used to view samples. Several spatially separated 2-D intensity distribution maps were collected from each sample. These maps were collected using laser power of 1.3 mW, image areas of $50x50 \mu m$, and an integration time of 0.1 s in order to minimize the occurrence of sample damage by heating.

Samples: We analyzed ordinary chondrites samples that encompass the known lithological types (H, L, and LL) and have petrologic types with a range of 3.2-6. The meteorites sampled are: Krymka (LL3.2), Tieschitz (H/L3.6), Tennasilm (L4), Ranchapur (H4), Barwell (L5), Limerick (H5), Kernouve (H6), and Mangwendi (LL6).

Results: The average spectra of PAH materials from each of the chondrite samples have been normalized to their respective largest peaks and have been overlain in Figure 1. The spectra, for the most part, have line shapes that closely resemble one another. Barwell, Krymka, Limerick, Ranchapur, and Tieschitz share similar FWHM_D and relative D-band peak intensity (Figure 1). Similarly, nearly all the collected spectra for our chondrite sample set have similar G-band parameters. This group excludes Tennasilm and Krymka, which have narrower G-bands and greater Raman shifts than the other samples.

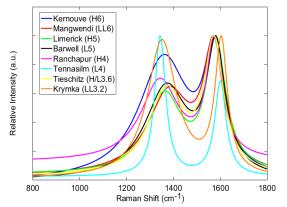


Figure 1. Normalized average spectra of PAH materials from sampled ordinary chondrites.

While the collected spectra have similar line shapes, they are distinct and vary among themselves. Figure 2 demonstrates the spectral variation among members of the high-iron (H) series of ordinary chondrites. While the peak position of the G-band is nearly identical for the selected chondrites of the H type, the peak position of the D-band shifts to higher frequencies with increasing petrologic type. Figure 3 demonstrates the quantitative relationship between the width of the D-band peak (FWHM_D) and the position of the D-band, for all measured chondrites.

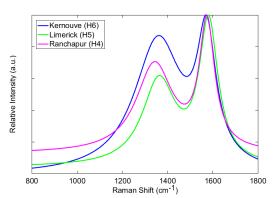


Figure 2. Normalized average spectra for the PAH contents of H type ordinary chondrite samples.

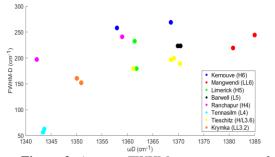


Figure 3. Average FWHM_D vs. average ω_D for all chondrite samples

As seen in Figure 3, there is clear spectral variation between the measured chondrite samples. An interesting phenomenon in the preliminary results is the intrasample variations of Kernouve, Mangwendi, and Ranchapur. These chondrite samples exhibit large variations in the position of their D-band peaks, however, the FWHM_D values for these samples do not share this large variation. More importantly, there appears to be a trend; as position or Raman shift of the D-band increases, the FWHM_D also increases. Additionally, this trend generally coincides with increasing petrologic grade; Chondrites belonging to grades 3 and 4 generally have lower FWHM_D and ω_D values than those of grades 5 and 6. However, this is only true in a general sense as there are exceptions. This concurrent trend can be seen more clearly in Figure 4.

From Figure 4, it becomes clear that as petrologic grade increases, so too does FWHM of the D-band. It should be reiterated that this is a general trend with obvious exceptions that do not fit. Specifically, in this case, the Tennasilm and Limerick chondrites do not fit the general trend.

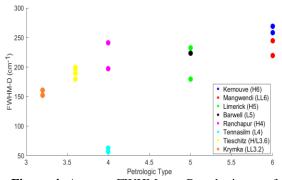


Figure 4. Average $FWHM_D$ vs. Petrologic type for all chondrites

Discussion: Per the preliminary results of this study, it is clear that changes in petrologic type result in distinct changes to Raman spectral properties. Specifically, increasing the petrologic grade of a given chondrite, and by extension peak metamorphic temperature, generally increases the FWHM_D and D-band peak position of observed PAH spectra.

The variation within samples regarding the D-band peak position of spectra collected from Kernouve, Mangwendi, and Ranchapur should also be addressed. It is possible that this variation is a result of brecciation. That is, pieces of the chondrites in question experienced different levels of thermal alteration before ultimately being joined together. More spectra will need to be collected to determine if this variation is a statistical error or if two or more distinct spectral populations coexist with the samples in question.

While the preliminary results show a general trend, we require additional data for each sample to determine how robust it is. In addition, while the samples of this study represent the breadth of ordinary chondrite types (H, L, and LL) and petrologic types, the sample population is in no way comprehensive. As such, future work will include collecting more data on the samples used by this study and collecting data on more ordinary chondrites of diverse petrologic grades and lithologic type.

References: [1] Sephton et al. (2002) *PSS*, 50, 711–716. [2] Quirico et al. (2003) *MPS*, 38, 795–811. [3] Chanet al. (2015) *AbsciCon* [4] Cloutis et al. (2016) *Icarus*, 274, 211-230. [5] Cody et al. (2008) *EPSL*, 272, 446-455. [6] Ronal et al. (2016) *GECA*, 189, 312-337. [7] Homma et al. (2015) *JMPS*, 110, 276-282. [8] Hughes D.W. (1966) *Meteoritics*, 16, 269-281.