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## Isotopic composition of carbon and nitrogen in ureilitic fragments of the Almahata Sitta meteorite

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**Abstract**—This study characterizes carbon and nitrogen abundances and isotopic compositions in ureilitic fragments of Almahata Sitta. Ureilites are carbon-rich (containing up to 7 wt% C) and were formed early in solar system history, thus the origin of carbon in ureilites has significance for the origin of solar system carbon. These samples were collected soon after they fell, so they among the freshest ureilite samples available and were analyzed using stepped combustion mass spectrometry. They contained 1.2–2.3 wt% carbon; most showed the major carbon release at temperatures of 600–700 °C with peak values of  $\delta^{13}\text{C}$  from  $-7.3$  to  $+0.4\text{‰}$ , similar to literature values for unbrecciated (“monomict”) ureilites. They also contained a minor low temperature ( $\leq 500$  °C) component ( $\delta^{13}\text{C} = \text{ca } -25\text{‰}$ ). Bulk nitrogen contents (9.4–27 ppm) resemble those of unbrecciated ureilites, with major releases mostly occurring at 600–750 °C. A significant lower temperature release of nitrogen occurred in all samples. Main release  $\delta^{15}\text{N}$  values of  $-53$  to  $-94\text{‰}$  fall within the range reported for diamond separates and acid leached material from ureilites, and identify an isotopically primordial nitrogen component. However, they differ from common polymict ureilites which are more nitrogen-rich and isotopically heavier. Thus, although the parent asteroid 2008TC<sub>3</sub> was undoubtedly a polymict ureilite breccia, this cannot be deduced from an isotopic study of individual ureilite fragments. The combined main release  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values do not overlap the fields for carbonaceous or enstatite chondrites, suggesting that carbon in ureilites was not derived from these sources.

### INTRODUCTION

Ureilites are ultramafic achondrite meteorites that are considered to represent the mantle residue of a single parent asteroid body (Takeda 1987; Scott et al. 1993; Goodrich et al. 2004; Downes et al. 2008; Herrin et al. 2010). As such they can be considered as analogues of the early mantle of terrestrial planets. Ureilites are the most carbon-rich of all meteorite types, typically containing 2–7 wt% C (Mittlefehldt et al. 1998; Smith et al. 2001a; Grady and Wright 2003;

Nakamuta 2005). Carbon occurs in ureilites as graphite, diamond and amorphous carbon (Vdovykin 1970; Berkley et al. 1976; Mittlefehldt et al. 1998; Hezel et al. 2008; Le Guillou et al. 2010; Ross et al. 2011a), with other rare carbon polymorphs (El Goresy et al. 2004; Ferroir et al. 2010; Kaliwoda et al. 2011) and carbide phases, e.g., cohenite (Goodrich and Berkley 1986; Warren and Kallemeyn 1994; Goodrich et al. 2013). Other compounds, such as carbonate weathering products (Grady et al. 1985) are likely to represent terrestrial contamination, whereas organic molecules

1 such as those found by Glavin et al. (2010), Sabbah  
2 et al. (2010) and Burton et al. (2011) are thought to be  
3 extraterrestrial but unlikely to be derived from the  
4 ureilitic precursor.

5 Despite occasionally occurring in veins (e.g., Corder  
6 et al. 2014), ureilitic carbon is usually intergranular and  
7 found on grain boundaries. It is considered to be a  
8 primary constituent (Berkley and Jones 1982; Rubín  
9 1988) rather than having been introduced late in the  
10 formation history of the meteorites. Carbon abundances  
11 are commonly higher in ureilites with low core olivine  
12 Fo contents (Nakamuta 2005). Diamond in ureilites is  
13 usually considered to have formed via shock  
14 metamorphism from graphite (Lipschutz 1964; Bischoff  
15 et al. 1999; Grund and Bischoff 1999; Nakamuta and  
16 Aoki 2000; El Goresy et al. 2004; Fisenko et al. 2004;  
17 Hezel et al. 2008; Ross et al. 2011a). However, some  
18 studies have suggested that ureilitic diamonds were  
19 formed by other processes such as chemical vapor  
20 deposition (e.g., Fukunaga et al. 1987; Nagashima et al.  
21 2012; Miyahara et al. 2013).

22 The first systematic study of carbon isotopes in  
23 ureilites was performed by Grady et al. (1985), using a  
24 stepped combustion technique on 14 of the 20 ureilites  
25 then known, including the only two polymict ureilites  
26 known at that time, Nilpena and North Haig. They  
27 showed that there is a window between 500 and 800 °C  
28 within which graphite/diamond combustion occurs, and  
29 suggested that diamond and graphite in ureilites have  
30 the same  $\delta^{13}\text{C}$  values, but showed that  $\delta^{13}\text{C}$  values  
31 clearly vary between different ureilites. Later work by  
32 Smith et al. (1999, 2001a) showed a somewhat greater  
33 range of  $\delta^{13}\text{C}$  and carbon contents for a wider variety  
34 of ureilite samples. Carbon in ureilites generally has  
35 bulk  $\delta^{13}\text{C}$  values of  $-11\%$  to  $0\%$  (Grady et al. 1985;  
36 Mittlefehldt et al. 1998; Smith et al. 2001a; Grady and  
37 Wright 2003; Hudon et al. 2004). However,  $\delta^{13}\text{C}$  values  
38 derived by bulk combustion techniques could include a  
39 contribution from material released at low  
40 temperatures. Consequently, we have applied a stepped  
41 heating technique to the Almahata Sitta samples.

42 Nitrogen is present in ureilites in much lower  
43 abundances than carbon (ca 10–150 ppm), but is mostly  
44 associated with the carbon phases. Rai et al. (2003)  
45 suggested that diamond is the major carrier of nitrogen.  
46 Nitrogen is far more complex than carbon in terms of  
47 its behavior during stepped combustion. The few  
48 nitrogen isotope analyses that have been made of  
49 ureilites (Grady et al. 1985; Grady and Pillinger 1988;  
50 Russell et al. 1993; Yamamoto et al. 1998; Rai et al.  
51 2002, 2003; Fisenko et al. 2004) have shown that there  
52 are multiple nitrogen isotopic components within these  
53 meteorites. Rai et al. (2002, 2003) showed that, in  
54 contrast to carbon isotopes,  $\delta^{15}\text{N}$  values differ between

graphite and diamond in ureilites, with diamond having  
values of  $-100\%$  and graphite having values of  $+19\%$ .  
Nitrogen in polymict ureilites differs from that in  
unbrecciated (monomict) ureilites but the paucity of  
nitrogen isotope data available for polymict ureilites  
means that only the studies of Grady and Pillinger  
(1988), Rooke et al. (1998) and Rai et al. (2003) are  
directly comparable to our investigation. In polymict  
ureilites, the main nitrogen release occurs below  
 $\sim 600$  °C and is isotopically heavy with  $\delta^{15}\text{N}$  up to  
 $+530\%$ .

The number of known ureilites has increased greatly  
in recent years because of numerous finds in both hot  
and cold deserts. In particular, the 2008 fall of the  
polymict ureilite Almahata Sitta (Jenniskens et al. 2009;  
Horstmann and Bischoff 2014) yielded around 600  
additional individual stones. These samples are very  
fresh, thereby minimizing the potential problems with  
terrestrial weathering reported by Ash and Pillinger  
(1995). The aim of the current study is to characterize  
the carbon and nitrogen isotopic values in several  
ureilitic fragments of Almahata Sitta using a stepped  
combustion technique that permits simultaneous  
analysis of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values. Stepped combustion  
mass spectrometry is a powerful tool for investigating  
the isotopic composition of carbonaceous components  
within meteorites (Swart et al. 1983; Wright et al. 1983;  
Pillinger 1984). Stepped heating allows identification of  
different carbon phases, and their isotopic  
compositions, by their characteristic combustion  
temperatures (Smith et al. 2001a; Grady et al. 2004).  
However, Smith et al. (2001b) have shown that  
combustion temperatures of graphite depend to some  
degree on grain size and crystallinity. Our new data will  
be compared with literature data for unbrecciated and  
polymict ureilites in order to assess whether the carbon  
and nitrogen components in Almahata Sitta samples are  
similar to, or different from, previously analyzed  
ureilites. We also aim to determine whether ureilitic  
carbon was derived from carbonaceous chondrites, as is  
commonly assumed (e.g., Rubín 1988), and to  
investigate the variation in  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in the early  
solar system.

## SAMPLES

The meteorites collectively known as Almahata  
Sitta fell in the Nubian desert of northern Sudan on  
October 7 2008 (Jenniskens et al. 2009; Jenniskens and  
Shaddad 2010). The rapid collection of the Almahata  
Sitta stones means that they can be considered as very  
fresh samples, having undergone minimal terrestrial  
weathering through processes such as rainfall. The  
samples studied in this paper were all recovered during

1 the first two collecting expeditions in December 2008—  
2 less than 3 months after asteroid 2008TC<sub>3</sub> collided with  
3 the Earth (Jenniskens et al. 2009; Shaddad et al. 2010).  
4 Almahata Sitta has been described as an anomalous  
5 polymict (i.e., brecciated) ureilite (Jenniskens et al.  
6 2009). Although not all Almahata Sitta samples have  
7 yet been studied, Horstmann and Bischoff (2014)  
8 collated the information available for 110 stones, of  
9 which 75 are ureilites and 35 are chondrites. This  
10 confirms the conclusions of initial mineralogical studies  
11 that Almahata Sitta is predominantly ureilitic (Bischoff  
12 et al. 2010; Zolensky et al. 2010); ureilites make up  
13 68% of the samples and 73% of the total mass of the  
14 stones studied. Of the Almahata Sitta ureilites, just over  
15 half (38 samples, ~65% of the ureilite mass (Horstmann  
16 and Bischoff 2014)) are coarse-grained akin to the main  
17 group of unbrecciated coarse-grained olivine-pyroxene  
18 ureilites that have long been dominant in ureilite  
19 collections (Goodrich 1992; Mittlefehldt et al. 1998;  
20 Goodrich et al. 2004). However, the first sample of  
21 Almahata Sitta analyzed (AS7, also studied here)  
22 represented a new fine-grained, porous lithology  
23 (Jenniskens et al. 2009). An additional 18 samples  
24 (making up 25% of ureilite samples but only 9% of the  
25 ureilite mass) are similarly anomalous ureilites (Bischoff  
26 et al. 2010; Warren and Rubin 2010; Zolensky et al.  
27 2010; Hutchins and Agee 2012; Horstmann and  
28 Bischoff 2014).

29 Four Almahata Sitta chondrites have been  
30 analyzed for cosmogenic radionuclides (Bischoff et al.  
31 2010; Meier et al. 2012), three of which are within  
32 error of values from the Almahata Sitta ureilites  
33 (Welten et al. 2010), indicating that they are all part  
34 of the same fall. The abundance of non-ureilitic  
35 material has been interpreted to indicate that  
36 Almahata Sitta was formed from an asteroid that was  
37 dominated by ureilitic material but on which a large  
38 proportion of non-indigenous debris had also collected  
39 (Horstmann and Bischoff 2014). Almahata Sitta  
40 provides important evidence that the source of ureilite  
41 meteorites is a brecciated impact-disrupted asteroid  
42 (Goodrich et al. 2004; Warren and Huber 2006;  
43 Downes et al. 2008; Borovička and Charvát 2009;  
44 Herrin et al. 2010).

45 Estimates of the mass of the 2008TC<sub>3</sub> asteroid  
46 before impact vary from 8 t (Kohout et al. 2011) to 108  
47 t (Jenniskens et al. 2009). The combined mass of all  
48 Almahata Sitta stones collected to date is approximately  
49 11 kg (Horstmann and Bischoff 2014). Therefore, no  
50 matter which calculation of pre-impact mass is used,  
51 more than 99% of the original asteroid was lost during  
52 the explosions observed as it entered the Earth's  
53 atmosphere (Borovička and Charvát 2009; Jenniskens  
54 et al. 2009; Jenniskens and Shaddad 2010; Jenniskens

et al. 2010b; Kwok 2009; Jenniskens et al. 2010a;  
Shaddad et al. 2010). Thus, it is not known to what  
4 extent the collected material is representative of the  
original asteroid.

Five Almahata Sitta stones were selected for analysis  
(AS7, AS22, AS27, AS36, and AS44). Samples were  
primarily chosen based on availability of material for  
destructive analysis, but also cover the range of  
heterogeneities found within Almahata Sitta. Apart  
from AS22, the stones studied here have already  
been investigated by various authors using different  
techniques. AS7 represents the unusual fine-grained  
porous lithology and is the same sample initially  
described from the Almahata Sitta fall and identified as  
an anomalous ureilite (Jenniskens et al. 2009). The rest  
are all coarse-grained compact main group ureilites.  
AS22 and AS427 were large stones (115.32 g and  
283.84 g, respectively), whereas AS7 (1.52 g) and AS44  
(2.21 g) were much smaller (Shaddad et al. 2010). Three  
of the samples (AS7, AS36, and AS44) have been  
described by Zolensky et al. (2010), while samples AS7,  
AS27, AS36, and AS44 were analyzed by Sandford et al.  
(2010). AS27 has also been investigated by Warren and  
Rubin (2010) and Burton et al. (2011) and AS44 was  
described by Goodrich et al. (2010a). Samples AS36 and  
AS44 were analyzed by Murty et al. (2010) for nitrogen  
and noble gases. Among our samples, the presence of  
diamond has been confirmed by Raman spectroscopy in  
AS7 (Ross et al. 2011a) and has also been identified in  
AS22, AS27, and AS44.

The forsterite (Fo) content of olivine cores in  
previously studied ureilites ranges from ~75 to 96;  
however, they are not evenly distributed, showing a  
significant peak at Fo~78–79 with a secondary lesser  
peak at Fo~90–92 (Goodrich et al. 2004; Downes et al.  
2008). Reported Fo values for Almahata Sitta ureilites  
cover this entire range from Fo~76 to Fo~95 (Bischoff  
et al. 2010; Warren and Rubin 2010; Kita et al. 2011;  
Horstmann and Bischoff 2014). Even within this small  
sample set, there is a major peak at ~Fo79. Oxygen  
isotope analyses of Almahata Sitta ureilites (Bischoff  
et al. 2010; Rumble et al. 2010; Kita et al. 2011;  
Horstmann et al. 2012) also cover the range of values  
previously found in ureilites (Clayton and Mayeda 1988,  
1996; Kita et al. 2004). These data are indicative of the  
heterogeneity within the original 2008TC<sub>3</sub> asteroid. Our  
studied samples range in Fo values from 79 (AS44) to  
90 (AS36) and their  $\Delta^{17}\text{O}$  values range from -0.4 to  
-1.5‰ (Rumble et al. 2010).

Polished blocks were prepared for petrographic  
study. AS22 and AS27 were split into petrographic and  
isotopic sub-samples, with AS36 being already  
completely disaggregated and requiring no further  
subdivision. AS7 was provided as two separate pieces,

Table 1. Summary data of C and N in Almahata Sitta samples (see Appendix for full data set). For AS22 N% yield in brackets for main release is value without the release at 1000 °C.

	AS7	AS22	AS27	AS36	AS44
Sample mass (mg)	4.736	5.129	5.466	4.41	4.948
Fo content in olivine	86	80	85	90	79
px*: px/(ol+px) %	0.31	0.28	0.22	0.35	0.15–0.23
Total C (wt%)	1.57	1.18	1.20	2.30	1.88
Carbon					
Average total $\delta^{13}\text{C}$ (‰)	0.0	-7.6	-2.8	-5.4	0.1
Average error (‰)	0.3	0.3	0.3	0.3	0.3
Main release range (° C)	500–700	500–700	500–700	500–700	500–700
C in main release (ng)	70895	58566	58409	91630	84041
C in main release (wt%)	1.49	1.14	1.07	2.06	1.70
% of total C in main release	94.7	96.6	89.3	89.6	90.2
Weighted av $\delta^{13}\text{C}$ in main release ‰	0.4	-7.3	-2.3	-5.2	0.4
Error (‰)	0.3	0.3	0.3	0.3	0.3
Nitrogen					
Total N (ppm)	18.11	27.11	21.19	9.35	11.78
Average total $\delta^{15}\text{N}$ (‰)	-48.2	-29.4	-7.5	-24.1	-52.1
Average error (‰)	0.9	4.5	4.3	2.2	2.4
Main release range (° C)	600–700	600–700	650–700	650–700	600–700
N in main release (ng)	54.63	36.69	34.11	12.41	37.19
N in main release (ppm)	11.47	7.15	6.24	2.79	7.52
% of N in main release	63.3	26.4 (60.5)	29.5	29.9	63.8
Weighted av $\delta^{15}\text{N}$ in main release ‰	-91.0	-69.0	-61.3	-95.6	-94.7
Error (‰)	0.3	4.8	5.3	1.0	1.2

one with fusion crust and one without; the former was made into the polished block and the latter used for isotope analyses. AS44 was provided as two distinct samples, but petrographic and mineralogical analysis of the polished blocks showed them to have identical features and so only one was sub-sampled for isotopic analysis.

The polished blocks of samples studied here are all olivine-dominated (Table 1). When compared with the results from infrared spectroscopy by Sandford et al. (2010), the px\* values (where px\* = modal px/(modal px + modal ol)) for AS27 are almost identical and our results for AS36 are very close to one of three analyses splits of AS36. However, our sample of AS44 is considerably more olivine-rich and our sample of AS7 is marginally more olivine-rich. All samples contain carbon, with one of the AS44 blocks appearing to contain a vein-like carbon-rich area. The samples also contain metal and sulfide; grain-boundary Fe-metal is the most common form, which can contain minor amounts of Ni and Si and trace amounts of Co and P. AS27 and AS44 both contain carbon-bearing metal phases. In AS27 these are included within silicate grains as “spherules”. In AS44, the C-bearing metal is present in the grain boundaries alongside Fe-metal, schreibersite, and sulfide (Goodrich et al. 2010a; Mikouchi et al. 2011; Agoyagi et al. 2013).

AS7 shows a distinctive porous texture. Olivine core compositions are Fo<sub>86</sub>. This sample was analyzed by micro Raman techniques (Ross et al. 2011a), which revealed the presence of diamond as well as graphite and amorphous carbon. AS22 has not previously been described. It shows a typical coarse-grained (up to 2 mm) olivine-dominated ureilitic texture with cores of Fo<sub>80</sub> (Ross et al. 2011b). Sample AS27 consists of olivine (Fo<sub>85</sub>) and low-Ca pyroxene. Warren and Rubin (2010) suggested that AS27 may have experienced some degree of shock, resulting in undulose extinction in olivine, and that its original texture may have been poikilitic. Our sample of AS36 was completely disaggregated into individual crystal grains with a small proportion of mixed grains, and therefore it tends to lack interstitial material. It is composed largely of coarse-grained (up to 3 mm) olivine Fo<sub>90</sub> crystals (Ross et al. 2011c). AS44 is a coarse-grained (>2 mm) olivine-dominated ureilite (Fo<sub>79</sub>) with some pigeonite. Its texture resembles those of impact-smelted ureilites (Goodrich et al. 2010a; Ross et al. 2011b).

## METHODOLOGY

Almahata Sitta samples were analyzed for C and N abundance and isotopic compositions using a

1 stepped combustion technique on the FINESSE static  
2 vacuum mass spectrometer at the Open University  
3 (UK). For detailed information regarding the  
4 components and processes within the FINESSE  
5 instrument, see Verchovsky et al. (1998, 2002). Care  
6 was taken to ensure that no sub-samples used for  
7 isotopic analyses had fusion crust attached. The  
8 meteorite chips were hand crushed to a powder in a  
9 class 10000 clean room, using an agate pestle and  
10 mortar. This was transferred to a class 100 clean  
11 room where approximately 5 mg of the powdered  
12 ureilites were weighed into 0.25  $\mu\text{m}$  thick Pt buckets  
13 that had been pre-cleaned by heating to 900  $^{\circ}\text{C}$  in the  
14 presence of CuO in a sealed quartz tube pumped to  
15  $10^{-7}$  mbar.

16 After being loaded in the mass spectrometer  
17 extraction system, the samples were heated  
18 incrementally from 200 to 1400  $^{\circ}\text{C}$  in the presence of  
19 oxygen derived from thermal decomposition, at 930  $^{\circ}\text{C}$ ,  
20 of CuO present in a separately heated unit with an inlet  
21 into the furnace, resulting in the liberation of individual  
22 components. Carbon (in the form of  $\text{CO}_2$ ) and  
23 molecular nitrogen were cryogenically separated from  
24 each other before analysis. Simultaneous analysis was  
25 made possible by using multiple mass spectrometers  
26 connected to a common extraction line: two magnetic  
27 sector mass spectrometers for determination of carbon  
28 isotopes and nitrogen abundance, and a quadrupole  
29 mass spectrometer for nitrogen isotopes. Abundance  
30 yields are recorded in nanograms with errors of  $<1\%$  in  
31 the carbon measurement, made on a capacitance  
32 manometer, and  $<5\%$  in the nitrogen measurements,  
33 which were determined from the previously calibrated  
34 relationship between  $m/z = 28$  intensity and the  
35 nitrogen volume. The system blank levels of the  
36 instrument are  $<10$  ng and  $<1$  ng for carbon and  
37 nitrogen, respectively. Isotopic compositions are  
38 reported in the delta ( $\delta$ ) notation as a deviation in  
39 parts per thousand (‰) from the Peedee Belemnite  
40 (vPDB) standard for carbon and from terrestrial air for  
41 nitrogen. Isotopic reproducibility is typically  $<1\%$ . As a  
42 result of the high yields normally found in ureilites  
43 (Grady et al. 1985; Smith 2002), blank correction was  
44 based on the average yield and isotopic composition of  
45 a system blank run prior to the experiment. Total  
46 isotopic error was calculated through an error  
47 propagation calculation, based on the contribution of  
48 the blank to the total yield. Summary results for  
49 carbon and nitrogen concentrations and isotopic  
50 compositions in the Almahata Sitta samples are given  
51 in Table 1, and full results for each temperature step  
52 can be found in the Appendix. No **N** analyses are  
53 available for the highest temperature step for AS7 and  
54 AS36 because of system failure.

## RESULTS

### Carbon

In Table 1 we report the average  $\delta^{13}\text{C}$  values of the carbon across all temperature ranges (equivalent to the bulk carbon composition), and the weighted average  $\delta^{13}\text{C}$  of the carbon released during the temperature steps where  $>\sim 90\%$  of carbon was liberated (referred to as the “main release carbon”). The main carbon release temperature range is the same in all the samples, occurring between 500 and 700  $^{\circ}\text{C}$ . The total amount of carbon released in all of the samples was 1.18 to 2.3 wt %. The  $\delta^{13}\text{C}$  value of the main carbon release is always heavier than that of the total C (Table 1), because of the contribution from an isotopically light, low-temperature component. Differences in  $\delta^{13}\text{C}$  between the main release component and the average  $\delta^{13}\text{C}$  across all temperatures (i.e., including the low-temperature component) range from 0.2 to 0.5‰.

The five Almahata Sitta ureilite samples have very similar carbon release patterns (Fig. 1). During the steps at  $<500$   $^{\circ}\text{C}$ , less than 2% of the total carbon was released and the  $\delta^{13}\text{C}$  values were typically around  $-25\%$ . As combustion reached about 600  $^{\circ}\text{C}$ ,  $\delta^{13}\text{C}$  showed a rapid increase to “peak” (main release) values ranging from  $-7.3\%$  (AS22) to  $+0.4\%$  (AS44). Following the main release of carbon, the samples behaved in slightly different ways. Samples AS7 and AS22 released less than  $\sim 4\%$  of their total carbon above 750  $^{\circ}\text{C}$ . The carbon stepped combustion plots (Fig. 1) show that these samples have a very subtle second minor release, at  $\sim 900$   $^{\circ}\text{C}$  (AS7) and  $\sim 1000$   $^{\circ}\text{C}$  (AS27). In AS7, the second release is almost identical in isotopic composition to the major release at 650  $^{\circ}\text{C}$ . Thus, the second release probably represents combustion either of higher crystallinity graphite or of genetically related diamond. The latter is more likely as this sample definitely contains diamond (Zolensky et al. 2010; Ross et al. 2011a). Samples AS27, AS36, and AS44 released a slightly larger amount of carbon ( $\sim 10\%$ ) at temperatures  $>750$   $^{\circ}\text{C}$ ; the isotopic composition of these releases ranges from  $+1.5\%$  (AS44) to  $-5.7\%$  (AS36). Two of these samples (AS27 and AS44) contain C-rich metal which may account for these high-temperature releases. The extreme change in isotopic composition of the highest temperature release of carbon from AS44 is almost certainly an artifact of the method (possibly furnace residue).

### Nitrogen

Table 1 shows the  $\delta^{15}\text{N}$  results for the five Almahata Sitta samples; step combustion profiles are

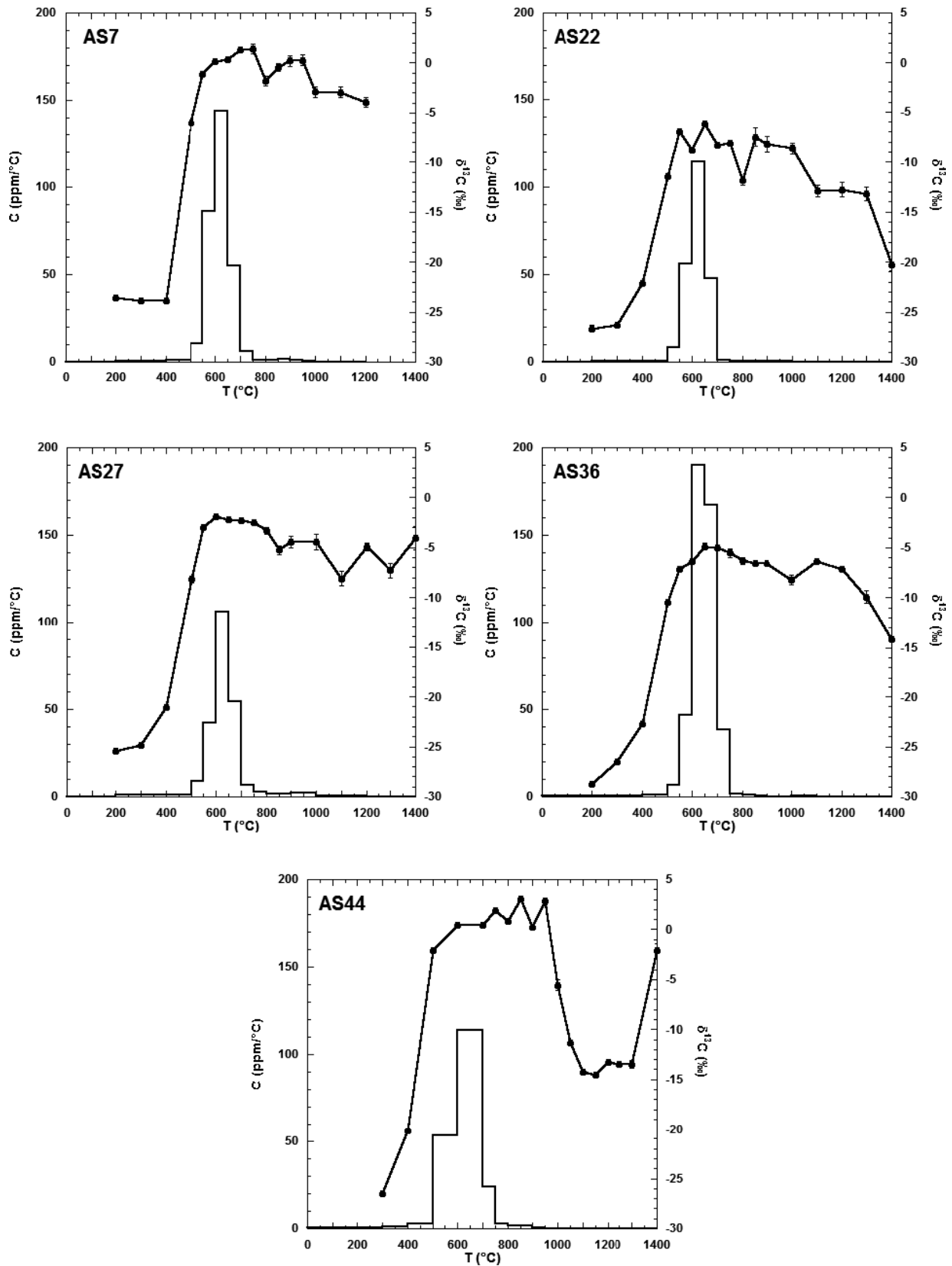


Fig. 1. Carbon release patterns (solid line) and  $\delta^{13}C$  compositions (points with error bars) of five samples of Almahata Sitta ureilites during stepped heating combustion. **8**

1 shown in Fig. 2. Both the nitrogen abundance  
2 combustion profiles and corresponding nitrogen isotopic  
3 values show more complexity than for carbon.  
4 Nevertheless, all of the samples show a clear main  
5 release of nitrogen at around 600–700 °C, over which  
6 the  $\delta^{15}\text{N}$  values decrease significantly. It is difficult to  
7 define the main release peaks because of the presence of  
8 lower temperature releases, but in general these main  
9 releases yield average  $\delta^{15}\text{N}$  values from  $-61\text{‰}$  (AS27)  
10 to  $-95.6\text{‰}$  (AS36). In sample AS22, a second large  
11 release peak is seen at a much higher temperature  
12 (1000 °C) which lacks these low  $\delta^{15}\text{N}$  values.

13 In contrast to the carbon release profiles, at  
14 temperatures below 400°C, up to ~20% of nitrogen was  
15 released from all of our samples with a weighted  
16 average  $\delta^{15}\text{N}$  of +8 to +19‰. Between 400 °C and  
17 600 °C, samples AS7, AS27 and AS36 released 20.8 to  
18 34.5% of total N, with a weighted average  $\delta^{15}\text{N}$  of +29  
19 to +42‰. In contrast, samples AS22 and AS44 released  
20 less nitrogen (8.7% and 14.0%, respectively) over this  
21 temperature range, and this nitrogen was significantly  
22 heavier than that seen in the other three samples (+64‰  
23 and +82‰). Sample AS27 was investigated by Burton  
24 et al. (2011) who identified the presence of  
25 extraterrestrial amino acids. This component may be  
26 responsible for the low temperature, heavy nitrogen  
27 release.

28 At temperatures between 600 °C and 900 °C,  
29 samples AS7 and AS44 released most (~70%) of their  
30 nitrogen. The isotopic composition of this release was  
31 the lightest of the analyzed samples in this study, with  
32 weighted average  $\delta^{15}\text{N}$  values of  $-84\text{‰}$  (AS7) and  
33  $-91\text{‰}$  (AS44). Samples AS27 and AS36 also released  
34 most of their nitrogen over this temperature range,  
35 however, it was a much smaller release in terms of total  
36 nitrogen (~43%). Its isotopic composition was slightly  
37 heavier than that seen in AS7 and AS44, with weighted  
38 average  $\delta^{15}\text{N}$  values of  $-40$  and  $-79\text{‰}$ .

39 AS22 is unique among the samples in that the  
40 600 °C to 900 °C release only made up 27.8% of total  
41 nitrogen, with a  $\delta^{15}\text{N}$  of  $-63\text{‰}$ . Its largest release  
42 (56.4% of total N) occurred at 1000 °C with a  
43 corresponding  $\delta^{15}\text{N}$  of  $-29\text{‰}$ . An extremely similar  
44 major release of nitrogen at 1000 °C was reported from  
45 Goalpara by Grady et al. (1985). This may suggest an  
46 additional, as yet unidentified, component of nitrogen in  
47 some ureilites, such as a nitride phase similar to those  
48 found in E chondrites. At temperatures >900 °C the  
49 other Almahata Sitta ureilite samples released <12% of  
50 their total nitrogen with weighted average  $\delta^{15}\text{N}$  values  
51 in the range  $-32$  to  $-3\text{‰}$ . In AS27 and AS36, the  
52 nitrogen isotopic composition changed from heavier  
53 values to lighter values with increasing temperature. In  
54 sample AS44, the opposite pattern was observed,

whereas for AS7, the high-temperature component had  
a much less variable isotopic composition.

The release patterns of carbon and nitrogen suggest  
that the two elements are clearly connected, as they are  
released at similar temperatures (Figs. 1 and 2),  
although the main release of nitrogen starts at a slightly  
higher temperature than that of carbon. This may be  
because the major carrier of nitrogen is diamond which  
combusts at a slightly higher temperature than graphite.  
There is a negative correlation between the total  
amount of carbon released and the total amount of  
nitrogen released during combustion of the Almahata  
Sitta samples (Fig. 3a). Sample AS22 represents the  
sample with the highest nitrogen and lowest carbon  
abundances (although this value includes the anomalous  
release of nitrogen at high temperature), whereas sample  
AS36 represents a low nitrogen, high carbon component  
(this sample was disaggregated and contains very little  
interstitial material). A stronger correlation is seen  
between the total amount of N and the ratio of N/C  
released from each sample (Fig. 3b). The correlation  
supports the contention that two separate components  
dominate the nitrogen isotopes. The nature of these  
components is discussed below.

## DISCUSSION

### Comparison of Carbon and Nitrogen With Ureilite Literature Data

The Almahata Sitta ureilite samples show a  
ubiquitous carbon component at <500 °C which is a  
very small (<2%) proportion of the total carbon yield  
and has a  $\delta^{13}\text{C}$  value of around  $-25$  to  $-30\text{‰}$  (Fig. 1).  
The low-temperature combustion behavior and isotopic  
composition of this carbon is suggestive either of  
terrestrial organic carbon (Hoefs 1997) or of extra-  
terrestrial, but non-ureilitic, organic components as  
described by Glavin et al. (2010) and Burton et al.  
(2011). While these are very minor components in  
Almahata Sitta samples, these low-temperature  
components could bias the carbon isotopic value  
determined from a bulk (i.e., non-stepped combustion)  
analysis, particularly if the component is terrestrial in  
origin and the sample has been in the terrestrial  
environment for a long time.

Data from three older ureilite falls (Dyalpur, Novo-  
Urei, and Haverö) show a range between 3.6 and 4.7%  
of total carbon released below 500 °C (Grady et al.  
1985), so the Almahata Sitta samples have almost half  
the abundance of contaminating carbon compared with  
other falls. For ureilite finds, however, the low-  
temperature component ranges from 1.7% (Y790981) to  
23.7% (Goalpara) of total carbon (Grady et al. 1985).



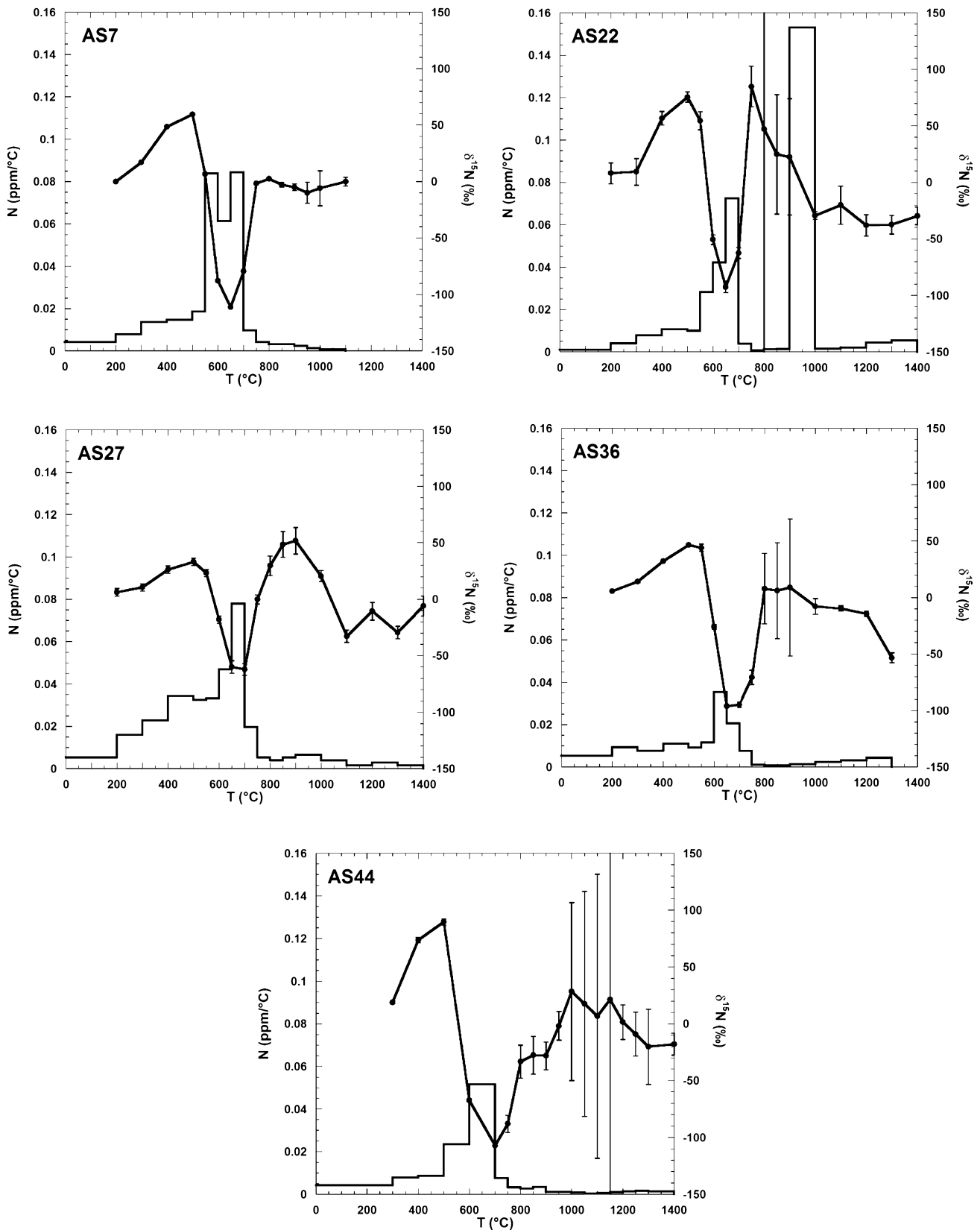
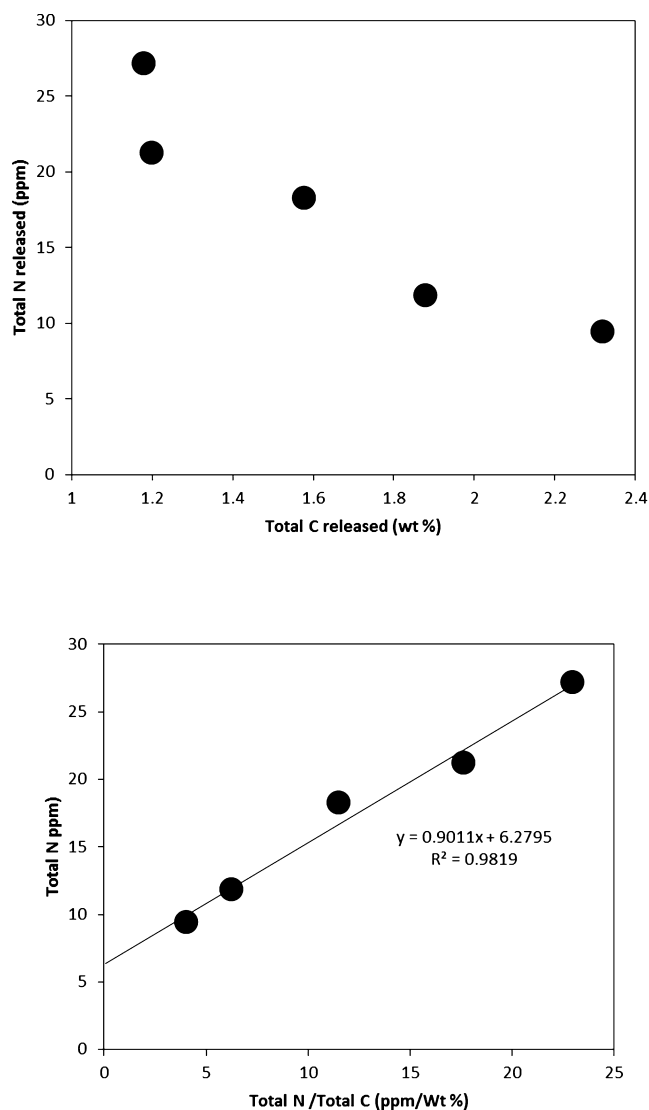


Fig. 2. Nitrogen release patterns (solid line) and nitrogen isotope compositions (points with error bars) of five samples of Almahata Sitta ureilites.



9 Fig. 3. a) Total amount of C versus total amount of N released during stepped combustion of five Almahata Sitta ureilites. b) Total amount of N released versus total N/total C released during stepped combustion of five Almahata Sitta ureilites. N.B. From a mathematical point of view, one of these two correlations cannot be a straight line relationship.

The low-temperature component is also significant in two polymict ureilite samples, North Haig (37.1%) and Nilpena (8.2%). Excluding the anomalously high Goalpara value, and the polymict samples, the low-temperature carbon released by unbrecciated ureilite finds ranges from 1.7% to 6.5%. Thus, in terms of carbon, the Almahata Sitta samples are among the least contaminated ureilites yet studied.

The main carbon components released at higher temperatures are considered to be indigenous ureilitic carbon. Previously analyzed unbrecciated ureilites all have similar ranges in carbon abundance and isotopic

composition (Grady et al. 1985; Smith et al. 2001a; Hudon et al. 2004) and main release  $\delta^{13}\text{C}$  values of the Almahata Sitta samples (+0.4 to  $-7.3\%$ ) fall well within these ranges. Grady et al. (1985) suggested that ureilites show a bimodality of peak  $\delta^{13}\text{C}$  values, with clusters around  $-2\%$  and  $-10\%$ . It appeared that the isotopically lighter values were associated with ureilites that had more Mg-rich olivine core compositions ( $>F_{0.83}$ ), whereas the isotopically heavier values belonged to samples with more Fe-rich ( $<F_{0.80}$ ) olivine cores (Fig. 4). However, the ureilites studied by Grady et al. (1985) cluster around only two different Fo compositions, although this bias results from the bimodal distribution of core Fo values in all ureilites (Goodrich et al. 2004; Downes et al. 2008). In a later study, Hudon et al. (2004) showed a more extensive data set for both Fo (74–95) and bulk  $\delta^{13}\text{C}$  ( $-11$  to  $+1\%$ ) values that also shows a negative correlation (Fig. 4). The reason for this correlation is not known, although it might relate to mixing of different nebula components in the source of the original UPB. Unlike the data of Grady et al. (1985) and Hudon et al. (2004), our Almahata Sitta ureilite data show no correlation between olivine core Fo composition and  $\delta^{13}\text{C}$  values. However, the small number of data points in our study precludes making any conclusion from this observation and, in any case, our data plot in or very near to the field defined by the data of Hudon et al. (2004).

Nitrogen is a trace element in unbrecciated ureilites (3–55 ppm according to Grady and Wright (2003) and Rai et al. (2003)). Our Almahata Sitta ureilite samples yield total nitrogen abundances in the range 9.4–27 ppm, well within this reported range. There are fewer measurements of nitrogen within polymict ureilites but nitrogen yields are generally higher, up to 152 ppm (Grady and Pillinger 1988; Rooke et al. 1998; Rai et al. 2003). Previous stepped combustion studies have suggested multiple components with distinct  $\delta^{15}\text{N}$  values with some differences in nitrogen composition and combustion behavior between unbrecciated and polymict ureilites (Grady et al. 1985; Grady and Pillinger 1988; Rooke et al. 1998; Yamamoto et al. 1998; Rai et al. 2003). Grady et al. (1985) measured the abundance and isotopic composition of nitrogen in five ureilite specimens using stepped combustion. Their results showed multiple nitrogen components with different isotopic compositions, released at different temperatures. However, the lightest nitrogen they measured had a  $\delta^{15}\text{N}$  value of only  $-62.4\%$  (the 800 °C step in Kenna) and bulk isotopic compositions  $\delta^{15}\text{N}$  values ranged from  $-24.8\%$  (Kenna) to  $+26.2\%$  in polymict sample North Haig. For unbrecciated (“monomict”) ureilites, Yamamoto et al. (1998) and Rai et al. (2003) described the main nitrogen release as

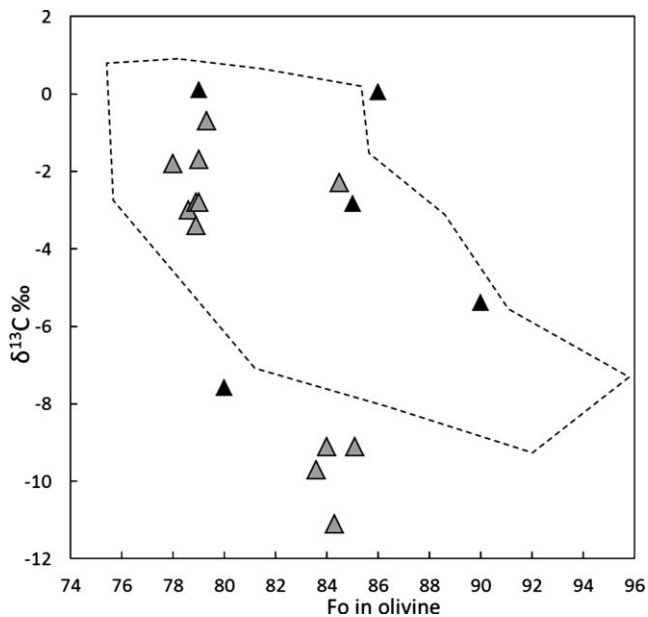


Fig. 4.  $\delta^{13}\text{C}$  versus core olivine mg# in analyzed Almahata Sitta ureilites (black triangles) and data for a variety of monomict (unbrecciated) ureilites (gray triangles) analyzed by Grady et al. (1985). Field outlined is for a larger number of unbrecciated ureilites (Hudon et al. 2004).

coinciding with the main carbon release at temperatures between 600 °C and 900 °C, and this nitrogen was isotopically light. The weighted average  $\delta^{15}\text{N}$  of the main nitrogen releases have been calculated as  $-53.6$  to  $-112.4\text{‰}$  (Yamamoto et al. 1998) and  $-26.6$  to  $-105\text{‰}$  (Rai et al. 2003). Unbrecciated ureilites can also contain a significant low temperature, isotopically heavy, component with  $\delta^{15}\text{N}$  compositions in the range 0 to  $+95\text{‰}$  (Yamamoto et al. 1998; Rai et al. 2003).

Only four polymict ureilites (North Haig, Nilpena, EET 83309, and EET 87720) have been previously analyzed for nitrogen isotopes. Thus, it is difficult to define what the combustion profile of a typical polymict ureilite looks like in comparison with a typical unbrecciated ureilite. However, Grady and Pillinger (1988) identified two components, one isotopically light with  $\delta^{15}\text{N}$  values that reached  $-94\text{‰}$  and one which is isotopically heavy nitrogen with  $\delta^{15}\text{N}$  up to  $+600\text{‰}$ . All polymict ureilites have a high-yield, low-temperature release ( $<500$  °C) that is isotopically heavy ( $\delta^{15}\text{N} \geq +50\text{‰}$  or even  $\geq +150\text{‰}$ ). On the other hand, two combustion profiles of Nilpena show clear evidence for a large release of light nitrogen over temperature intervals of 700–800 °C (weighted average  $\delta^{15}\text{N}$  over this temperature range =  $-66.6\text{‰}$ ; Grady and Pillinger 1988), and from 700 to 1050 °C ( $\delta^{15}\text{N} = -89.5\text{‰}$ ; Rai et al. 2003).

Our Almahata Sitta results also show  $^{15}\text{N}$  enriched values at low temperatures with weighted average

compositions of  $+24$  to  $+55\text{‰}$  at temperatures between 200 °C and 550 °C, although these values are somewhat less  $^{15}\text{N}$  enriched than have been measured previously in polymict ureilites. Their major nitrogen release was over a narrow temperature range within the main carbon release temperature range. Their main release isotopic compositions (from  $-61\text{‰}$  to  $-96\text{‰}$ ; Table 1) tend to be heavier than the values reported for unbrecciated ureilites by Yamamoto et al. (1998) and Rai et al. (2003). Among our samples, only two (AS7 and AS44) released any nitrogen with a  $\delta^{15}\text{N}$  value of  $<-100\text{‰}$  and this was only seen in one temperature step in each sample (see Appendix).

Murty et al. (2010) also carried out step-wise combustion analyses of nitrogen isotopes in AS36. Their bulk sample of AS36 contained almost twice the amount of nitrogen (21.1 ppm compared with our value of 9.35 ppm). The main release (63% N) occurred between 800 and 1200 °C, which is much higher than the main release temperature we obtained. However, a very different experimental protocol was used whereby the samples were combusted at 200 °C steps between 300 °C and 1000 °C and then pyrolyzed the sample at 1200 °C, 1400 °C, and 1800 °C, compared with our protocol where the sample was combusted at 50 °C or 100 °C degree steps (combustion involves heating the sample in the presence of oxygen whereas pyrolysis does not). The bulk  $\delta^{15}\text{N}$  isotopic composition obtained by Murty et al. (2010) of  $-36.8\text{‰}$  is lighter than our bulk value of  $-24\text{‰}$  but their peak release, i.e., the single temperature step where the largest amount of N was released, occurred at 1000 °C with a  $\delta^{15}\text{N}$  of  $-94.3\text{‰}$ , which is within error of the composition ( $-95.6\text{‰}$ ) seen in the 650 °C main release in our analysis of AS36.

In a stepped combustion study of carbon-bearing acid residues and diamond concentrates from ureilites, Russell et al. (1993) showed that  $\delta^{15}\text{N}$  values reach  $-100$  to  $-120\text{‰}$  at temperatures  $>600$  °C, whereas at low temperatures the isotopic composition was  $\sim 0\text{‰}$ . Fisenko et al. (2004) obtained  $\delta^{15}\text{N}$  values for acid residue samples of Novo-Urei (the oldest ureilite fall and the meteorite that gives the ureilites their name) of  $-108\text{‰}$  and  $-114\text{‰}$ . Murty et al. (2010) also analyzed an acid residue sample from AS44 using stepped combustion, in which the major release occurred at temperatures  $\geq 600$  °C with corresponding  $\delta^{15}\text{N}$  values ranging from  $-96.0\text{‰}$  to  $-115.3\text{‰}$ , very similar to our main release value of  $-94.7\text{‰}$  for this sample.

For nitrogen, any terrestrial atmospheric contamination would be expected to be most apparent in the steps below 500 °C, although Hashizume and Sugiura (1995) describe terrestrial nitrogen as being detectable at temperatures up to 700 °C in ordinary chondrites. Nitrogen stepped combustion data are

available for three ureilite falls: Dyalpur (Grady et al. 1985), Lahrauli and Haverö (Rai et al. 2003). The low-temperature nitrogen abundance ranges from 19.5% (Dyalpur; Grady et al. 1985) to 38.5% (Haverö; Rai et al. 2003). For unbrecciated ureilite finds, the nitrogen abundance at <500 °C is highly variable and ranges from 11.2% to 65.9% of total nitrogen (Grady et al. 1985; Yamamoto et al. 1998; Rai et al. 2003). The Almahata Sitta samples yield 5–30% of total nitrogen at <500 °C and so, on the basis of abundance alone, it is not obvious that they show less terrestrial nitrogen contamination than other ureilites. Atmospheric nitrogen adsorbed by interaction with the terrestrial environment would push the  $\delta^{15}\text{N}$  values toward 0‰, whereas terrestrial biological nitrogen has  $\delta^{15}\text{N}$  values from +5.1‰ to +10.2‰ (Boyd et al. 1993), which would also cause a similar trend to higher values if the samples had been contaminated by biological material. However, it is difficult to ascertain the contribution of contaminating terrestrial nitrogen by looking at the isotopic composition at these temperatures as at least some of this low-temperature component is probably indigenous nitrogen. This contribution may be from extraterrestrial but non-ureilitic organic material (Glavin et al. 2010; Burton et al. 2011) that has been found in Almahata Sitta ureilites and which would combust in this temperature range. Acapulcoites and lodranites have recently been shown to contain a similar extraterrestrial organic component (Charon et al. 2014).

### C & N in the Solar System

Figure 5 compares our carbon and nitrogen isotope results with literature values for different groups of carbonaceous chondrites and enstatite chondrites. Two sets of results are shown for the Almahata Sitta ureilitic fragments: one set (open symbols) shows the bulk release of carbon and nitrogen whereas the second set (closed symbols) shows the isotopic composition for the main release (Table 1). Two of our samples (AS7 and AS44) are almost indistinguishable on this plot, despite having different olivine core compositions and different textural characteristics. All of our data fall outside the fields of known carbonaceous chondrites (Fig. 5). We therefore suggest that, despite the well-known similarity of oxygen isotopes between ureilites and carbonaceous chondrites (Clayton and Mayeda 1988), on the basis of carbon and nitrogen isotopes, we agree with Grady et al. (1985) and Smith (2002) that it is unlikely that ureilites were formed from a carbonaceous chondritic precursor. This conclusion is also in agreement with that of Warren (2011) based on non-traditional stable isotope ratios. Data for acid residues and diamond concentrates from ureilites (Russell et al. 1993; Fisenko

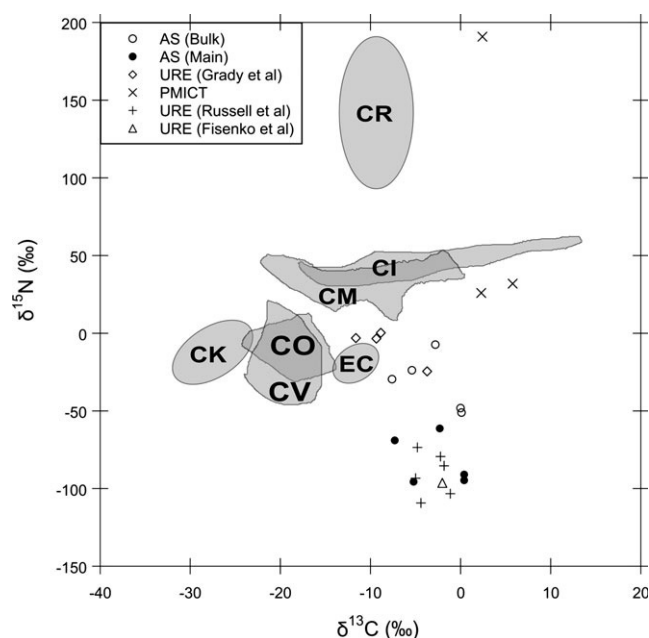


Fig. 5. Comparison of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in the Almahata Sitta ureilites with literature data from other ureilites and carbonaceous and enstatite chondrites (data from Kerridge 1985; Grady et al. 1986; Sephton et al. 2003; Pearson et al. 2006). Almahata Sitta data are shown as “Bulk” (average isotopic composition of total carbon and nitrogen, Table 1) and “Main” (weighted average isotopic composition taken from the main release temperature range, Table 1). “Ure (Russell et al.)” are acid-treated ureilites (Russell et al. 1993), “Ure (Fisenko et al.)” are ureilite diamond separates (Fisenko et al. 2004), averaged across the whole combustion range.

et al. 2004), in which the low-temperature N-isotope component has been removed, overlap the main release data for AS7, AS44, and AS36 (Fig. 5) and point toward a primordial component.

Objects within the solar system vary in their carbon isotope ratios because of three processes: (1) fractionation of carbon isotopes by nebula condensation; (2) the presence of unequilibrated carbon-rich presolar grains (e.g., SiC, diamond); and (3) fractionation effects related to secondary processes on the parent body, e.g., aqueous alteration, thermal metamorphism and shock. There is very little possibility of the presence of presolar grains in the Almahata Sitta samples, as a result of the extensive processing ureilites have experienced (although any presolar grains that may have been present might have been reworked and their carbon liberated during parent body processing). Furthermore, although ureilites display ample evidence for shock, there is no obvious difference between highly shocked and low shock ureilites in terms of carbon isotopes. Therefore, the range in  $\delta^{13}\text{C}$  observed in ureilites is most probably a result of variations within the primordial solar nebula. An alternative process,

isotopic fractionation during redox reactions on the original ureilite parent asteroid, is considered to be unlikely according to the results of a study of Smith et al. (2000a).

The origin and extent of variation in N-isotopes in the solar system are not well understood (Aleon 2010; Marty et al. 2011). Nitrogen isotopic variation is generally considered to have been produced by mixing between isotopically light primordial solar nitrogen ( $\delta^{15}\text{N} = -380\text{‰}$ ) and an isotopically heavy (up to  $+4000\text{‰}$ ) component. The latter seems to be formed by isotope fractionation processes such as a self-shielding effect (Clayton 2002) or ion-molecular reactions from solar nitrogen (Franchi et al. 1986). Some solar system objects contain a high fraction of solar nitrogen, e.g., osbornite in CAIs have  $\delta^{15}\text{N} = -360\text{‰}$  (Meibom et al. 2007) whereas iron meteorites have  $\delta^{15}\text{N} = -96\text{‰}$  (Franchi et al. 1993). A diamond-bearing extraterrestrial pebble found in the Libyan desert also yields  $\delta^{15}\text{N}$  values of  $-100\text{‰}$  (Marty et al. 2014). Ureilites formed early in solar system history (no later than 4564 Ma, according to Goodrich et al. (2010b) and Qin et al. (2010)) and contain a significant fraction of light nitrogen. Acapulcoites also show a similar component with even lighter nitrogen of  $\delta^{15}\text{N} = -150\text{‰}$  (El Goresy et al. 2005). This suggests that nitrogen-bearing carbonaceous material was formed very early in the solar system and escaped significant processing in the ureilite parent body. Terrestrial diamonds generally have  $\delta^{15}\text{N}$  values of  $-5\text{‰}$  and the lightest values found for Earth ( $\delta^{15}\text{N} = -39\text{‰}$ ) occur very rarely (Palot et al. 2012). Thus, Earth may also retain a very small remnant of primordial nitrogen.

### Nature of Almahata Sitta Parent Asteroid

One of the puzzling issues with regards to the origin and history of the asteroid from which the Almahata Sitta samples were derived is the similarities and differences with samples traditionally described and classified as polymict ureilites (i.e., brecciated and with inhomogeneous olivine core compositions) and those described as monomict ureilites (i.e., those that are unbrecciated and have homogeneous olivine core compositions). Previous investigations of polymict ureilites suggested that, because of the presence of a wide-variety of exotic components such as carbonaceous chondrite-like dark clasts (Brearley and Prinz 1992) and other chondritic (OC, EC, RC) and angritic clasts (Ikeda et al. 2003; Goodrich et al. 2004; Kita et al. 2004; Downes et al. 2008), these meteorites originated as regolith materials on the ureilite daughter asteroidal body. Such regolith breccias also show evidence for solar wind implantation (Ott et al. 1990).

In terms of the mineralogy and petrography of the Almahata Sitta samples studied here, the individual fragments are unbrecciated lithologies, displaying dominant olivine and pyroxene with abundances and compositions similar to those found within the range of unbrecciated ureilites. However, the abundance of non-ureilitic stones found within the strewn field clearly indicates that the Almahata Sitta parent body was extremely heterogeneous in its composition and was undoubtedly a brecciated asteroid (Herrin et al. 2010; Bischoff et al. 2010). However, despite knowing that Almahata Sitta was a polymict breccia, the carbon and nitrogen results from the ureilitic fragments do not suggest the presence of any non-ureilitic material, other than perhaps the high- $T$  ( $\sim 1000$  °C) nitrogen component in AS22. It is therefore possible that other areas where ureilite finds are common (e.g., Dar al Gani; Frontier Mountains) may be relics of unrecognized previous falls of polymict ureilites (Smith et al. 2001b; Downes et al. 2010).

### CONCLUSIONS

1. Ureilite fragments from the Almahata Sitta fall are not unusual in their C isotopes; the  $\delta^{13}\text{C}$  results obtained for their main carbon release ( $+0.4$  to  $-7.3\text{‰}$ ) fall within the same range as those previously analyzed in unbrecciated ureilites. However, they are somewhat anomalous in that they have low carbon contents ( $<2.5$  wt% C) and do not clearly lie on a trend of Fo in olivine cores versus  $\delta^{13}\text{C}$  in carbon (Fig. 4).
2. Bulk carbon and nitrogen analyses of ureilite meteorites always include an isotopically light component which may be terrestrial in origin, even when freshly collected like those from Almahata Sitta. Stepped combustion analyses can help compensate for this and produce more accurate values for the indigenous light isotope component in meteorites. In terms of C abundance and isotopes, the Almahata Sitta samples are some of the least contaminated.
3. Although derived from a polymict ureilite breccia, the analyzed Almahata Sitta ureilite fragments more closely resemble unbrecciated (monomict) ureilites, rather than polymict ones, in terms of their carbon and nitrogen abundances and isotopic compositions. They are missing the low- $T$ , very heavy N component seen in other polymict ureilites. Thus, they do not represent the surficial regolith from which other polymict ureilites are considered to originate. A ubiquitous low- $T$  high N/C component may be related to the presence of non-indigenous extraterrestrial organic material.

4. Three of the Almahata Sitta ureilites show main release  $\delta^{15}\text{N}$  values that are around  $-90\text{‰}$ , similar to the values for acid residues and diamond concentrates from ureilites. On a diagram of  $\delta^{15}\text{N}$  vs  $\delta^{13}\text{C}$ , the Almahata Sitta ureilites do not overlap the fields of any of the carbonaceous or enstatite chondrites, and hence ureilitic carbon was not derived from these sources but shows evidence for a primordial nebula origin.

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*Editorial Handling*—Dr. Cyrena Goodrich

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## APPENDIX

Table A1. Full data set of stepped combustion analyses of five ureilitic fragments from Almahata Sitta.

Sample	Temperature (°C)	Carbon yield (ng)	Carbon yield (ppm/°C)	$\delta^{13}\text{C}$ (‰)	$\pm$	Nitrogen yield (ng)	Nitrogen yield (ppm/°C)	$\delta^{15}\text{N}$ (‰)	$\pm$
AS7	200	259.71	0.27	-23.53	0.26	4.02	0.00	-0.08	0.27
	300	322.40	0.68	-23.90	0.27	3.73	0.01	17.11	0.50
	400	369.99	0.78	-23.87	0.28	6.53	0.01	48.53	0.67
	500	420.29	0.89	-5.98	0.27	6.98	0.01	59.59	0.75
	550	2570.06	10.85	-1.17	0.29	4.46	0.02	6.61	0.30
	600	20508.76	86.61	0.18	0.30	19.94	0.08	-88.02	0.40
	650	34261.67	144.69	0.29	0.00	14.61	0.06	-111.08	0.50
	700	13134.67	55.47	1.30	0.26	20.08	0.08	-79.30	0.32
	750	1460.37	6.17	1.39	0.46	2.30	0.01	-1.68	0.39
	800	292.59	1.24	-1.79	0.49	0.98	0.00	2.49	0.81
	850	331.60	1.40	-0.41	0.39	0.76	0.00	-2.94	2.03
	900	385.75	1.63	0.21	0.55	0.74	0.00	-5.03	3.07
	950	283.25	1.20	0.25	0.54	0.54	0.00	-9.84	9.30
	1000	151.84	0.64	-2.91	0.54	0.32	0.00	-5.97	15.50
1100	55.53	0.12	-2.94	0.53	0.28	0.00	0.00	3.82	
1200	62.65	0.13	-3.93	0.48					
AS22	200	270.61	0.26	-26.65	0.35	1.10	0.00	8.06	9.24
	300	170.82	0.33	-26.29	0.19	2.04	0.00	9.41	11.79
	400	172.77	0.34	-22.08	0.16	4.00	0.01	56.76	6.10
	500	356.96	0.70	-11.46	0.16	5.53	0.01	75.49	4.48
	550	2089.76	8.15	-6.96	0.16	2.55	0.01	54.50	7.92
	600	14398.76	56.15	-8.80	0.17	7.24	0.03	-50.68	4.15
	650	29421.67	114.73	-6.20	0.24	10.84	0.04	-92.43	5.02
	700	12298.67	47.96	-8.27	0.00	18.60	0.07	-62.49	4.91
	750	316.11	1.23	-8.09	0.23	1.01	0.00	84.84	17.98
	800	125.64	0.49	-11.83	0.44	0.22	0.00	47.03	203.03
	850	156.98	0.61	-7.51	0.91	0.33	0.00	24.79	52.85
	900	175.86	0.69	-8.21	0.75	0.35	0.00	22.57	51.32
	1000	238.73	0.47	-8.64	0.54	78.45	0.15	-29.27	3.32
	1100	119.57	0.23	-12.85	0.60	0.81	0.00	-20.18	16.81
1200	85.48	0.17	-12.76	0.71	1.00	0.00	-37.98	9.38	
1300	117.88	0.23	-13.20	0.66	2.26	0.00	-37.30	8.23	
1400	103.22	0.20	-20.27	0.59	2.71	0.01	-29.61	7.83	
AS27	200	286.66	0.26	-25.46	0.17	5.78	0.01	6.18	3.36
	300	484.57	0.89	-24.82	0.19	8.67	0.02	10.56	3.22
	400	519.72	0.95	-21.03	0.23	12.39	0.02	26.25	3.31
	500	693.43	1.27	-8.16	0.30	18.71	0.03	33.22	3.27
	550	2367.46	8.66	-2.95	0.25	8.84	0.03	23.23	3.17
	600	11554.76	42.28	-1.85	0.25	9.07	0.03	-17.94	3.22
	650	28952.67	105.94	-2.20	0.00	12.80	0.05	-59.93	5.45
	700	14840.67	54.30	-2.33	0.00	21.30	0.08	-62.18	5.14
	750	1770.77	6.48	-2.51	0.23	5.32	0.02	-0.15	3.89
	800	775.35	2.84	-3.31	0.32	1.45	0.01	29.73	8.71
	850	510.96	1.87	-5.24	0.45	1.10	0.00	48.63	11.47
	900	532.36	1.95	-4.41	0.55	1.44	0.01	51.77	11.76
	1000	1149.65	2.10	-4.43	0.79	3.56	0.01	20.62	4.88
	1100	455.96	0.83	-8.09	0.71	2.16	0.00	-32.81	5.51
1200	270.67	0.50	-4.89	0.38	0.81	0.00	-10.53	7.81	
1300	122.01	0.22	-7.25	0.73	1.57	0.00	-29.44	5.54	

Table A1. *Continued.* Full data set of stepped combustion analyses of five ureilitic fragments from Almahata Sitta.

Sample	Temperature (°C)	Carbon yield (ng)	Carbon yield (ppm/°C)	$\delta^{13}\text{C}$ (‰)	$\pm$	Nitrogen yield (ng)	Nitrogen yield (ppm/°C)	$\delta^{15}\text{N}$ (‰)	$\pm$
AS36	1400	133.68	0.24	-4.05	1.16	0.86	0.00	-5.95	8.85
	200	299.16	0.34	-28.73	0.18	4.76	0.01	5.62	0.95
	300	356.50	0.81	-26.54	0.17	4.09	0.01	14.13	1.09
	400	278.69	0.63	-22.70	0.20	3.44	0.01	32.40	1.42
	500	415.63	0.94	-10.42	0.18	4.93	0.01	46.69	1.26
	550	1544.86	7.01	-7.13	0.23	2.03	0.01	44.22	3.16
	600	10339.76	46.89	-6.38	0.19	2.58	0.01	-25.85	1.82
	650	42181.67	191.30	-4.88	0.36	7.85	0.04	-96.12	1.28
	700	37147.67	168.47	-5.00	0.00	4.55	0.02	-94.77	2.32
	750	8491.17	38.51	-5.54	0.42	1.68	0.01	-70.49	6.33
	800	410.69	1.86	-6.32	0.31	0.23	0.00	7.98	31.07
	850	227.31	1.03	-6.52	0.24	0.17	0.00	6.22	42.41
	900	88.39	0.40	-6.56	0.28	0.17	0.00	8.93	60.59
	1000	126.29	0.29	-8.19	0.47	0.58	0.00	-7.81	7.03
AS44	1100	170.93	0.39	-6.35	0.25	1.07	0.00	-9.55	2.41
	1200	99.39	0.23	-7.11	0.22	1.43	0.00	-14.19	2.20
	1300	54.28	0.12	-9.94	0.61	1.95	0.00	-53.29	4.31
	1400	17.56	0.04	-14.17	0.26				
	300	912.56	0.61	-26.54	0.13	6.13	0.00	18.79	1.44
	400	505.72	1.02	-20.21	0.21	3.89	0.01	73.75	2.40
	500	1292.42	2.61	-2.09	0.18	4.25	0.01	89.62	2.63
	600	26498.76	53.55	0.40	0.00	11.64	0.02	-67.32	1.46
	700	56249.76	113.68	0.43	0.16	25.55	0.05	-107.11	1.17
	750	5880.36	23.77	1.94	0.23	1.85	0.01	-87.99	7.39
	800	629.40	2.54	0.81	0.19	0.83	0.00	-33.25	14.66
	850	482.77	1.95	3.03	0.25	0.69	0.00	-27.60	16.67
	900	380.67	1.54	0.25	0.18	0.84	0.00	-28.12	12.18
	950	106.41	0.43	2.82	0.30	0.28	0.00	-1.83	12.59
	1000	49.51	0.20	-5.55	0.53	0.28	0.00	28.36	78.27
	1050	30.21	0.12	-11.28	0.29	0.19	0.00	17.45	99.01
	1100	17.39	0.07	-14.23	0.32	0.10	0.00	6.71	125.02
1150	22.89	0.09	-14.56	0.33	0.15	0.00	21.47	180.95	
1200	22.54	0.09	-13.27	0.29	0.22	0.00	1.55	15.22	
1250	24.30	0.10	-13.48	0.31	0.33	0.00	-9.00	19.39	
1300	30.44	0.12	-13.49	0.44	0.37	0.00	-20.14	33.05	
1400	57.24	0.12	-2.06	0.69	0.72	0.00	-17.97	9.65	