Uranium isotopic composition of carbonaceous chondrites. A. Kaltenbach[1], C. H. Stirling[1] and Y. Amelin[2], [1]Department of Chemistry and Centre for Trace Element Analysis, University of Otago, NZ (akaltenbach@chemistry.otago.ac.nz), [2]RSES, Australian National University, Canberra, Australia (yuri.amelin@anu.edu.au).

Introduction: The assumption that the $^{238}U/^{235}U$ ratio is homogeneous at 137.88 throughout the Solar System was negated by recent findings in calcium-aluminum rich inclusions (CAIs) [1-3] and terrestrial samples [4, 5]. However, no significant variations could yet been found in bulk meteorite samples [6, 7, 8]. Variations in the $^{238}U/^{235}U$ ratio have several implications for the knowledge about the nucleosynthesis of elements and the chronology of the early Solar System. Anomalies that show an excess of ^{235}U suggest that the short-lived isotope ^{247}Cm ($t_{1/2}$ = 16 My), formed in a r-process source, was probably present at the very beginning of the Solar System. Furthermore, heterogeneity in the distribution of uranium isotopes means that all samples have to be analyzed for their $^{238}U/^{235}U$ composition when determining ages via Pb-Pb dating [3, 9]. Here we present $^{238}U/^{235}U$ data of 12 different carbonaceous chondrites (subgroups CV3, CI1, CR2, CM2), including Allende CAI and chondrules data (Allende data revised from [3]).

Methods: The samples were processed under clean laboratory conditions in the Centre for Trace Element Analysis at the Department of Chemistry, University of Otago. Sample sizes varied from as little as 3ng U (Leoville) up to 440ng U (Dar al Gani 521). All samples were digested using HF, HNO_3 and HCl double-distilled acids in Savillex PFA vials. The uranium was extracted and purified via a two-stage ion exchange protocol, as described in [3, 8]. A $^{236}U/^{233}U$ double spike was added prior to digestion to correct for instrumental mass fractionation. Uranium isotopes were analyzed using a Nu Instruments Nu Plasma multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS). Sample measurements were bracketed with standard measurements (CRM 145) to monitor instrument performance. A standard value of $^{238}U/^{235}U$=137.84 was adopted based on results from [10, 11]. For easier assessment of differences, results are shown as the parts-per-10,000 deviation from the standard value composition in ε-notation ($\varepsilon^{238}U$). All blanks were <3pg U and therefore negligible. The errors are reported as ±2SE, scaled based on the errors of standard measurements on the same day.

Results: The results are summarized in Table 1. The average of all 12 bulk samples is 4.6 ±1.3ε (2S.E., $^{238}U/^{235}U$ = 137.776 ± 0.018). Only the ratio in Allende CAI SJ101 deviates from the bulk carbonaceous chondrite average outside the 2 SD error limits and Allende CAI and chondrules are not being within error of 2SE around the bulk average (Figure 1). Anomalous U isotopic ratios of the CAIs [1-3] suggest that the ratios of bulk samples might be influenced depending on the amount and type of inclusions they contain.

No systematic variations in $^{238}U/^{235}U$ can be resolved between the different meteorite groups, indicating that the increasing oxidation (CR-CV-CM-CI) and varying abundance of refractory inclusions and volatile contents do not necessarily result in an increased fractionation of uranium isotopes. It is especially noteworthy that Orgueil (CI group) does not show any deviation from the average, despite being the only meteorite in this study which does not contain chondrules, and being the chemically most primitive.

Subtle variability in the uranium isotopic composition between samples, even within meteorite groups, is observed, although all bulk samples are within 2SE error of the bulk average. The largest variation is seen in the ungrouped C3-chondrite Dar al Gani 430, which is not in agreement with several other chondrite values. Another example are the average values of DaG 521 and Northwest Africa 2140 that are not in agreement with the average values for Allende and the ungrouped C3-chondrite DaG 430. If these differences are caused by U isotopic variations between inclusions and matrices or 'stable' isotope fractionation of uranium throughout the meteorite can only be determined by analyzing different mineral phases and inclusions from the same sample and comparing the results to the closure times and metamorphic states of the mineral phases.

The variations of several ε-units between single samples (e.g. Mighei (CM2, -0.1ε) and DaG 430 (C3, ungrouped, 8.2ε)) show that it is necessary to analyze and apply the $^{238}U/^{235}U$ ratios of samples, especially of inclusions, to ensure accurate Pb-Pb dating and a reliable chronology.

Conclusions: Although the observed variations in the $^{238}U/^{235}U$ ratios of the carbonaceous chondrites analyzed in this study are relatively small, they do show that meteorites can have statistically relevant differences, even within one meteorite group. Further investigation with improved resolution and of different mineral phases might reveal bigger anomalies within meteorite groups or differences between them and might give evidence whether U isotopic variability is dependent on their oxidation state, metamorphic history or other causes.

References: [1] Brennecka G. A. et al. (2010) *41st LPS*, Abstract #2117. [2] Brennecka G. A. et al. (2010) *Science*, *327*, 449-451. [3] Amelin Y. et al. (2010) *EPSL*, *300*, 343–350. [4] Stirling C. H. et al. (2007) *EPSL*, 264, 208-225. [5] Weyer S. et al. (2008), *GCA*, *72*, 345-359. [6] Friedrich J. M. et al. (2004) *LPS XXXV*, Abstract #1575. [7] Stirling C. H. et al. (2006) *EPSL*, *251*, 386-397. [8] Stirling C. H. et al. (2005) *GCA*, 69, 1059-1071. [9] Amelin Y. (2010) *41st LPS*, Abstract #1648. [10] Richter S. et al. (2010) *IJMS*, *295*, 94-97. [11] Condon, D. J. et al. (2010) *GCA*, *74*, 7127-7143

Figure 1: ε^{238}U data for the averages of 12 bulk carbonaceous chondrites, one CAI and one set of chondrules. The average of all bulk carbonaceous chondrites analyzed (solid line), as well as the 2SE (dashed lines) and 2SD (dotted lines) error limits of the bulk samples are also given

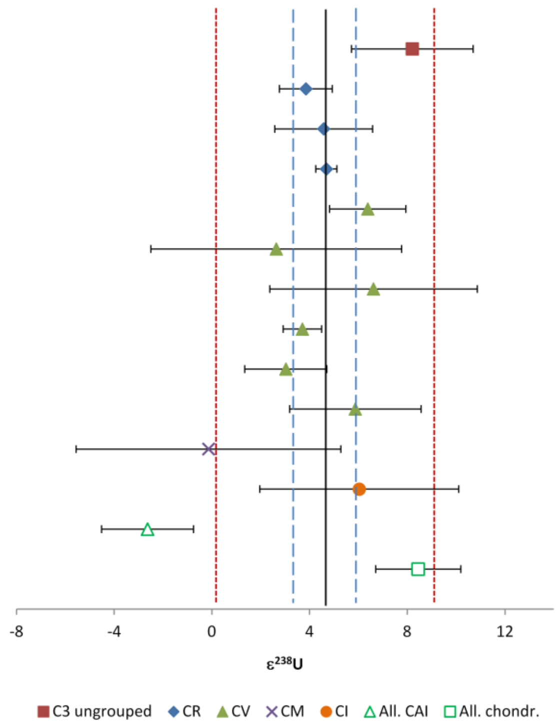

Table 1: ε^{238}U and uranium concentration data for samples of 12 different carbonaceous chondrites. Av. stands for weighted averages, depending on length (= ng used) of analysis.

Sample	Type	ε^{238}U	2SE	n	ppb U
DaG 430	C3 ungr., bulk	8.2	2.5	1	14
Acfer 059 a	CR2, bulk	3.4	1.8	4	247
Acfer 059 b	CR2, bulk	4.4	0.5	3	491
Acfer 059 av.	*CR2, bulk*	*3.9*	*1.1*	*7*	
NWA 721	CR2, bulk	4.6	2.0	1	69
NWA 801	CR2, bulk	4.7	0.4	2	103
Allende a*	CV3, bulk	9.0	2.4	1	12
Allende b*	CV3, bulk	5.0	2.2	1	13
Allende c*	CV3, bulk	7.3	1.6	1	15
Allende d*	CV3, bulk	5.2	1.8	1	12
Allende e*	CV3, bulk	5.9	1.5	1	14
Allende av.	*CV3, bulk*	*6.4*	*1.6*	*5*	
Leoville	CV3, bulk	2.6	5.1	1	18
Vigarano a	CV3, bulk	4.0	4.3	1	16
Vigarano b	CV3, bulk	8.2	4.4	1	17
Vigarano av.	*CV3, bulk*	*6.6*	*4.3*	*2*	
DaG 521 a	CV3, bulk	3.6	0.7	2	417
DaG 521 b	CV3, bulk	3.8	0.5	2	441
DaG 521 av.	*CV3, bulk*	*3.7*	*0.8*	*4*	
NWA 2140	CV3, bulk	3.0	1.7	3	127
NWA 4818 a	CV3, bulk	5.2	1.7	1	13
NWA 4818 b	CV3, bulk	7.9	2.5	1	17
NWA 4818 av.	*CV3, bulk*	*5.9*	*2.7*	*2*	
Mighei	CM2, bulk	-0.1	5.4	1	14
Orgueil a	CI1, bulk	3.8	3.6	1	25
Orgueil b	CI1, bulk	7.8	5.0	1	23
Orgueil av.	*CI1, bulk*	*6.0*	*4.1*	*2*	
Average total		4.6	1.3		
Allende*	CAI SJ101	-2.6	1.9	1	65
Allende*	chondrules	8.5	1.7	1	26

*previously published in [3]