Title: The ungrouped chondrite El Médano 301 and its comparison with other highly reduced ordinary chondrites

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Abstract

El Médano (EM) 301 is an ungrouped chondrite with overall texture and trace-element distribution similar to those of ordinary chondrites (OC), but with silicate (olivine and low-Ca pyroxene) compositions that are more reduced than those in OCs, with average olivine and low-Ca pyroxene of $Fa_{3.9 \pm 0.3}$ and $Fs_{12.4 \pm 4.8}$, respectively. These values are far lower than the values for OCs and even for chondrites designed as "reduced" chondrites. Low-Ca pyroxene is the dominant mineral phase and shows reverse zoning along the crystal rims and cracks. Similar to lower Fa and Fs values than OCs, Co content of kamacite is also much lower than the concentrations observed in them. Oxygen-isotope composition is slightly different from those of OCs.

The lower modal olivine/pyroxene, different IR spectra, lower Co content of kamacite, higher MgO contents of olivine and pyroxene, different kamacite texture, and different oxygen-isotope composition show that EM 301 is different from the typical OCs. In sense of their similarities to the OCs but showing more reduced mineralogy, chondritic clasts in Cumberland Falls aubrite (CFC), Northwest Africa (NWA) 7135 and Acfer 370 and EM 301 are similar to each other. However, dissimilar to NWA 7135 and CFC, it does not contain highly reduced mineral phases.

Our observations suggest the formation of EM 301 in a nebular region compositionally similar to OCs but with a different redox state, with oxygen fugacity in this region lower than that of OCs and higher than that of enstatite chondrites (EC) condensation region. Experiencing a second phase of reduction by the production of reducing gas phases (e.g., C-rich) could be responsible for the subsequent reduction of the primary material and the occurrence of reverse zoning in the low-Ca pyroxene and lower average Fa/Fs ratio. Based on the IR spectra of EM 301, which shows resemblance to V-type asteroids, we discuss the possibility of its ejection from a Vesta family asteroid.

1. INTRODUCTION

Chondrites account for the vast majority of meteorites in our collections. Ordinary chondrites (OCs) are the most abundant and form a class divided into H, L, and LL groups. This division corresponds to variable oxidation state among the groups, as reflected in their mineralogy, whole-rock chemistry and oxygen-isotope composition. The oxygen fugacity increases from H to LL chondrites (Rubin, 1990). Much more reduced than OCs, enstatite chondrites (ECs) show different whole-rock chemistry, modal mineral abundances, mineral chemistry, and oxygen-isotope composition (e.g., Keil, 1989; Weisberg and Kimura, 2012; El Goresy et al., 2017). A significant compositional hiatus exists between the H and E chondrites, which may be an artefact due to an incomplete sampling of an original more continuous spectrum of chondritic material (Bild and Wasson, 1977). Ungrouped chondrites, especially those showing intermediate compositions between H and E chondrites, may be rare members of such original spectrum (Kallemeyn and Wasson, 1985). Their study may shed light on the condensation/accretion processes occurring in the nebula and the unsampled parent bodies.

Among these rare samples are dark chondritic clasts in the brecciated Cumberland Falls aubrite. These clasts have whole rock chemical compositions similar to those of OCs, but have Mg-rich olivine ($Fa_{0.08-3.66}$) and pyroxene ($Fs_{0.07-14.5}$) compositions (e.g., Neal and Lipschutz, 1981). It was suggested that these clasts are the fragments of an otherwise unsampled “F chondrite” (F for “Forsterite”) parent body which upon a collision with the aubrite parent body led to the formation of Cumberland Falls polymict breccia (Neal and Lipschutz, 1981). Another interpretation suggests that the reduction of a LL chondritic component in presence of highly reduced aubrite host could form clasts with the observed composition (Kallemeyn and Wasson, 1985). Acfer 370 (ungrouped chondrite of petrologic type 3) (Moggi-Cecchi et al., 2009) and Northwest Africa (NWA) 7135 (ungrouped chondrite of petrologic type 3/4) (Irving et al., 2015) are two chondrites that show affinities with Cumberland Falls Chondritic clasts (CFC).

Other chondrites (defined as low-FeO ordinary chondrites) with OC whole rock chemical composition but with olivine and pyroxene richer in Mg than OCs have been described (Wasson et al., 1993). Chondritic clasts (defined as HH chondrites) with reduced olivine and pyroxene compositions and higher concentration of siderophile elements than OCs found inside IIE iron meteorites, are thought to be fragments of a different parent body than H chondrites (Bild and Wasson, 1977; Bogard et al., 2000; Schrader et al., 2017).

Here we report on the petrography, mineral chemistry, whole-rock trace element composition, oxygen-isotope composition, and IR-spectroscopy of El Médano (EM) 301, a chondrite meteorite that is unique amongst other chondrites but shows similarities with NWA 7135, Acfer 370 and the CFC. Thus, NWA 7135 and CFC are also studied during this work.

Comparison with the previously known meteorites gives insights into the existence and origin of chondrites intermediate between H and E chondrites, their formation and evolution.

2. SAMPLES AND METHODS

EM 301 was found in 2013 during a systematic search for meteorites in the Atacama Desert (Chile). It is composed of two stones found less than 1 m apart, and totaling 17.9 g. The whole meteorite is deposited at CEREGE. Magnetic susceptibility was measured on the two pieces using a KLY2 instrument from Agico (Rochette et al., 2003). Thick and thin polished sections were prepared from the two pieces of EM 301 for textural and mineralogical observations. In addition, to compare the textural and mineralogical characteristic, thick sections of NWA 7135 and Cumberland Falls aubrite (section #2840-2 from Muséum National d'Histoire Naturelle-Paris) were examined. Textural and qualitative mineral studies were conducted with a Leica DM2500P optical microscope and a Hitachi S3000-N Scanning Electron Microscope

(SEM) equipped with a Bruker X-ray Energy Dispersive Spectrometer (EDS) at CEREGE (Aix-en-Provence). In addition, a Zeiss Gemini 500 SEM at CP2M (Marseille) was used for semi-quantitative analyses. Chemical compositions of the mineral phases were determined with JEOL JXA-8200 electron microprobe at the Camparis facility (Paris University), using natural and synthetic standards, focused electron beam (~ 1 µm in diameter), an accelerating voltage of 15 kV and a beam current of 25 nA. To correct the deviation of Co content produced by the occurrence of an interference between Co and Fe of the metal grains (Afiattalab and Wasson, 1980), a correction for the Co content of by a factor equal to 0.0012 (Fe concentration ????) is done.

The whole-rock trace-element content of EM 301 was determined by Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) using a Perkin-Elmer NexION® 300x spectrometer at the Pisa University's Dipartimento di Scienze della Terra. The geochemical reference samples with basaltic composition WS-E and PM-S, and the Allende carbonaceous chondrite reference sample (USNM 3529, splite 20, position 22) were dissolved and analyzed along with EM 301 to check the accuracy of the results. About 50-100 mg of each powder were dissolved in a mixture of HF and HNO_3 on a hot plate at ~120 °C inside screw-top perfluoroalkoxy (PFA) vessels. Then the sample solutions were diluted to 50 mL in polypropylene vials. In each step of sample preparation, Mill-Q® purified water (18.2 M cm), ultrapure HF and HNO_3 were used..The sample solutions were introduced into the plasma after online mixing with a solution containing 20 ng/mL each of Rh, Re and Bi as internal standards. The elements Li, Be, Ga, Rb, Sr, Y, Zr, Nb, Mo, Cs, Ba, REE, Hf, Ta, W, Pb, Th, U were determined in "standard mode", whereas the elements Sc, V, Cu were determined in "kinetic energy discrimination mode, KED" using a He flow of 3.7 mL/min. Analyses were done using an external calibration performed with a solution of the BE-N (alkaline basalt) geochemical reference sample. In Table X are reported the results of the ICP-MS analyses of EM 301, Allende (two separate dissolutions), PM-S and WS-E, along with their reference values. In the same table are reported the detection limits for each analyte calculated as three times the standard deviation of the procedural blank concentrations. The analytical precision is between 5 and 10% RSD for elements with concentrations > 0.5 µg/g and between 10 and 20% RSD for elements with concentrations < 0.5 µg/g.

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Reflectance spectra of powders of EM 301 and NWA 7135 were obtained using the spectro-gonio-radiometer at the Institut de Planétologie de Grenoble (Brissaud et al., 2004; Beck et al., 2011).

Measurements of $\delta^{18}O$ and $\delta^{17}O$ of two 1.5 mg aliquot of silicates hand-picked from a powdered and acid-washed 200 mg bulk sample were carried out at the Stable Isotopes Laboratory of CEREGE, by laser fluorination coupled with isotope ratio mass spectrometry (IRMS) technique (Alexandre et al., 2006; Crespin et al., 2008) adapted for measurement of extraterrestrial materials (Suavet et al., 2010). The three oxygen isotopic compositions were measured with a dual-inlet mass spectrometer Thermo-Finnigan Delta Plus. The oxygen isotope results are expressed in ‰ versus the international reference standard V-SMOW: $\delta^{18}O = (^{18}O/^{16}O)_{sample}/(^{18}O/^{16}O)_{V\text{-}SMOW}-1)*1000$ and $\delta^{17}O = (^{17}O/^{16}O)_{sample}/(^{17}O/^{16}O)_{V\text{-}SMOW}-1)*1000$. The $\delta^{18}O$ and $\delta^{17}O$ values of the reference gas were calibrated with measurements of NBS28 standard ($\delta^{18}O$=9.60‰, Gröning, 2004). $\Delta^{17}O$ is computed as $\Delta^{17}O= \ln(1+ \delta^{17}O)- \lambda\ln(1+ \delta^{18}O)$ with λ=0.5247 (Miller, 2002). The $\delta^{17}O$ value of the NBS28 standard ($\delta^{17}O$ =5.026‰) was computed so as to give $\Delta^{17}O$=0‰. The measurements were corrected on a daily basis using 1.5 mg quartz internal laboratory standard "Boulangé" (Alexandre et al., 2006; Suavet et al., 2010). During the analyzing period, the analytical uncertainties derived from repeated measurement (n= 29) of this internal laboratory standard are 0.09‰, 0.17‰, 0.05‰ for $\delta^{17}O$, $\delta^{18}O$ and $\Delta^{17}O$ respectively.

3. RESULTS

3.1. Petrography

3.1.1. EM 301

Due to wind abrasion in the desert environment, both stones are devoid of fusion crust. Cut surfaces of both pieces show a dark brown interior with visible chondrules and metal grains. Optical and electron microscope observations reveal well-preserved and closely packed chondrules and chondrule fragments (Fig. 1a). Modal abundances of chondrules and matrix, measured on a section (n=720), are about 80 vol.% and 20 vol.%, respectively. Different chondrule types are present: porphyritic (POP, PO and PP), cryptocrystalline, radial pyroxene, barred olivine, barred pyroxene, and granular olivine-pyroxene (Fig. 1b). Average apparent chondrule diameter is 502 ± 319 µm (n=99), with a median, mode, and maximum diameters of 400, 300, and ~ 2000 µm, respectively (Fig. 2). Ca-poor pyroxene is the main mineral and Mg-rich olivine, Ca-rich pyroxene, chlorapatite, albitic feldspar, chromite, troilite, (Fe,Ni) metal are the minor phases. Primary (Fe,Ni) metal and sulfides have been extensively (>95%) replaced by Fe oxides/hydroxides by terrestrial weathering, which corresponds to a weathering degree W4 using the ordinary chondrite scheme of Wlotzka (1993). Magnetic susceptibility is log χ = 4.62 (χ in 10^{-9} m^3/kg), but because of the extensive weathering, this value only gives a very lower limit of about 5 vol.% for the initial metal content. Point counting (n=597) under reflected light optical microscopy, with a magnification of X500, yields the following proportions: 66 vol.% silicates, 31 vol.% weathering products, 2 vol.% troilite, 0.7 vol.% (Fe,Ni) metal and 0.5 vol.% chromite. By assuming an average density of 4.5 g/cm^3 for the weathering products (intermediate value between goethite and magnetite), and correcting for the average abundance of troilite in OCs, the initial metal content can be estimated to about 13 wt.%. This value is near the lower limit of the values reported for H chondrites (14.2-19.8 wt.%; Keil, 1962). SEM and transmitted light microscopy of thin section shows that pyroxene is more abundant than olivine (Fig. 1b). Olivine grains show a sharp optical extinction and no strong fracturing, indicating a shock stage S1 (unshocked) using the classification of Stöffler et al. (1991), originally designed for ordinary chondrites. XRF-EDS analysis reveals that pyroxenes are Mg-rich and the abundant presence of type I chondrules (Fig. 3a). Olivine is homogenous in MgO content, whereas the majority of the Ca-poor pyroxene grains show a reverse zoning (compared to the normal silicate zoning observed in the thermally metamorphosed chondrites) with higher amount of MgO in the crystal rims and along cracks (Fig. 3b, c). Chondrule rims contain smaller grains of euhedral crystals (Fig. 3a, d). A pyroxene-dominated chondrule (Fig. 3a, d) shows aligned silica grains, (Fe,Ni) metal and regions differing in the MgO contents. Feldspar (plagioclase) is rare, but reaches up to 10 µm.

Troilite is monocrystalline, anhedral to subhedral in shape, and most often well separated from the (Fe,Ni) metal (Fig. 3e, f). Silicates with absorbed and rounded margins (and in some cases laths) occur inside some of the troilite grains (Fig. 3f). As mentioned, most of the metal grains are replaced by terrestrial weathering products, but those preserved show unusual textures (Fig. 3e, g). Figure 3g displays a metal grain hosting smaller Ni-rich (taenite) euhedral metal crystals. Taenite grains are smaller than 20 µm and show semi-oriented texture. The majority of chondrules in Semarkona (LL3.0), contain metal spherules that show a fine-grained plessitic intergrowth with submicrometer size Ni-rich grains (Kimura et al., 2008). The unique (Fe,Ni) metal texture in EM 301, show similarities with those grains though at a larger scale. Terrestrial alteration of (Fe,Ni) metal grains initiates from the kamacite/taenite contact zone and develops until complete replacement of the kamacite (Fig. 3e). Beside (Fe,Ni) metal grains in the matrix, some Ca-poor pyroxene-dominated chondrules contain frequent (Fe,Ni) metal blebs which are less weathered and show a homogenous texture (Fig. 3d). Chromite is present as euhedral to subhedral grains (Fig. 3f). The matrix is composed of euhedral enstatite laths set in a mixture of different weathering products (Fig. 3h).

3.1.2. NWA 7135

NWA 7135, also described in Irving et al., (2015), shows a very similar texture (Fig. 4). Well-defined chondrules are set within a matrix of enstatite laths set in a background of Fe oxide/oxyhydroxides (Fig. 5a). Apparent chondrule diameter has maximum of 1880 µm and a mean value of 476 ± 271 µm (n=132) (Fig. 2). Median and mode values for the measured chondrules are 400 and 280 µm, respectively. Ca-poor pyroxene is the dominant phase. Neither pyroxene nor olivine show the reverse zoning observed in EM 301. However, some chondrules show zones with different Mg-content and (Fe,Ni) metal blebs, identical to those in EM 301 (Fig. 5b). Except for chromite, the majority of the other opaque phases outside of the chondrules have been altered in the terrestrial environment. As reported in (Irving et al., 2015), we observed a few preserved grains of oldhamite and daubréelite.

3.1.3. CFC

The studied thin section of Cumberland Falls consists of white enstatite-rich aubrite host and a dark colored chondritic clast (Fig. 6). The highly shocked nature of the clast is evidenced by the presence of troilite melt veins and droplets, and by shock darkening. The texture of the clast and the visual (??) structure of the silicates does not show similarities to the textures of EM 301 or NWA 7135. Showing a chondrule size ranging from 500 to 2000 µm (Rubin, 2010a), CFC generally have larger chondrule sizes than EM 301 and NWA 7135. An observation that cannot be attributed to thermal metamorphism (e.g., Schrader et al., 2017) as most of the studied CFC clasts are only very slightly metamorphosed (type 3) (Binns, 1969; Neal and Lipschutz, 1981).

3.2. Mineral Chemistry

Table 1, 2 and 3 report the chemical compositions of olivine, low-Ca pyroxene and (Fe,Ni) metal of EM 301. Low-Ca pyroxene shows a range of chemical compositions, whereas olivine has a relatively narrow chemical distribution (Fig. 7). Average olivine (n=19) and orthopyroxene (n=14) compositions are $Fa_{3.9 \pm 0.3}$ and $Fs_{12.4 \pm 4.8}$, respectively. As shown in Fig. 8, these values are far from the olivine and low-Ca pyroxene chemical composition ranges of ordinary chondrites as well as those reported in chondrites and chondritic clasts described as HH, low-FeO, and reduced ordinary chondrites. Together with CFCs, NWA 7135 and Acfer 370, it forms a well-separated cluster. Compared to Acfer 370 and NWA 7135, EM 301 shows more reduced ferromagnesian silicate compositions. Olivine and low-Ca pyroxene show percent mean deviations (PMD) of 4.4% and 44%, respectively, which suggest a petrologic type 4. The Co concentration of kamacite (Table 3) is below detection limit (0.18 wt.%).This is well below the observed concentrations for H (0.44-0.51 wt%), L (0.70-0.95 wt%) and LL (0.42-37 wt%) chondrites (Rubin, 1990). The same with Si, the analyzed kamacite grains contain less than detection limit (0.04 wt.%) of this element.

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3.3. IR-Spectroscopy

The infrared reflectance spectrum of EM 301 and NWA 7135 were obtained on powdered samples leached with HCl to remove weathering products (oxyhydroxides) that otherwise would dominate the spectrum. The spectrum reveals the occurrence of two strong absorptions around 0.92 µm (Band I) and 1.9 µm (Band II) (Fig. 9). The presence and position of these two bands are typical of a pyroxene signature (Cloutis and Gaffey, 1991). Addition of olivine, would tend to decrease the area of the 1.9 µm feature and to shift the band center of Band I toward higher wavelength, as is observed for ordinary chondrites.

3.4. Trace Element Bulk Chemistry

The whole rock chemical composition of EM 301 is reported in Table 4. The CI-normalized trace-element pattern shows enrichments of Ba (× 43.6), Sr (× 3.2) and LREE (up to × 2.4 for La), which are indicative of terrestrial weathering (Pourkhorsandi et al., 2017). Keeping in mind these effects, the trace-element contents are in the range of ordinary chondrites (Fig. 10; Wasson and Kallemeyn, 1988).

3.5. Oxygen-Isotope Composition

Oxygen-isotope composition analyses yielded the following results: $\delta^{17}O$ = +3.61, +3.78‰, $\delta^{18}O$ = +5.38, +5.71‰, $\Delta^{17}O$ = +0.79, +0.78‰. Figure 11 depicts the $\Delta^{17}O$ versus $\delta^{18}O$ values of EM 301 along with literature data for H, L, and LL chondrites, and the chondrites/clasts with more reduced compositions than the ordinary chondrites. EM 301 shows higher $\delta^{18}O$ values than those of H chondrites and together with NWA 7135, Burnwell, Suwahib (Buwah) and the CFCs forms a different cluster. Since the samples were acid washed and clean hand-picked silicate crystals were used for the analyses, we believe that the effect of the terrestrial alteration on the oxygen-isotope composition of EM 301 is insignificant.

4. DISCUSSION

4.1. Classification

Whole rock trace-element composition of EM 301 shows an affinity to OCs. The average apparent chondrule size of EM 301 is intermediate between values for H (Weisberg et al., 2006) and L (Rubin, 2010b) chondrites. However, a higher pyroxene/olivine ratio, higher MgO contents of olivine and pyroxene, lower Co contents of kamacite, and different oxygen-isotopic composition, hinders its classification as a member of any of the typical OC groups (H, L, LL). In fact, the chemical range of the ferromagnesian minerals (olivine and pyroxene) in EM 301 is much closer to ECs than OCs. From a spectral point of view, it does not show similarities to ordinary chondrites and its spectrum is closer to that of howardites, i.e. V-type related material (Fig. 12).

Whole rock trace-element composition, oxygen-isotopic composition, and chemistry of olivine/pyroxene/kamacite shows similarities to CFC, Acfer 370, and NWA 7135. However, its general texture (e.g., chondrule size) is different from CFC and the highly reduced opaque minerals observed in NWA 7135 are absent in EM 301.

Chondrites with affinities to OCs but with higher contents of MgO in their ferromagnesian minerals have been reported with different terms such as, “HH” (Bild and Wasson, 1977), “reduced OCs” (Wasson et al., 1993), and “low-FeO” (Russel et al., 1998; Troiano et al., 2011) (hereafter we will use “reduced chondrites” for all these). Beside these, the term “forsterite chondrites” has been used to designate samples such as CFC (Graham et al., 1977) and NWA 7135 (Kuehner et al., 2015) whose ferromagnesian minerals have even higher MgO contents (similar to ones in EM 301). The problem with using this name for EM 301, is its textural and opaque mineralogical differences with CFC and NWA 7135, and also the poorly discriminant character of this term, since forsterite is abundant in many chondrite groups. Eventually, we use a "highly reduced ordinary chondrite" for EM 301 in the following.

4.2. Origin of EM 301 chondrite

4.2.1. Introduction

To investigate the origin of EM 301, we compare it with OCs, ECs, and the reduced chondrites. Based on the mean Fa (mole%) and Fs (mole%) contents, OCs are divided into H ($Fa_{16.0-20.0}$; $Fs_{14.5-18.0}$), L ($Fa_{22.0-26.0}$; $Fs_{19.0-22.0}$) and LL ($Fa_{26.0-32.0}$; $Fs_{22.0-26.0}$) groups (Brearley and Jones, 1998). Olivine is the dominant mineral in OCs and its chemical composition along with the Co concentration of kamacite is an accepted proxy of the oxidation-reduction state of the sample (Rubin, 1990). FeO content of olivine and Co concentration in kamacite, with increasing oxidation, increase from H to L, and LL chondrites. By formation of (Fe,Ni) metal (kamacite/taenite) in a reducing environment (low-fO_2), FeO content of the silicate phases decreases and in contrast, in a more oxidant environment (high-fO_2), iron is incorporated in silicates. As a result, in low-fO_2 conditions, ferromagnesian silicates are richer in MgO and the concentration of Co (and Ni) is lower in kamacite grains (Rubin, 1990). The extreme effect of formation in an environment with low-fO_2 is observed in ECs, in which almost pure enstatite is the dominant silicate phase and the majority of the Fe forms kamacite grains, as well as a variety of reduced minerals (e.g. carbides, phosphides, Ca or Mg sulfides, Keil, (1989)).

4.2.2. The reduced chondrites

As mentioned in Sec. 4.1., beside typical OCs and ECs, some chondrites show intermediate characteristics, still with stronger affinities with OCs. Such reduced chondrites (relative to OCs), occur in at least three distinct "clusters": 1) low-FeO, 2) HH, and 3) CFC.

Based on the MgO-rich olivine/pyroxene and oxygen-isotopic compositions of Willaroy ($Fa_{14.1}$; $Fs_{13.3}$) and Suwahib (Buwah) ($Fa_{13.5}$; $Fs_{13.2}$), Scott et al. (1985) interpreted these meteorites as representatives of a distinct chondritic group. Cerro los

Calvos, with similar characteristics ($Fa_{12.5}$; $Fs_{11.7}$), was classified as an H chondrite defining the extreme limit of H compositional field (Whitlock et al., 1991). Reduction of normal H and L chondritic material during thermal metamorphism within the parent body in the presence of a reducing agent (e.g., graphite) was suggested by (Wasson et al., 1993) to account for the formation of these chondrites as well as of Moorabie ($Fa_{15.9}$; $Fs_{15.3}$). In contrast, based on evidences such as the low Co content of kamacite, and the lack of a reducing agent, McCoy et al. (1994) proposed their formation in nebular rather than asteroidal setting. The lack of clasts with characteristics similar to low-FeO chondrites in H chondrites made them to consider these samples originating from different objects than the H chondrites parent body(ies). Burnwell anomalous H4 chondrite ($Fa_{15.8}$; $Fs_{13.4}$), is a relatively large reduced chondrite (measuring 15.5 × 7 × 5 cm), much larger than the size of the reduced clasts in aubrites and IIE irons. Russel et al. (1998) suggest the necessity of a period of intense parent body thermal activity to reduce a volume of rock with the size of Burnwell with a reducing process similar to that proposed by Wasson et al. (1993). However, the occurrence of such high temperature period is in conflict with the low petrologic degree of Burnwell. Therefore, they consider a nebular origin for the reduced nature of Burnwell. Other studies of low-FeO chondrites, also propose either nebular or parent body origins (Troiano et al., 2011; Yamaguchi et al., 2015).

IIE irons (such as Netschaëvo, Techado, Garhi Yasin) host chondritic clasts with lower Fa and Fs ($Fa_{14.3}$, $Fs_{14.0}$; in Netschaëvo), distinct chromite composition, lower Co content in kamacite, and different oxygen-isotopic composition compared to H chondrites (Bild and Wasson, 1977; McDermott et al., 2016; Schrader et al., 2017). Based on these properties and the higher concentration of siderophile elements in whole rock chemical composition, Bild and Wasson, (1977) named them HH chondrites and proposed an origin from a distinct parent body.

The chondritic clasts of Cumberland Falls are mostly of petrologic type 3 or 4 (Binns, 1969), but type 6 clasts and impact melt clasts have also been reported (Rubin, 2010a; Kuehner et al., 2016). The whole rock chemistry of CFC is in the range of LL chondrites (Kallemeyn and Wasson, 1985). However, unlike in OCs, pyroxene is the dominant silicate phase in CFC and their olivine/pyroxene and kamacite chemistry is different from OCs. In addition, their oxygen-isotopic composition is also different from OCs and ECs. Two different alternatives have been given to their formation: 1) condensation in a nebular region more reduced than the OCs formation region and thus provenance from a distinct parent body (Graham et al., 1977; Neal and Lipschutz, 1981; Verkouteren and Lipschutz, 1983; Grady and Pillinger, 1986; Lipschutz et al., 1988; Keil, 2010; Kuehner et al., 2016), 2) reduction of LL chondritic material in the presence of the reducing aubrite host after an impact event and the later modification in mineral and oxygen-isotopic composition (Wasson and Kallemeyn, 1984; Kallemeyn and Wasson, 1985; Rubin, 2010a).

4.2.3. EM 301

Chemical and oxygen-isotopic composition, and general texture of EM 301 indieate its affinity to OCs. Still, there are significant difference between EM 301 and OCs. They include: 1) lower modal olivine/pyroxene ratio, 2) lower Co content of kamacite, 3) higher MgO contents of olivine and pyroxene, 4) different kamacite texture, and 5) different oxygen-isotopic composition.

Similar oxygen-isotopic ratios and ferromagnesian minerals composition indicate a close relationship between EM 301, NWA 7135, CFC, and Acfer 370. However, there are some differences among these meteorites. The occurrence of daubréelite, oldhamite, schreibersite, and djerfisherite (reported by Irving et al., 2015) in NWA 7135 indicates its formation in very reduced conditions. Similar minerals are reported from CFC (Rubin, 2010a). In contrast, despite a low weathering degree (W1) in some parts, none of these minerals are reported in Acfer 370 (Moggi-Cecchi et al., 2009). During SEM survey of EM 301, we did not find any of these mineral either. However, owing to the strong weathering, their preservation (if they ever existed) would have been unlikely.

The texture and silicate mineralogy of EM 301 and NWA 7135 show some resemblance to ECs. The matrix is composed of enstatite laths set in an iron oxide/oxyhydroxide mélange of weathering products. This texture is somehow similar to a “remnant” of the “metal/sulfide-silicate intergrowths” observed in ECs, which form as a result of impact melting in a reduced lithology (Lin and Kimura, 1998; van Niekerk and Keil, 2006; Rubin and Wasson, 2011; Horstmann et al., 2014). However, the preserved kamacite grains do not host such enstatite laths which rejects the idea of their possible relevance to "metal/sulfide-silicate intergrowths". Kamacite is Si-poor (below detection limit of 0.04 wt%) in EM 301, and also in CFC (Rubin, 2010a), and NWA 7135 (Irving et al., 2015), which is in contrast to their Si-rich nature in ECs (up to about 3.0 wt.% Si in the metal of EH3s and up to about 1.4 wt.% in EL3s) (Weisberg et al., 1995). In addition, olivine is a rare silicate in ECs (e.g., Keil, 1989), but abundant in the mentioned chondrites. The olivine and pyroxene composition of EM 301 is close to the reported values for the Kakangari chondrites (Graham et al., 1977; Weisberg et al., 1996), but the matrix/chondrule ratio, oxygen-isotopic composition and whole rock chemistry are different. Putting all the data together, we observe the effects of low-fO_2 conditions (compared to OCs) during formation/evolution of EM 301. These conditions could have prevailed in the nebula and/or the parent body (asteroidal processes).

The solar nebula contained regions with different level of fO_2 (Larimer and Bartholomay, 1979; Rubin and Wasson, 1995; Grossman et al., 2008). It is believed that the formation region of ECs had a C/O ratio higher than the solar value (Larimer and Bartholomay, 1979). The presence of reducing C-rich phases, such as organic matter and graphite, resulted in the formation of the observed EC mineralogy (high modal pyroxene/olivine ratio, Mg-rich nature of the mafic minerals, etc.).

However, some processes in the parent body can also lead to the reduction. Among these are: impact events and the resulting melting (Horstmann et al., 2014), impact and reduction in the presence of reduced material in the host (Rubin, 2010a), and the reducing effect of C-rich gases during parent body degassing (Sugiura et al., 1985). As being proposed for the formation of CFC (Kallemeyn and Wasson, 1985), and low-FeO chondrites (Wasson et al., 1993), parent body reduction can change the oxidation state of the minerals forming Mg-rich ferromagnesian silicates and Co-poor kamacite (Wasson et al., 1993). An inverse process (parent body oxidation), is

Commented [MD3]: We never give an explanation of the Co-poor kamacite found in reduced chondrites proposed to describe the occurrence of EH clasts in Galim LL/EH polymict breccia (Rubin, 1997), which show similar composition to CFC.

We suggest that EM 301 formed in a nebular location with fO_2 intermediate between OCs and ECs. Indeed, EM 301 is devoid of shock metamorphic features (no shock veins, polycrystalline troilite, native copper, fractures in ferromagnesian minerals) and of traces of melting or annealing (low metamorphic grade) to transform the olivine to pyroxene during a high temperature period. This points to a nebular rather than asteroidal origin for the primary characteristics of EM 301.

Olivine and pyroxene can record a secondary reduction event as reverse zoning (Mg-rich rims) (Weisberg et al., 1994; Goodrich et al., 2006; Rubin, 2010a) and higher mean Fs to higher mean Fa ratio (Wasson et al., 1993; Keil, 2007; Rubin, 2010a). The outer rims and cracks walls in pyroxene grains in EM 301 are Mg-rich (Fig. 3b, c). A reverse zoning along a Ca-poor pyroxene is shown in Fig. 12. This can be an evidence of a secondary reduction event. As a consequence of material mixing and injection of reducing components to the formation region of the grains in the nebula (e.g., Fegley, Jr., 2000; Bockelée–Morvan et al., 2002; Zanda et al., 2006) or while releasing reducing gases (especially C-rich agents) to the shallower regions of the parent body through the high permeability zones and along the cracks during the degassing of a parent body after its accretion (Sugiura et al., 1984; Sugiura et al., 1985; Sugiura et al., 1986; Krot and N., 1994; Hashizume and Sugiura, 1998). It is noteworthy that formation of EM 301 in a region more reduced than the one of the OCs means a higher abundance of C-rich material in the resulting parent body and higher amount of reducing gasses during asteroidal processes (such as metamorphism). Considering its relatively unequilibrated character and low petrologic type, and the necessity for high degrees of thermal metamorphism for parent body reduction (e.g., Russel et al., 1998; Schrader et al., 2017), a secondary nebular reduction event is more likely than a one occurring in the parent body (Rubin, 2017; Schrader et al., 2017).

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Olivine coexisting in equilibrium with pyroxene has a higher content of FeO than pyroxene (e.g., Keil and Fredriksson, 1964). With increasing reduction, Fe forms metallic phases and available FeO to incorporate in olivine and pyroxene decreases and in result mean Fa and Fs contents become lower than the original precursor. Because diffusion in olivine is faster than pyroxene (e.g., Freer, 1981), during a subsequent reduction event, it equilibrates faster than pyroxene. If the equilibrium does not reach completion, the pyroxene grains show wider compositional range and retain more ferrous compositions than olivine. This is what is observed in EM 301 and a secondary reduction event can be responsible for that. The same idea is suggested for CFC (Wasson et al., 1993; Rubin, 2010a), and could also apply to NWA 7135 and Acfer 370.

Contrary to CFC where oxygen-isotopic composition might have been shifted by impact and asteroidal reduction (Wasson and Kallemeyn, 1984), this scenario is unlikely for EM 301. A study by Tait et al., (2014) on Watson 012 H7 chondrite (which is slightly more reduced than typical H chondrites), reveals an oxygen-isotopic composition close to EM 301. Intense thermal metamorphism and associated mass dependent fractionation has been suggested to be responsible for such a shift in oxygen-isotopic composition, but this cannot be the case for type 4 EM 301.

We suggest that EM 301 formation region in the nebula had a trace element composition pattern similar to OCs, but fO_2 intermediate between OCs and ECs. This reducing condition (in comparison with OCs) has resulted in the formation of a higher modal pyroxene/olivine ratio, Mg-rich olivine and pyroxene, different oxygen-isotopic composition, and, for NWA 7135, formation of highly reduced opaque phases. During a secondary reduction event which might have happened in the nebula, Mg-rich rims have formed in EM 301 and also the mean Fs has become higher than the mean Fa, which is also observed in NWA 7135, CFC, and Acfer 370. A reason for the lack of reverse zoning in pyroxene of NWA 7135 may be more intense reduction and associated diffusion of Fe-Mg, similar to the chemically homogenous olivine and pyroxene in the highly reduced acapulcoites and lodranites (Rubin, 2007).

Recent studies on chondrites with compositions intermediate between H and E chondrites have suggested their formation as nebular condensates. For instance, Weisberg et al. (2015) described the occurrence of such chondrites (termed “G chondrites”) which have however oxygen-isotopic composition different from EM

301. Rubin et al. (2016) describe a “new kind” of OC, occurring in a LL3 breccia, with mean $Fa_{13.6}$ and $Fs_{4.3}$, that because of being in a more oxidized host, is a nebular product rather than being reduced during the impact.

4.2.4 Possible parent body

The reflectance spectrum of EM 301 is quite different from that of ordinary chondrites. This difference is explained by a higher pyroxene/olivine ratio, but also by the low Fe content of olivine ($Fa_{3.9 \pm 0.3}$), which makes it almost spectrally neutral. The reflectance spectrum of EM 301 is similar to that of material typically interpreted as differentiated: it resembles typical spectra of HED meteorites (Fig. 13). The position of Band I and Band II of EM 301 (0.92 and 1.92 µm) are typical of Fe-poor pyroxene. They are reminiscent of, but not identical to values found for 4-Vesta (on average 0.93 µm and 1.96 µm; De Sanctis et al., 2012)).

While the spectrum of the surface of 4-Vesta is not a perfect match of the spectrum of EM 301 (and while we now have the confirmation from the DAWN mission that 4-Vesta is related to HED meteorites), there are a number of V-type asteroids (with spectra similar to 4-Vesta) that do not have a dynamical affinity with 4-Vesta and could sample a different parent body (Nesvorný et al., 2008). The spectra of V-type asteroids show the presence of both Band I and II, with a high Band II to Band I ratio, (i.e. spectra dominated by pyroxene). Several V-type families have been distinguished: i) vestoids (with dynamical affinities with Vesta) ii) fugitives (with a<2.3 a.u. but inclination and eccentricity similar to vestoids, Nesvorný et al., 2008) iii) low-inclination V-types (with 2.3 a.u. < a < 2.5 a.u, and i<6°, according to Nesvorný et al., 2008)) iv) Near-Earth Asteroid with V-type spectra and last v) Middle or Outer Belt V-type, MOV (Ieva et al., 2016). Among all these V-types, an important variability is present in the Band I and Band II positions (from 0.90 to 0.95 µm for Band I and from 1.89 to 2.05 µm for Band II). This variability includes objects with band positions similar to those measured for EM 301. While vestoids and fugitives are most likely related to HED meteorites, a number of V-types might in fact be related to chondritic material similar to EM 301.

5. CONCLUSION

EM 301 is a chondrite containing Mg-rich olivine and pyroxene with average compositions of $Fa_{3.9 \pm 0.3}$ and $Fs_{12.4 \pm 4.8}$, respectively, which is intermediate between ordinary and enstatite chondrites. Its oxygen isotopic composition is different from other ordinary chondrites. It shows a whole rock trace-element composition similar to OCs. However, its olivine/pyroxene modal abundance, and olivine/pyroxene/kamacite chemical composition suggest its formation in nebular region with lower fO_2 than OCs. Mg-rich rims in pyroxene and higher mean Fs/mean Fa ratio suggest the occurrence of a secondary reduction event, which probably has occurred in the nebula. The general similarities between EM 301 and NWA 7135 and probably CFC and Acfer 370, suggests that these meteorites may have formed in region with relatively similar physico-chemical conditions and more reduced than OCs.

The IR spectra of EM 301 is markedly different from those of typical ordinary chondrites. The spectrum is dominated by a pyroxene signature. Similar spectra when observed among main-belt asteroids are usually interpreted as "basaltic", as is the case of V-type asteroids. The presence of chondritic material with pyroxene-like reflectance spectra suggests that a number of V-type, in particular those that are not dynamically related to Vesta, might in fact be chondritic.

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Table headings:

Table 1: Representative olivine compositions (in wt.%) from EM 301.

Table 2: Representative and low-Ca pyroxene compositions (in wt.%) from EM 301.

Table 3: (Fe,Ni) metal compositions (in wt.%) from EM 301.

Table 4: Whole rock trace element composition (in µg/g) of EM 301.

Figure captions:

Fig.1: Full section optical mosaic images of EM 301. a) Thick polished section in reflected light showing the terrestrial weathering products as light grey patches and veinlets. Dark grey portions are silicates and white spots are the (Fe,Ni) metal grains. b) Thin section in cross polarized light showing the chondritic textures. Porphyritic chondrules with higher abundance of pyroxene (low birefringence colors) are the dominant chondrule types. The other chondrule types based on their abundance are: radial pyroxene, granular, cryptocrystalline, and barred olivine.

Fig. 2: Size frequency distribution of the chondrule diameters in EM 301 and NWA 7135. Horizontal axis values mark the upper limits of the size bins.

Fig. 3: Electron and optical microscope images of EM 301. a) Backscattered electron (BSE) image showing the higher abundance of Ca-poor pyroxene in the chondrules and matrix. Few olivine grains (darker grains) are visible. Note the occurrence of chondrule with different MgO and FeO concentration contents. .b) BSE image showing mostly elongated enstatite with Mg-rich rims in. c) BSE image showing reverse zoning in Ca-poor pyroxene. Yellow bar shows the location of the chemical profiles shown in Fig. 12. d) BSE image of a pyroxene dominated chondrule, showing region with different MgO contents and silica and (Fe,Ni) metal blebs. Note the MgO regions along the rim and a major crack. e) Reflected light image showing that Ni-rich metal is more resistant than Ni-poor metal to the terrestrial weathering. f) Troilite, (Fe,Ni) metal and chromite constitute the main opaque phases in El Médano 301. Majority of the (Fe,Ni) metal is weathered to Fe oxides (reflected light image). g) Some (Fe,Ni) metal grains in EM 301, show the exsolution of two Ni-rich and Ni-poor metal phases (BSE image). h) An RGB image of EM 301 with Al in red, Si in green, and Mg in blue channels, respectively. The dominance of Ca-poor pyroxene is clear in this image.

Fig. 4: Full thick section optical mosaic image of NWA 7135 in reflected light showing the chondritic textures and severe terrestrial weathering as evidenced by Fe oxides/oxyhydroxides mineral assemblages shown as (light grey patches and veinlets). Dark grey portions are silicates and rare white to yellowish spots (right up side for example) are the (Fe,Ni) metal and troilite grains.

Fig. 5: BSE images of NWA 7135 showing a) the dominance of Mg-rich pyroxene; b) a pyroxene dominated chondrule showing regions with different MgO contents and silica and (Fe,Ni) metal blebs. Note the MgO-rich regions along the rim and a major crack. The white regions in the matrix and along the cracks are terrestrial weathering products. Metal grains occur as white spots inside the silicate grains.

Fig. 6: Full thick section reflected light image of Cumberland Falls (section #2840-2 from MNHNP). Ca-poor pyroxene is the main component of the aubrite host. The darker chondritic region constitutes chondrules, (Fe,Ni) metal, and troilite grains that in most cases form shock assemblages indicative of an impact event.

Fig. 7: Histograms showing the compositional distributions of randomly chosen a) low-Ca pyroxene (n=14) and b) olivine (n=19) grains in EM 301.

Fig. 8: Scatter diagram showing the average Fa content of olivine (mol%) in El Médano 301, CFC, reduced, and OCs. Density contour plots drawn using petrologic type ≥ 3.9, H (n=4696), L (n=2040) and LL (n=360) chondrites. For Cumberland Falls, average values for different clasts are shown (data from Neal and Lipschutz, 1981). The mean compositional values of OCs from MetBase (Koblitz, 2005), and reduced chondrites and chondritic clasts (both shown as hexagons) are plotted for comparison. The plotted reduced chondritic meteorites and clasts are: LaPaz Icefield (LAP) 04757 and 04773 (Connolly et al., 2007), Suwahib (Buwah) and Willaroy (Scott et al., 1985), Cerro los Calvos and Moorabie (Wasson et al., 1993), Burnwell (Russel et al., 1998), Allan Hills A77221 (Grossman, 1994), Elephant Moraine (EET) 96031 (Grossman, 1998), Beni Semguine (Grossman, 1999), Miller Range 07273 (Weisberg et al., 2010), Yamato 982717 (Ruzicka et al., 2015), Garhi Yasin clast (McDermott et al., 2016), Techado and Netschaëvo clasts (Van Roosbroek et al., 2015), NWA 10214 unique clast (Rubin et al., 2016), Hammadah al Hamra (HaH) 180 (Grossman, 1997), Acfer 370 (Moggi-Cecchi et al., 2009) and NWA 7135 (Irving et al., 2015).

Fig. 9: IR spectra of EM 301 and NWA 7135.

Fig. 10: CI-normalized REE chemical composition of EM 301 is identical to the mean composition of OCs. Mean OCs data from (Wasson and Kallemeyn, 1988).

Fig. 11: $\Delta^{17}O$ versus $\delta^{18}O$ values of EM 301 compared to type 4-6 OCs and the reduced chondrites. Data list and references are: OCs (Clayton and Mayeda, 1991), CFC (Kuehner et al., 2016), NWA 7135 (Kuehner et al., 2015), Acfer 370 (Moggi-Cecchi et al., 2009), Burnwell (Russel et al., 1998; Troiano et al., 2011), LAP 04757 (Connolly et al., 2007; Troiano et al., 2011), EET 96031 (Troiano et al., 2011) Suwahib (Buwah) and Willaroy (Scott et al., 1985), Garhi Yasin, Techado and Netschaëvo chondritic clasts (McDermott et al., 2016). $\Delta^{17}O$ values are calculated using a slope of 0.52.

Fig. 12: Higher Mg (wt.%) and lower Fe (wt.%) contents in a representative low-Ca pyroxene (shown in Fig. 3c) in EM 301 are the representatives of a reverse zoning.

Fig. 13: A comparison of the IR spectra of IR spectra of EM 301 with V-type asteroids, H chondrites and an howardite.

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Table EA-1: XXX.

Table 1 (Pourkhorsandi et al.)

SiO_2	**FeO**	**MnO**	**MgO**	**Total**	**Fa (mol.%)**
41.5	3.65	0.52	55.78	101.61	3.54
42.48	3.60	0.58	53.70	100.41	3.63
41.73	3.75	0.50	55.68	101.69	3.64
42.06	3.76	0.54	55.38	101.92	3.67
42.47	3.67	0.57	53.52	100.42	3.71
43.29	3.70	0.47	53.31	100.89	3.75
42.77	3.76	0.45	54.14	101.34	3.75
41.8	3.91	0.57	56.06	102.52	3.77
42.68	3.79	0.50	53.64	100.78	3.81
41.79	4.05	0.55	56.46	103.08	3.87
42.45	3.98	0.56	54.92	102.39	3.9
42.4	4.08	0.45	56.14	103.19	3.91
41.21	4.02	0.46	55.46	101.23	3.91
43.5	3.88	0.53	53.42	101.47	3.92
43.2	3.93	0.51	54.01	101.75	3.92
43.49	3.94	0.44	53.60	101.56	3.96
42.74	3.91	0.43	53.01	100.27	3.97
41.95	4.92	0.46	52.76	100.4	4.97
41.93	4.28	0.56	55.58	102.66	4.14

bdl = below detection limit.

Detection limits (%wt) = Si (0.07), Al (0.06), Ti (0.13), Fe (0.16), Cr (0.07), Mn (0.14), Mg (0.07), Ca (0.05), Ni (0.18).

The concentrations of Al_2O_3, TiO_2, Cr_2O_3, CaO, are below detection limit.

Table 2 (Pourkhorsandi et al)

SiO2	Al_2O_3	TiO_2	FeO	Cr_2O_3	MnO	MgO	CaO	Total	Fs (mol.%)
59.34	0.90	0.13	3.71	0.42	0.71	35.49	0.61	101.83	5.47
60.43	0.07	bdl	4.70	0.08	0.50	36.59	0.19	102.69	6.7
59.77	0.10	0.14	4.69	bdl	0.59	36.21	0.30	101.87	6.74
57.61	0.14	0.22	5.48	0.50	0.46	36.73	0.43	101.59	7.67
57.19	bdl	0.13	5.44	0.08	0.68	36.19	0.44	100.33	7.72
54.31	0.33	bdl	7.54	0.49	0.70	37.99	0.38	101.89	9.95
59.13	0.12	0.12	7.60	0.18	0.58	33.39	0.47	101.69	11.23
58.86	0.45	bdl	9.47	0.27	0.51	31.54	0.46	101.72	14.29
57.13	0.22	bdl	11.08	0.36	0.27	32.47	0.47	102.17	15.94
55.22	0.26	bdl	11.12	0.19	0.55	32.09	1.01	100.56	15.97
56.88	0.57	bdl	10.96	0.40	0.39	30.48	0.65	100.39	16.57
58.32	bdl	bdl	11.27	0.21	0.49	31.06	0.24	101.68	16.83
56.41	0.53	bdl	11.87	0.51	0.61	29.69	0.75	100.41	18.05
56.53	0.21	bdl	13.36	0.41	0.51	28.87	0.73	100.73	20.32

bdl = below detection limit.

Detection limits (%wt) = Si (0.07), Al (0.06), Ti (0.13), Fe (0.16), Cr (0.07), Mn (0.14), Mg (0.07), Ca (0.05), Ni (0.18).

The concentrations of Na_2O, K_2O, P_2O_5, Ni, and S are below detection limit.

Table 3 (Pourkhorsandi et al.)

	Kamacite	**Kamacite**	**Kamacite**	**Kamacite**	**Kamacite**	**Kamacite**	**Kamacite**	**Kamacite**	**Taenite**	**Taenite**	**Taenite**
Si	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.04	bdl
S	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Cr	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	0.08	0.07
Fe	92.74	90.85	92.93	91.00	91.90	91.49	90.48	91.30	71.81	45.33	44.36
Co	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ni	5.71	5.79	5.99	6.15	6.17	6.50	6.55	6.70	25.19	52.25	52.94
Total	98.51	96.71	99.01	97.25	98.06	98.02	97.05	98.09	97.00	97.65	97.36

bdl = below detection limit.

Detection limits (%wt) = Si (0.04), S (0.04), Cr (0.07), Fe (0.21), Co (0.18), Ni (0.18).

Table 4 (Pourkhorsandi et al.)

Li	**Be**	**Sc**	**V**	**Cu**	**Ga**	**Rb**	**Sr**	**Y**	**Zr**	**Nb**	**Mo**	**Cs**	**Ba**	**La**	**Ce**	**Pr**	**Nd**	**Sm**	**Eu**	**Gd**	**Tb**	**Dy**	**Ho**	**Er**	**Tm**	**Yb**	**Lu**	**Hf**	**Ta**	**W**	**Pb**	**Th**	**U**
2.65	0.04	5.50	41	72.0	5.9	2.71	23.1	2.04	5.9	0.37	4.10	0.516	105.0	0.609	1.563	0.188	0.828	0.229	0.087	0.311	0.054	0.341	0.074	0.222	0.033	0.208	0.033	0.170	0.025	0.3	0.97	0.115	0.061