Contents lists available at ScienceDirect

Earth and Planetary Science Letters

journal homepage: www.elsevier.com/locate/epsl

Meteoritic and asteroidal amino acid heterogeneity: Implications for planetesimal alteration conditions and sample return missions

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ARTICLE INFO

Edited by Dr O Mousis

Keywords:
Carbonaceous chondrite
Heterogeneity
Planetesimal
Aqueous alteration
Amino acid and meteorite

ABSTRACT

Carbonaceous chondrites (CC) and asteroid return samples contain amino acids (AA), which are essential for an origin of life on the early Earth and can provide important information concerning planetesimal alteration processes. While many studies have investigated AA from CC, separate studies have often found differing abundances for the same meteorite. Accordingly, analytical bias, differing terrestrial contamination levels and intrinsic sample heterogeneity have been proposed as potential reasons. However, current analytical techniques allow for the analysis of several mg-sized samples and can thus enable an investigation of AA heterogeneity within single meteorite specimens. Here, such an analytical technique is applied to characterise the AA in triplicate aliquots of three CCs. The results indicate that CCs are heterogenous in terms of their AA at the mm-scale. Furthermore, the results help to further constrain the effects of planetesimal alteration on organic matter and the requirements of future sample return missions that aim to obtain organic-bearing extraterrestrial materials.

1. Introduction

Carbonaceous chondrites (CC) and C-type asteroids (e.g. Ryugu) represent the surviving fragments of planetesimals, which formed within the early Solar System. Such planetesimals accreted from dust, ice and organic matter (OM) distributed within the protosolar nebula (PSN)/protoplanetary disk (PPD) and preserve information from both before and after the formation of the solar system (Nakamura et al., 2022; Ota et al., 2023; Potiszil et al., 2023b). For instance, gas and dust emitted from stars is thought to have collected in the interstellar medium (ISM) and given rise to molecular clouds, where the gases condensed to yield ice that formed upon the dust grain surfaces (Potiszil et al., 2023b; Sandford et al., 2020). Subsequent UV and cosmic ray interactions then transformed the original materials to generate new products, which could have included a large array of organic compounds (Glavin et al., 2018; Martins, 2019; Potiszil et al., 2023b). Meanwhile, within the PSN, UV-X-ray interactions with gas and ice could have led to the formation of OM and the alteration of existing OM (Henning & Seme 2013; Sandford et al., 2020; Walsh et al., 2014).

After the formation of planetesimals, those that possessed enough ice and which formed neither too early nor too late, could have supported liquid water, as a result of the heat supplied by the decay of short-lived radionuclides (McSween & Huss, 2022). During the period of liquid water activity, aqueous alteration converted the anhydrous amorphous and mineral dust components into secondary phases (Brearley, 2006). Additionally, aqueous alteration is also thought to have synthesised new OM from inorganic precursors and/or existing OM (Glavin et al., 2018; Potiszil et al., 2023b). While the main phase of aqueous alteration is indicated to have only lasted for several million years (Fujiya et al., 2013; 2012; Tanaka et al., 2024), subsequent impact events have been suggested to result in later limited episodes of aqueous alteration (Jilly-Rehak et al., 2017).

Organic matter is itself comprised of insoluble or macromolecular organic matter (IOM/MOM) and soluble or free organic matter (SOM/FOM) (Glavin et al., 2018; Sephton, 2013). The IOM/MOM component represents a complex macromolecular polymer that is not soluble in solvents. Meanwhile, the SOM/FOM component comprises smaller molecules than the IOM/MOM component, which are free from any macromolecules, and are soluble in solvents. Amino acids (AA) are one constituent of the SOM/FOM fraction and have received particular interest due to their essential role in an origin of life on Earth (Naraoka et al., 2023; Parker et al., 2023; Potiszil et al., 2023b, 2023a). While AA

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https://doi.org/10.1016/j.epsl.2025.119205
Received 2 September 2024; Received in revised form 7 January 2025; Accepted 9 January 2025

have been formed in various ISM/outer PSN ice analogue experiments, no amino acids have yet been definitively detected within the ISM or the PSN/PPD of other solar systems (Glavin et al., 2018; Henning & Seme, 2013; Martins, 2019; Potiszil et al., 2023b; Sandford et al., 2020; Walsh et al., 2014).

Alternatively, a variety of AA have been synthesised in experiments simulating planetesimal aqueous alteration (Glavin et al., 2018; Martins, 2019; Potiszil et al., 2023b). Furthermore, certain characteristics of AA in CC have been proposed to result from the conditions of aqueous alteration that relate to different CC and asteroid samples. For example, more aqueously altered CC (e.g. CI1 chondrites) tend to have larger abundances of β-AA (the amine is located on the 2nd C after the carboxyl group) compared to less aqueously altered CC (e.g. CM2 chondrites), which have higher abundances of α-AA (the amine is located on the 1st C after the carboxyl group) (Glavin et al., 2018; 2010). Such observations have been suggested to arise due to the higher stability of β-AA, compared to α-AA, during oxidation associated with higher levels of aqueous alteration, or as a result of a greater generation of α-AA via Strecker synthesis at lower levels of aqueous alteration(Martins et al., 2015). Accordingly, the β-alanine/glycine ratio has been found to increase with increasing aqueous alteration level for a given group of CC (Glavin et al., 2010; Martins et al., 2015; Modica et al., 2018).

Alternatively, it has been recently proposed that α-AA could be converted to β-AA and γ-AA (the amine is located on the 3rd C after the carboxyl group) during aqueous alteration due to distinct geo-electrochemical conditions that could arise if steep pH and redox gradients were present on planetesimals (Li et al., 2023). The aforementioned conditions could be present on planetesimals if they became gravitationally differentiated in terms of their water and rock components.

Another potential route to the formation of γ-AA, may be through thermal decarboxylation or deamination of α-amino dicarboxylic acids or diamino acids, which predominantly result in γ-AA and δ-AA (the amine is located on the 4th C after the carboxyl group) (Burton et al., 2012). Additionally, lactams (amino acid precursors) can be converted to γ-AA and δ-AA during aqueous alteration or during the acid hydrolysis steps employed in analytical procedures (Cooper & Cronin, 1995). Lactams have been shown to be thermally stable during heightened levels of aqueous alteration and may thus explain why γ-AA and δ-AA are comparatively more abundant in more aqueously altered CC, relative to α-AA and β-AA, than in less aqueous altered CC (Glavin et al., 2018). Meanwhile, some *n*-ω-AA (linear straight-chained amino acids), including β-alanine and γ-aminobutanoic acid, could have been formed via Fischer-Tropsch type (FTT) synthesis or Haber-Bosch process (HBP) reactions, during the cooling of CO, H_2, and N_2 gases that had become trapped within pore space(Burton et al., 2012). Therefore, there are many potential ways to form the AA present in CC and asteroids and the types of AA present are indicative of these different synthetic processes.

In the case of meteorite samples, such as CC, AA can be introduced from the terrestrial biosphere once they have fallen and even during their curation, once they are retrieved (Chan et al., 2020). In order to mitigate this, the interior portions of meteorites are often studied and the abundances of common protein forming amino acids that are not typically abundant in CC (e.g. phenylalanine and tyrosine) can be evaluated (Engel & Nagy, 1982). Additionally, chiral information can be used to assess the likelihood of terrestrial contamination, as all living organisms on Earth utilise l-AA (left-handed AA enantiomers) in their proteins (Glavin et al., 2018). However, excesses of l-AA, compared to d-AA (right-handed AA enantiomers), have been observed for non-protein forming AA, such as isovaline (Pizzarello et al., 2003), and so this is not a full-proof method for assessing the level of terrestrial contamination in meteorite samples (Chan et al., 2020). Alternatively, C, H and N isotopes can be used to evaluate the level of terrestrial contamination, with extraterrestrial AA often possessing enrichments in the heavy isotopes of these elements (Glavin et al., 2018). Nevertheless, large sample sizes (0.1 g to >1 g) are required to get precise compound specific isotope data (Chan et al., 2016; Elsila et al., 2012; Engel et al., 1990; Pizzarello et al., 1994; Pizzarello & Huang, 2005) and this makes it difficult to use such isotopic analyses for particularly precious samples, such as asteroid return samples or for certain rare CC (e.g. CI chondrites).

Conversely, for asteroid return samples, which have been kept in ultra-clean environments, the level of terrestrial contamination can be more easily minimised, compared to meteorite samples that have been exposed to the terrestrial biosphere. Accordingly, due to the expectedly low levels of terrestrial contamination, recent analytical developments and the very small Hayabusa2 return sample amounts (~5.4 g in total), the analysis of very small sample sizes (several mg to 10′s of mg) were employed for the Ryugu SOM/FOM analyses, including AA analysis (Nakamura et al., 2022; Naraoka et al., 2023; Parker et al., 2023; Potiszil et al., 2023a). However, the use of such small sample sizes raised the question of how representative the analytical results are of the materials they were sourced from? This question forms part of a wider debate on the heterogeneity of AA within CC samples (Elsila et al., 2016; Glavin et al., 2018; Potiszil et al., 2023b) and whether the differences in AA abundance observed between studies are truly due to indigenous heterogeneity, or possibly differences in the amount of contamination introduced during curation (Aerts et al., 2016; Chan et al., 2020), or even biases introduced from the different analytical procedures utilised (Potiszil et al., 2023b; Simkus et al., 2019). Despite such issues, the recent analytical advancements, which were applied to the Ryugu samples, have made the determination of AA abundances in milligram-sized meteorite samples possible (Potiszil et al., 2023a). Therefore, it is now possible to investigate the heterogeneity over the mm-scale within individual meteorite samples.

The aim of the current study is to better understand the heterogeneity of AA within specific meteorites and asteroid samples (intra-sample heterogeneity) and between different meteorites and asteroid samples (inter-sample heterogeneity). Therefore, the current study will discuss the results of quantitative AA analyses of ~2 mg triplicate aliquots of three different CCs, separated by more than several mm. The samples were analysed using the same analytical technique employed for two Ryugu particles (Potiszil et al., 2023a), including blank correction. Furthermore, the whole analytical procedure was carried out in the same clean-room environment used for the Ryugu analyses and eight serpentine blanks were put through the entire procedure, along with the meteorite samples, in order to ensure terrestrial contamination was accounted for.

2. Methods

2.1. Sample preparation and extraction

Three ~2 mg aliquots (see supplementary materials) each of Murchison (CM2), Aguas Zarcas (CM2) and Orgueil (CI1) were prepared by ultra-microtoming three mm-sized chips of each meteorite. The chips were generated from interior fragments of each meteorite and were obtained from regions of the fragment that were separated by >2 mm. The method for sample processing and analysis is the same as that reported previously (Potiszil et al., 2023a), but it is outlined in detail in the supplementary materials document.

2.2. UHPLC-OT-MS analysis

The amino acids were analysed using a binary pump ThermoFisher Scientific Vanquish^{TM} UHPLC unit, equipped with an Accucore^{TM} 150 Amide HILIC column. An Orbitrap Fusion Mass Spectrometer (Thermo Scientific) was used to conduct mass spectrometric measurements (see the supplementary information for more details). Amino acids were identified and quantitated using an in-house standard, which contained all the amino acids of interest and at concentrations ranging from 0.5 to 0.00075 μg g^{-1}.

2.3. Statistics

In order to understand how similar different aliquots of the three meteorites were to one another and thus deduce a sense of intra-sample heterogeneity, a Bayesian statistical approach was applied to the AA abundance data and the ratios calculated from this data. A Markov chain Monte Carlo (MCMC) algorithm was used, because this allowed for the generation of probability distributions for the abundance and ratio data for each aliquot. A normal distribution was assumed as the prior distribution for the AA concentration data of each aliquot that was analytically obtained by the current study and this was used to obtain a representative value. Subsequently, the representative value was used to generate the posterior distributions for the aliquots, but using a Cauchy distribution as the prior distribution. The deviation among the aliquots was then calculated from the posterior distributions. For more information concerning the Bayesian statistical approach employed here, please see the supplementary material.

3. Results

3.1. Intra-sample heterogeneity

The AA were detected and quantitated (Table 1) using a highly sensitive technique recently applied to the Ryugu return samples (Potiszil et al., 2023a). Chiral information was not obtained, due to the focus of the method on obtaining reliable AA abundances from the very small sample sizes associated with the Hayabusa2 mission (Potiszil et al., 2023b) (~1–2 mg of sample was used per an extraction). The use of small sample sizes is also essential for understanding the heterogeneity at and above the mm-scale, which is a major aim of the current study. Additionally, all samples recorded low levels of phenylalanine and tyrosine and reported higher abundances of certain non-protein AA, such as isovaline and allothreonine, compared to the protein forming isomers of these AA (i.e. valine and threonine) (Figs. 1–3). As will be mentioned in later sections, the average abundance ratios for a number of AA for Murchison and Orgueil reported here are similar to previous studies which used chirality to investigate terrestrial contamination. Therefore, the influence of terrestrial contamination for the data reported here is likely very limited.

All of the CCs demonstrated heterogeneity for the majority of their AA among their 3 aliquots. AA concentration data was determined to be heterogenous, on comparison of the data for two aliquots of the same meteorite, if the overlap of the probability distributions of the 2 groups of data were low (see table S1 and S2 in the supplementary information for more details).

On comparing the β-alanine/glycine (β-Ala/Gly) ratios for the two Murchison aliquots that recorded β-alanine above the limit of quantitation (LOQ), the MCS1 value (0.42) was found to have a low overlap (0.09) of its probability distribution with that of the MCS2 value (0.86). Furthermore, as the full data of other studies is not available, it is not possible to calculate probability distributions, to compare the data reported here and elsewhere. However, the values for Murchison from other studies (0.41–0.43) (Ehrenfreund et al., 2001; Glavin et al., 2010), which used 10′s of mg to 100′s of mg, are similar to the MCS1 value, but not the MCS2 value (Fig. 1). Moreover, the average value for Murchison from the current study (0.28), calculated by summing all the β-alanine abundances and dividing them by the sum of the glycine abundances, appeared different when compared to both MCS1 and MCS2, but was similar to the values reported by previous studies (Ehrenfreund et al., 2001; Glavin et al., 2010). Orgueil demonstrated a similar relationship, with the average value (3.24) appearing different to the values for ORG1 and ORG3, but being similar to those of previous studies (Ehrenfreund et al., 2001; Glavin et al., 2010) (2.90–3.16) (Fig. 2). Note that the large difference between the β-Ala/Gly ratios of the aliquots and average

Table 1
Blank corrected amino acid concentration data in ng/g for the three aliquots of the three carbonaceous chondrites investigated here. *Av.* = average, S.D. = standard deviation, BDL = below the detection limit, BQL = below the quantitation limit, MCS = Murchison, AZC = Aguas Zarcas and ORG = Orgueil. Note that the aliquot weight is reported in the supplementary materials. Gly = glycine, β-Ala = β-alanine, Ser = serine, Dmg = N,N-dimethylglycine, α-Aba = α-aminobutyric acid, Mal = N-methylalanine, α-Aib = α-aminoisobutyric acid, γ-Aba = γ-aminobutyric acid, Thr = threonine, Alt = allothreonine, Hse = homoserine, Asp = aspartic acid, Val = valine, Nva = norvaline, Iva = isovaline, Glu = glutamic acid, Pip = pipecolic acid, Tle = tert-leucine, Ale = alloleucine, Ile = isoleucine, Leu = leucine, Nle = norleucine, α-Adp = aminoadipic acid, Hcl = homocycloleucine, α-Apm = α-aminoapimelic acid, Phe = phenylalanine and Tyr = tyrosine.

Analyte	MCS1		MCS2		MCS3		AZC1		AZC2		AZC3		ORG1		ORG2		ORG3	
	Av.	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.	*Av.*	S.D.
Gly	2925	205	2225	272	6132	327	4015	1016	BDL	BDL	3055	356	1532	264	2579	617	1332	239
β-Ala	1226	209	1924	341	BDL	BDL	4252	853	BDL	BDL	2824	662	8531	1720	BQL	BQL	9083	708
Ser	791	167	900	238	2301	240	BQL	BQL	BDL	BDL	BQL	BQL	BDL	BDL	BQL	BQL	BDL	BDL
Dmg	373	35	532	111	660	43	BQL	BQL	BDL	BDL	BDL	BDL	1966	149	617	168	476	27
a-Aba	420	0	BQL	BQL	505	15	BQL	BQL	BDL	BDL	BQL	BQL	BQL	BQL	BQL	BQL	98	7
Mal	78	39	600	131	525	52	BQL	BQL	BDL	BDL	402	74	BDL	BDL	69	20	BQL	BQL
a-Aib	1410	398	882	228	2030	91	BDL	BDL	223	29	512	27	616	106	1132	241	1974	645
γ-Aba	946	108	2423	583	1264	253	10,719	1502	1227	279	BDL	BDL	5677	1400	BDL	BDL	4416	848
Thr	170	17	BQL	BQL	246	13	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Alt	BQL	BQL	470	155	457	60	BQL	BQL	BDL	BDL	BDL	BDL	BDL	BDL	280	73	BDL	BDL
Hse	6534	1169	BDL	BDL	BDL	BDL	BQL	BQL	BDL	BDL	BQL	BQL	578	72	1068	268	BQL	BQL
Asp	861	86	BDL	BDL	2472	68	597	26	96	2	218	17	BDL	BDL	BDL	BDL	124	9
Val	376	11	BQL	BQL	729	40	188	7	BQL	BQL	80	14	BDL	BDL	99	36	BQL	BQL
Nva	57	2	115	24	206	28	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Iva	4024	855	2485	785	4120	842	376	116	227	54	232	70	1558	494	1543	441	3328	546
Glu	1220	68	165	42	1289	23	831	26	450	8	401	11	54	1	585	102	308	49
Pip	BDL	BDL	BDL	BDL	BDL	BDL	159	50	BQL	BQL	BDL	BDL	BQL	BQL	BQL	BQL	BQL	BQL
Tle	BQL	BQL	BDL	BDL	110	11	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Ale	40	1	59	15	153	9	BQL	BQL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Ile	62	10	BQL	BQL	104	36	76	20	BQL	BQL	71	2	BDL	BDL	67	26	BQL	BQL
Leu	87	8	104	33	183	11	108	17	BQL	BQL	BDL	BDL	BDL	BDL	194	73	BDL	BDL
Nle	61	4	247	44	139	16	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	120	37	BQL	BQL
a-Adp	416	61	BQL	BQL	411	72	75	10	BQL	BQL	BQL	BQL	BQL	BQL	BQL	BQL	BQL	BQL
Hcl	BQL	BQL	BDL	BDL	50	3	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
a-Apm	169	2	BDL	BDL	123	10	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BQL	BQL	BDL	BDL
Phe	47	2	BDL	BDL	68	5	60	1	21	2	BQL	BQL	BDL	BDL	62	20	BDL	BDL
Tyr	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL

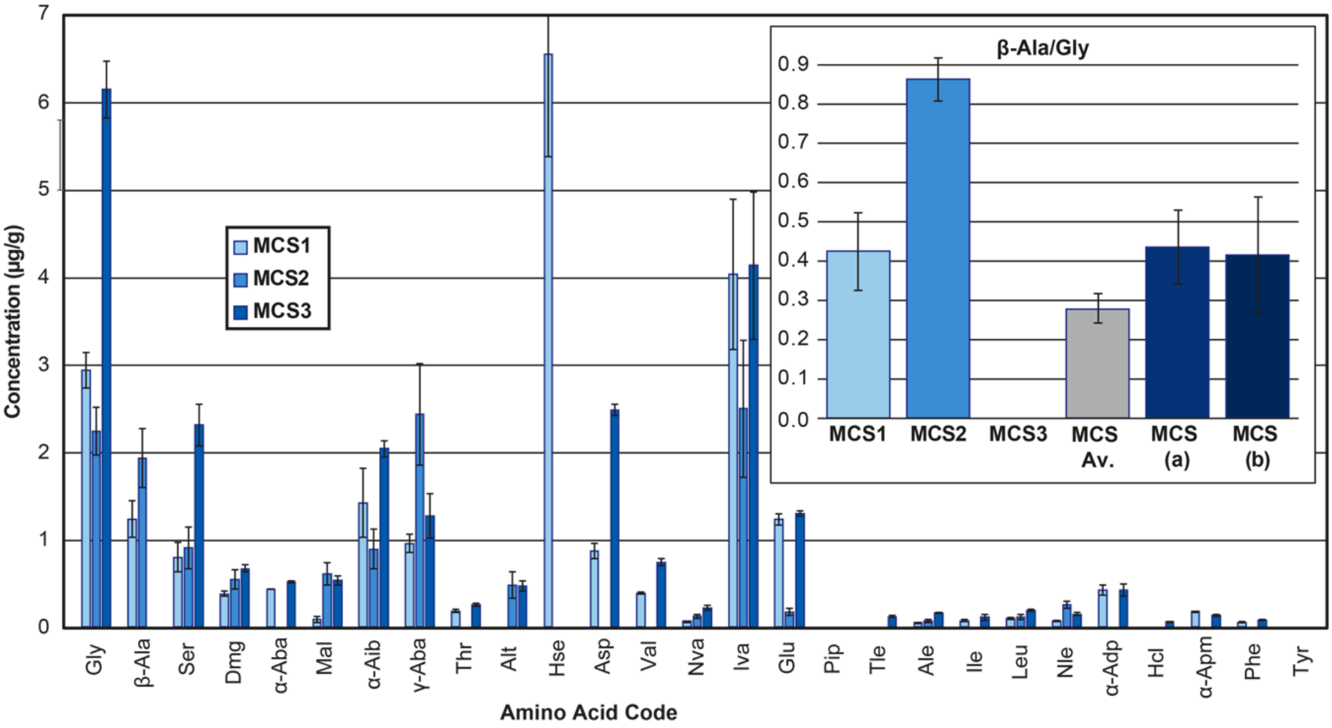

Fig. 1. A bar chart demonstrating the amino acid abundances in part per million (µg/g) for the three Murchison aliquots studied here. To the top right-hand side of the diagram is another bar chart showing the β-alanine/glycine (β-Ala/Gly) ratios of the Murchison aliquots reported here, as well as the average value calculated through summing all the β-alanine abundances and dividing these by the sum of the glycine abundances for the three aliquots. Additionally, the β-Ala/Gly ratios of two previous studies, (a) Ehrenfreund et al. (2001) and (b) Glavin et al. (2010), are also included for reference.

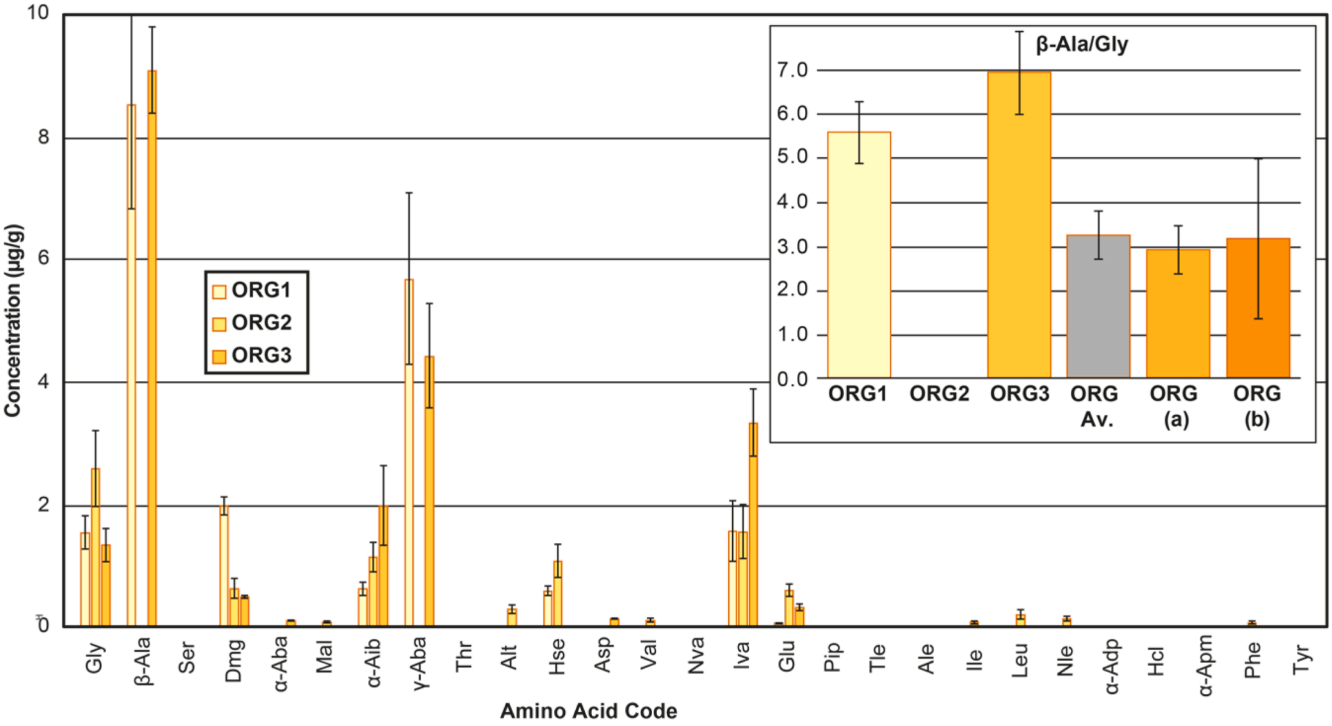

Fig. 2. A bar chart demonstrating the amino acid abundances in part per million (µg/g) for the three Orgueil aliquots studied here. To the top right-hand side of the diagram is another bar chart showing the β-alanine/glycine (β-Ala/Gly) ratios of the Orgueil aliquots reported here, as well as the average value calculated through summing all the β-alanine abundances and dividing these by the sum of the glycine abundances for the three aliquots. Additionally, the β-Ala/Gly ratios of two previous studies, (a)Ehrenfreund et al. (2001) and (b)Glavin et al. (2010), are also included for reference. The AA codes are the same as in Fig. 1.

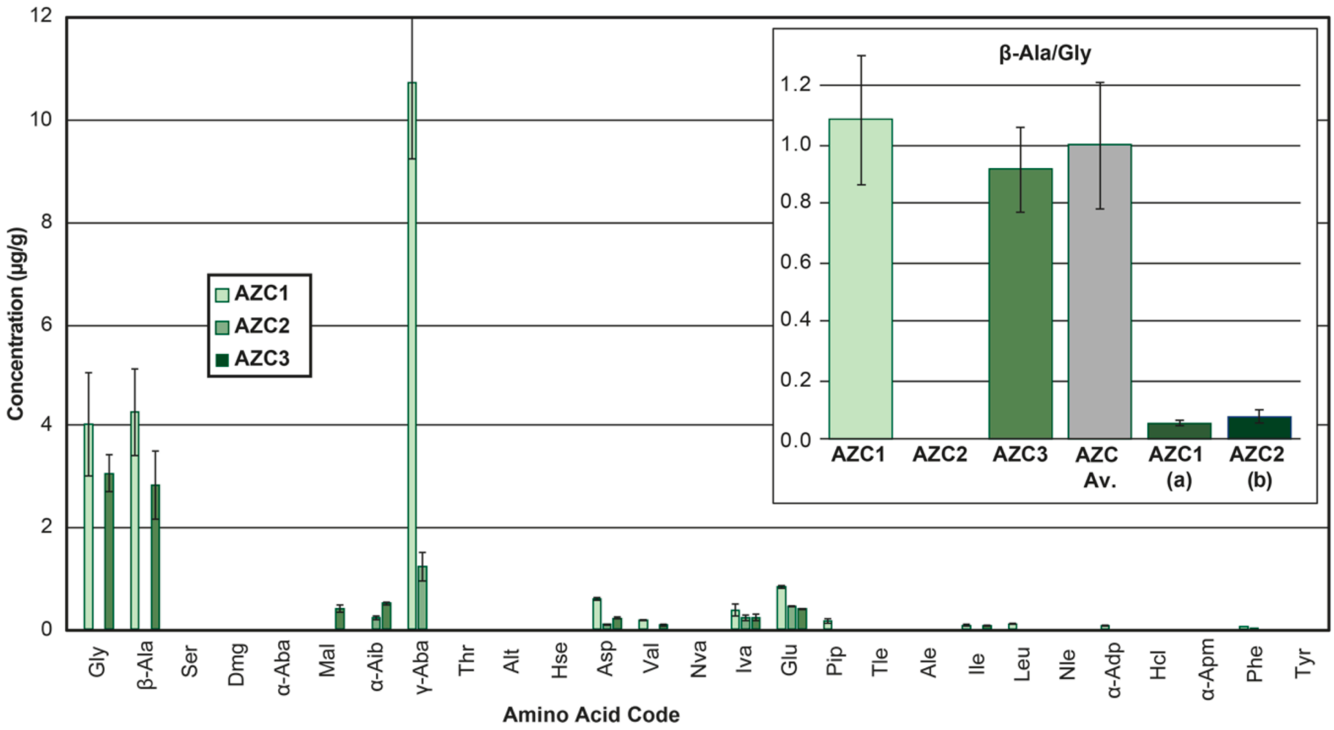

Fig. 3. A bar chart demonstrating the amino acid abundances in part per million (µg/g) for the three Aguas Zarcas aliquots studied here. To the top right-hand side of the diagram is another bar chart showing the β-alanine/glycine (β-Ala/Gly) ratios of the Aguas Zarcas aliquots reported here, as well as the average value calculated through summing all the β-alanine abundances and dividing these by the sum of the glycine abundances for the three aliquots. Additionally, two β-Ala/Gly ratios from a previous study (Glavin et al., 2021), (a) = UA 2741 and (b) = UA 2746, are also included for reference. The AA codes are the same as in Fig. 1.

value, for Murchison and Orgueil, arise from β-alanine being either below the limit of detection (LOD) or LOQ in one aliquot for both Murchison and Orgueil.

Aguas Zarcas gave a much more homogenous set of β-Ala/Gly ratios, with AZC1 and AZC3 having values (1.09 and 0.92, respectively) that had a high overlap of their probability distributions (0.62) and appeared very similar to the average value reported here (1.00). Nevertheless, AZC1, AZC2 and the AZC average value appeared to be very different to those of a previous study (0.05–0.08) (Glavin et al., 2021) (Fig. 3).

Meanwhile, the abundance of the AA N,N-dimethylglycine (Dmg) was found to have a low overlap on comparison of MCS1 with MCS2 and on comparison of MCS1 with MCS3 (Table S1). Similarly, for Orgueil there was a low overlap of the Dmg abundance probability distributions, when comparing ORG1 with ORG2 and ORG1 with ORG3. Whereas, in the case of Aguas Zarcas, none of the aliquots recorded Dmg above the LOQ of the method employed here.

3.2. *Inter-sample heterogeneity*

The Murchison CC recorded the largest variety of different AA for its aliquots, among all the three different meteorites analysed by this study (Fig. 1) and the Ryugu particles analysed previously (Potiszil et al., 2023a). Murchison also recorded the most instances where it was possible to quantify a given AA's abundance for all triplicate aliquots (13 out of the 27 AA investigated here). Furthermore, Murchison was the most heterogenous in terms of its glycine and β-alanine abundances, when considering the overlap of the probability distributions for its aliquots (Table S1), when compared to Aguas Zarcas and Orgueil.

Isovaline is a particularly rare AA on the earth (Pizzarello et al., 2003), and, as such, it can give an indication of AA heterogeneity among different meteorite and asteroid samples, with little potential for contamination to bias the results. When the β-Ala/Gly ratio is plotted against the isovaline/glycine (Iva/Gly) ratio (Fig. 4), it is clear that the Iva/Gly ratio appears distinct when comparing the values measured for ORG1, ORG3 and their average value to those reported for Orgueil from a previous study (Ehrenfreund et al., 2001). Nevertheless, both ORG2 and ORG3 show low overlap of their probability distributions to ORG1 (Table S2). Meanwhile, the values measured here for MCS1, MCS2, the average value and those reported by previous studies (Ehrenfreund et al., 2001; Glavin et al., 2010) appear very similar to each other, with MCS1 and MCS2 recording a high overlap of their probability distributions. For Aguas Zarcas, the probability distributions of the values reported by the current study are also highly overlapped with one another but appear very different to some of the values reported previously (Glavin et al., 2021). However, AZC1 (UA 2741) from the previous study (Glavin et al., 2021), appears to have a similar Iva/Gly ratio to the Aguas Zarcas samples measured here.

When comparing the probability distributions of the Dmg/glycine ratios (Dmg/Gly) for MCS1 to MCS2 and MCS2 to MCS3, there is a minimal overlap between them (Table S2). A similar relationship can be seen between ORG1 and ORG3, and between ORG2 and ORG3. Meanwhile, when plotting the Dmg/Gly against the β-Ala/Gly for the aliquots measured here and a Ryugu particle measured in a previous study (Potiszil et al., 2023a) (Fig. 5), it is clear that both the Murchison and Orgueil aliquots plot in very distinct regions from that of the Ryugu particle. Such an observation indicates that Dmg is distributed heterogeneously in most of the CC samples measured here. Furthermore, a moderate correlation is also observed for an increase in Dmg/Gly with increasing β-Ala/Gly ($R^2 = 0.4$), when excluding the Ryugu measurement (Fig. 5).

In the case of the γ-aminobutyric acid/glycine ratios (γ-Aba/Gly), these are observed to increase with increasing β-Ala/Gly. However, the slopes of this trend appear different for CM2 ($R^2 = 0.82$) and CI1 ($R^2 = 0.77$) CC (Fig. 6). Interestingly, Aguas Zarcas shows highly variable γ-Aba/Gly ratios, which arise due to the very high γ-aminobutyric acid abundance within one Aguas Zarcas aliquot.

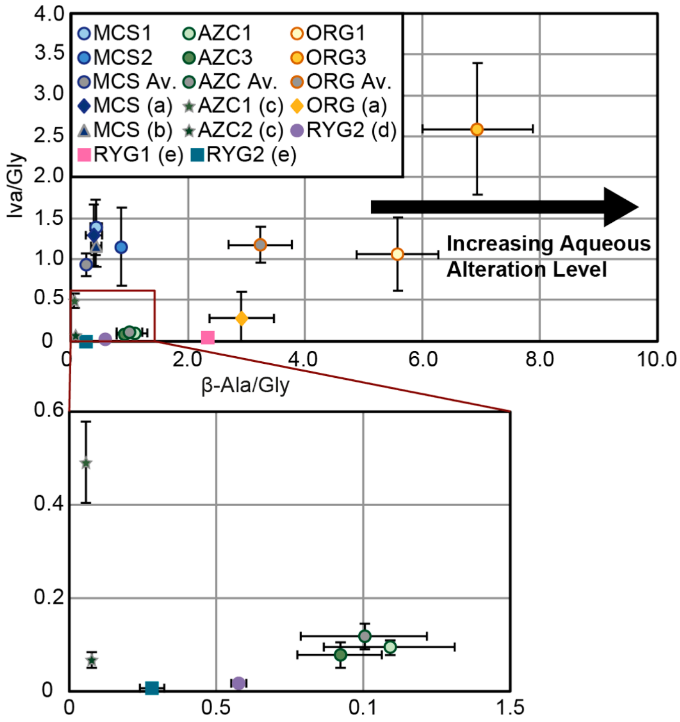

Fig. 4. A plot of the β-alanine/glycine ratio (β-Ala/Gly) against the isovaline/glycine ratio (Iva/Gly) for the aliquots of the three CC samples analysed here and data from previous studies. *Av.* = average, MCS = Murchison, AZC = Aguas Zarcas, ORG = Orgueil and RYG = Ryugu. (a) is fromGlavin et al. (2010), (b) is from Ehrenfreund et al. (2001), (c) is fromGlavin et al. (2021), (d) is C0008 fromPotiszil et al. (2023a) and (e) is from Parker et al. (2023) (RYG1 is A0106 and RYG2 is C0107). The black solid arrow indicates the effects of increasing aqueous alteration level, proposed previously (Glavin et al., 2010; Martins et al., 2015).

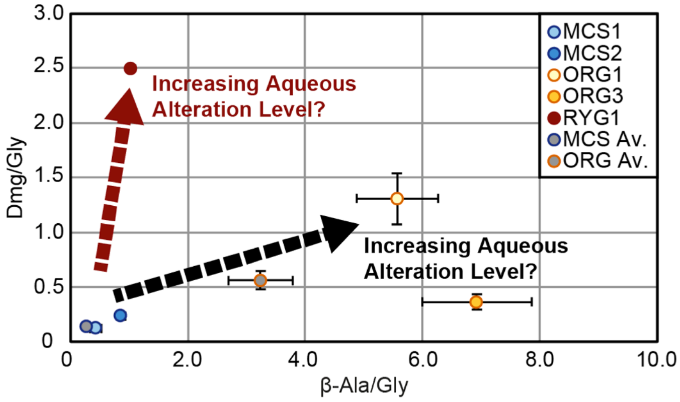

Fig. 5. A plot of the β-alanine/glycine ratio (β-Ala/Gly) against the N,N-dimethylglycine/glycine ratio (Dmg/Gly) for the aliquots of the three CC samples analysed here and data from a previous study. *Av.* = average, MCS = Murchison, ORG = Orgueil and RYG = Ryugu. The data for RYG1 is for A0022 from (Potiszil et al., 2023a). The black dashed arrow highlights a potential aqueous alteration trend between the Murchison aliquots and the Orgueil aliquots. The red dashed arrow highlights a potential aqueous alteration trend between the Murchison aliquots and a Ryugu particle.

4. Discussion

4.1. Progenitor planetesimal accretion and alteration

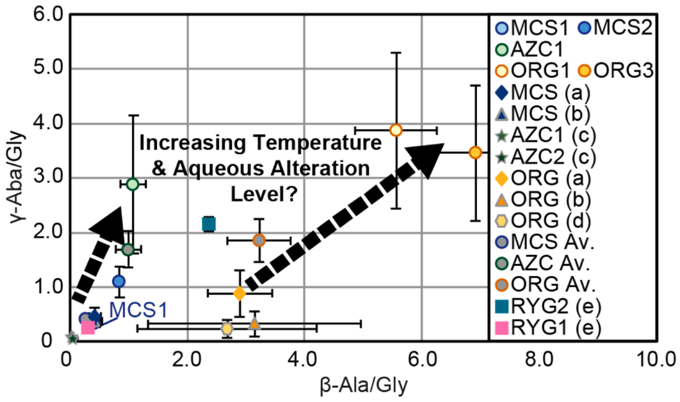

Fig. 6. A plot of the β-alanine/glycine ratio (β-Ala/Gly) against the γ-amino-butyric acid/glycine ratio (γ-Aba/Gly) for the aliquots of the three CC samples analysed here and data from previous studies. *Av.* = average, MCS = Murchison, AZC = Aguas Zarcas, ORG = Orgueil and RYG = Ryugu. (a) is from Glavin et al. (2010), (b) is from Ehrenfreund et al. (2001), (c) is from Glavin et al. (2021). The black dashed arrows indicate potential trends relating to an increase in temperature and aqueous alteration level.

All of the carbonaceous chondrites (CC) analysed here recorded heterogeneity among the majority of their ammino acids (AA) between their aliquots. Potential processes that can explain the heterogeneity are either the accretion of materials that are heterogenous in terms of their AA abundances and/or AA-precursors, or the presence of heterogenous conditions during aqueous alteration, with respect to space and possibly time. For example, if the AA were heterogeneously distributed among ice or dust grain surfaces, then these heterogeneities could be preserved after accretion into a planetesimal (Potiszil et al., 2023a). Alternatively, if the water/rock ratio (W/R), temperature, pH, fluid composition or composition/structure of the associated minerals was different throughout the planetesimal at and below the mm-scale, then heterogeneities in the AA abundances could be generated through the synthesis and/or alteration/destruction of AA (Glavin et al., 2010; Li et al., 2023; Martins et al., 2015). Nevertheless, a combination of both processes is also possible (Potiszil et al., 2023a).

Murchison is a petrographic type-2 CC and has been shown from both its mineralogy and β-Ala/Gly ratios to have been less affected by aqueous alteration, than the petrographic type-1 Orgueil (Brearley, 2006; Glavin et al., 2010). For instance, Orgueil is more hydrated than Murchison (Brearley, 2006) and higher W/R ratios have been proposed for Orgueil during planetesimal aqueous alteration (Glavin et al., 2018). Furthermore, β-alanine has been indicated to be more resistant to the effects of higher levels of aqueous alteration, compared to α-AA. The higher resistance of β-alanine to aqueous alteration, may relate to its higher stability at elevated temperatures or in more oxidising environments compared to α-AA (Martins et al., 2015). Accordingly, the β-Ala/Gly ratios of CC have been suggested to give an indication of the levels of aqueous alteration that a CC has experienced (Glavin et al., 2010; Martins et al., 2015).

Indeed, here it was found that similar to previous studies (Glavin et al., 2021, 2010; Martins et al., 2015), the β-Ala/Gly ratios recorded by Murchison and Aguas Zarcas, both CM2 chondrites, were generally lower than those recorded by Orgueil (Fig. 7). Moreover, the trend observed between Murchison and Orgueil in the β-Ala/Gly against the Dmg/Gly plot (Fig. 5) may also point towards differences in the aqueous alteration experienced on the Orgueil progenitor planetesimal, compared to the progenitor planetesimal of Murchison (Fig. 8) and potentially between CM2 and CI1 progenitor planetesimals in general. For instance, meteoritic Dmg has been suggested to form from glycine, formic acid and formaldehyde in aqueous mixtures (Potiszil et al., 2023a), through the Eschweiler-Clarke reaction (Clarke et al., 1933). As such, an increase in Dmg/Gly may be expected with higher aqueous alteration levels (indicated by higher β-Ala/Gly), which was observed

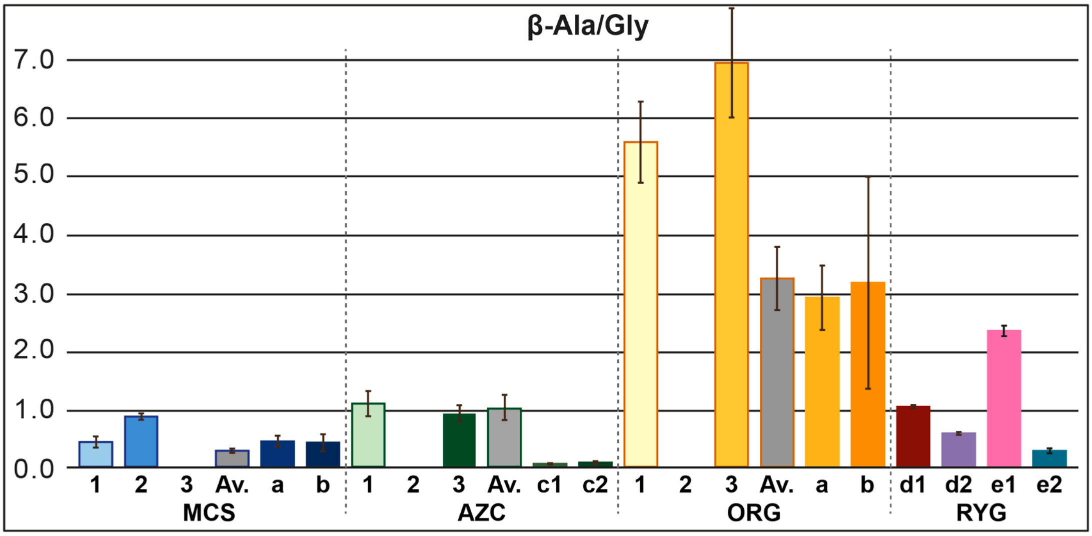

Fig. 7. A bar chart demonstrating the β-alanine/glycine ratios (β-Ala/Gly) for the three aliquots of the three CC samples studied here and for previous studies of these CC and Ryugu. The average value for each CC analysed by the current study was calculated through summing all the β-alanine abundances and dividing these by the sum of the glycine abundances for the three aliquots of a given CC. *Av.* = average, MCS = Murchison, AZC = Aguas Zarcas, ORG = Orgueil and RYG = Ryugu. (a) is from (Glavin et al., 2010), (b) is from (Ehrenfreund et al., 2001), (c) is from (Glavin et al., 2021), (d) is from (Potiszil et al., 2023a) (d1 is A0022 and d2 is C0008) and (e) is from (Parker et al., 2023) (e1 is A0106 and e2 is C0107).

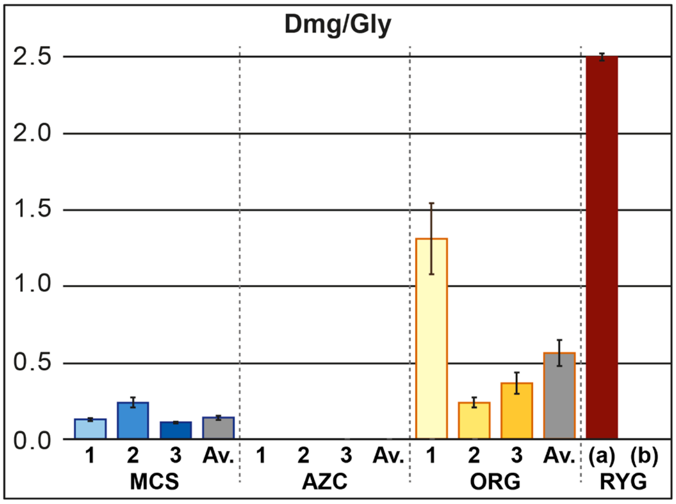

Fig. 8. A bar chart demonstrating the N,N-dimethylglycine/glycine ratios (Dmg/Gly) for the three aliquots of the three CC samples studied here and for a previous study of Ryugu. The average value for each CC analysed by the current study was calculated through summing all the Dmg abundances and dividing these by the sum of the glycine abundances for the three aliquots of a given CC. *Av.* = average, MCS = Murchison, AZC = Aguas Zarcas, ORG = Orgueil and RYG = Ryugu. (a) is A0022 and (b) is C0008 from (Potiszil et al., 2023a).

previously when comparing two Ryugu particles (Potiszil et al., 2023a), in terms of both their modal mineralogy and AA. Nevertheless, the trend observed here between Murchison and Orgueil is moderate, and so it is necessary for future experimental studies to establish if the Eschweiler-Clarke reaction could explain the moderate trend observed here.

Despite the apparent trend between Murchison and Orgueil in Fig. 5, Ryugu (RYG1;A0022) plots way off of the trend to much higher Dmg/Gly values, but with comparatively low β-Ala/Gly ratios compared to Orgueil. Meanwhile, Ryugu has been shown to be very heterogenous in terms of its fine scale aqueous alteration, as suggested by its β-Ala/Gly ratios and the heterogenous survival of presolar grains that are highly susceptible to aqueous alteration. Therefore, it is possible that while mineralogically similar, both Ryugu and Orgueil accreted distinct abundances of AA or AA precursor compounds. In such a case, both Ryugu and Orgueil may have experienced similar overall aqueous alteration conditions, which have generated similar modal mineral assemblages, but yielded different relationships between the level of aqueous alteration they experienced and the β-Ala/Gly ratio of their aliquots. As such, RYG1 may not follow the general trend potentially present in Fig. 6 between CM2 and CI1 chondrites, possibly due to accreting in a distinct region of the PSN/PPD. Indeed, it was previously indicated that Ryugu accreted material which was the least thermally processed of any material so far collected (Nakamura et al., 2022; Tanaka et al., 2024). Therefore, Ryugu may have accreted further out within the PSN/PPD than the progenitor planetesimals of Orgueil and the CM2 chondrites analysed here.

Similar to Ryugu, Aguas Zarcas also demonstrates considerable differences in its β-Ala/Gly ratios. However, such differences were observed on comparing the data obtained by the current study and that from a previous study (Glavin et al., 2021). Nevertheless, as mentioned above, the average data value from the current study and the value calculated for bulk Murchison samples from several previous studies (Ehrenfreund et al., 2001; Glavin et al., 2010) appear very similar. Therefore, differences in the analytical techniques applied are unlikely to be the reason for the differences in the above AA ratios between different studies. Furthermore, Aguas Zarcas shows extensive evidence of brecciation and consists of a series of different lithologies (Kerraouch et al., 2022; 2021). Additionally, the different Aguas Zarcas lithologies are thought to have experienced distinct aqueous alteration conditions (Kerraouch et al., 2022). Therefore, Aguas Zarcas could contain substantially different AA between these different lithologies as a result of a disparity in the planetesimal processing conditions they each experienced.

Indeed, the AZC1 sample analysed by the current study, contains a very high γ-Aba/Gly ratio, compared to those reported by a previous study (Glavin et al., 2021). As γ-AA can be produced during or survive at higher temperatures and degrees of aqueous alteration (Burton et al., 2012; Glavin et al., 2018; Kebukawa et al., 2017), compared to α-AA or β-AA, it may be that the high γ-Aba/Gly ratio, as well as the higher than Murchison β-Ala/Gly ratio, observed for AZC1 could arise from a localised heating event. Such an event might relate to an impact, which could help to explain why Aguas Zarcas appears brecciated. Indeed, Aguas Zarcas was found here to contain lower overall abundances and have a lower diversity of amino acids compared to Murchison, which could be explained if it experienced a higher amount of impact processing relative to Murchison. Essentially, the Aguas Zarcas parent body may have been exposed to many small impactors, which worked to brecciate the surface of the parent body and mix its various lithologies together, while also potentially destroying some of the amino acids originally present. Similarly, Orgueil has also been described as brecciated and several of the aliquots measured by the current study also recorded high γ-Aba/Gly compared to the average value and those reported for previous studies that used larger sample sizes (Ehrenfreund et al., 2001; Glavin et al., 2010).

Furthermore, such impact heating events could have melted ice and continued aqueous alteration (Jilly-Rehak et al., 2017), but at distinct temperature conditions to that experienced during aqueous alteration on the Aguas Zarcas and Orgueil progenitor planetesimals. The resulting localised aqueous alteration events (Brearley, 2006; Le Guillou et al., 2014; Le Guillou & Brearley, 2014), may help to explain the large variation in aqueous alteration level observed among different Aguas Zarcas lithologies and also the differences in the AA ratios measured here and in previous studies for the Aguas Zarcas and Orgueil, compared to Murchison.

4.2. *The formation and evolution of carbonaceous chondrites and Ryugu*

On integrating all the separate lines of evidence (Fig. 9), it would seem that the γ-Aba/Gly versus β-Ala/Gly plot (Fig. 6) suggests the presence of aqueous alteration dependent relationships between Aguas Zarcas and Murchison, but not between these two CM2 chondrites and Orgueil. Instead, Orgueil may have evolved along a different aqueous alteration pathway. Such a phenomena is also indicated by the distinct carbonate compositions found within Orgueil, compared to those in CM2 chondrites (De Leuw et al., 2010).

Furthermore, while Aguas Zarcas and Murchison (CM2) have both been determined as petrographic type 2, Aguas Zarcas has been shown to contain type 1 lithologies, indicating that some portions of Aguas Zarcas are more aqueously altered than Murchison (Brearley, 2006; Kerraouch et al., 2022; Le Guillou & Brearley, 2014). Such evidence supports the trend observed between Murchison and Aguas Zarcas in Fig. 6. Meanwhile, Orgueil is a type 1 CC and has distinct mineralogy to Murchison and Aguas Zarcas, containing mostly saponite and serpentine, instead of cronstedtite-tochilinite intergrowths and serpentine (Brearley, 2006; Howard et al., 2015; King et al., 2015; Le Guillou et al., 2014). Therefore, the mineralogy and petrographic type of the CCs studied here also supports a scenario where differences in aqueous alteration could lead to different trends for the amino acid ratios for CM2 and CI1 chondrites.

However, it is not clear if Orgueil and CM2 chondrites evolved along different aqueous alteration pathways due to the presence of different conditions during aqueous alteration (e.g. W/R ratio or temperature), or due to differences in the components accreted by two different planetesimals (e.g. AA or AA precursor abundances or primary mineralogy). Whereas, the Dmg/Gly versus β-Ala/Gly (Fig. 5) plot suggests that the CM2 chondrites studied here and Orgueil could have at least accreted similar AAs and/or AA precursor materials, which have evolved differently due to the different degrees of aqueous alteration experienced by their progenitor planetesimals. Meanwhile, Ryugu shows a very heterogenous β-Ala/Gly ratio and RYG1 has a very high Dmg/Gly ratio. Therefore, Ryugu may have accreted distinct abundances of AAs or AA precursor compounds compared to the CM2 chondrites and Orgueil.

Nevertheless, all the CCs and Ryugu likely accreted their AA and/or AA precursor-containing phases (such as ice or residues on mineral

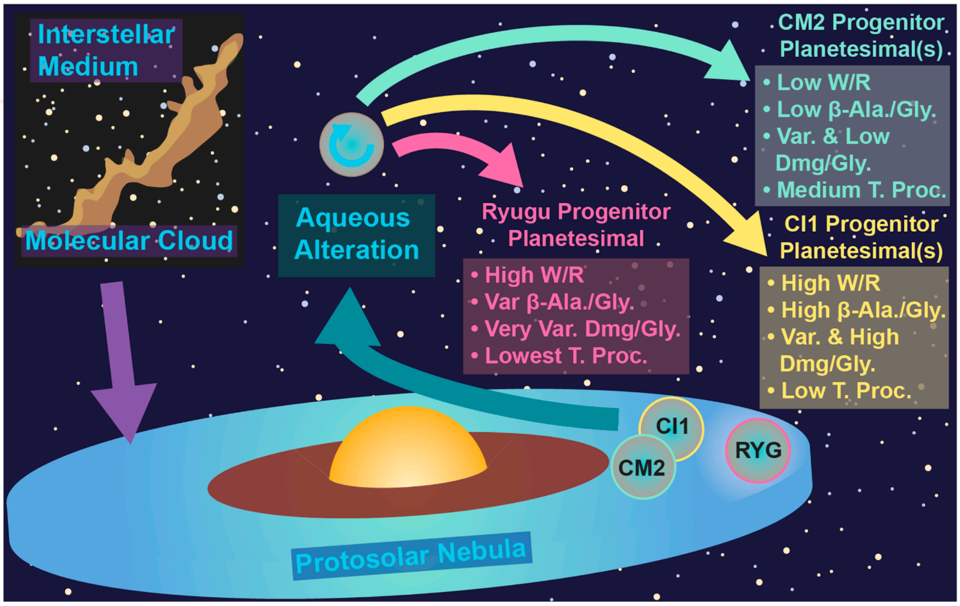

Fig. 9. The proposed origin of the progenitor planetesimals of the CC analysed here and Ryugu. AA and/or AA precursor compounds are generated within the ISM/PSN. The compounds are then accreted into icy planetesimals. Heat from radionuclides causes the melting of ice. The water-rich fluid transforms primary mineral phases into secondary mineral phases and alters and generates AA. The SOM/FOM produced depends on the conditions operating on the planetesimal, such as the W/R ratio and temperature, but also on the abundances of the AA and AA precursor compounds originally accreted. The Dmg/glycine (Dmg/Gly), γ-aminobutanoic/glycine and β-alanine/glycine (β-Ala/Gly) ratios determined here suggest the level of aqueous alteration (relating to W/R ratio and temperature) is likely the most important factor to explain the heterogeneity in AA abundances and ratios between CM2 and CI1 chondrites. However, initial differences in the accreted components may be the largest factor in determining the differences in the AA abundances and ratios observed between CI1 chondrites and Ryugu. As such, the progenitor planetesimal of CM2 and CI1 chondrites may have sourced similar, yet different parts of the PSN/PPD. The probably higher W/R ratio for CI1 chondrites (Zolensky et al., 1989), and their accretion of lower abundances of thermally processed (T. Proc.) materials (Nakamura et al., 2022; Tanaka et al., 2024), indicate that their progenitor planetesimal likely formed further out in the PSN/PPD, than that of CM2 chondrites. Meanwhile, the progenitor planetesimal of Ryugu likely accreted within a distinct region of the PSN/PPD to that of Orgueil and formed further out, as indicated by its thermal processing history (Nakamura et al., 2022; Tanaka et al., 2024).

surfaces) heterogeneously above the mm scale within their progenitor planetesimals. Such a finding is important, because it puts constraints on the style of aqueous alteration that could have occurred on icy planetesimals. If fluid routinely flowed through a lithified body over distances much greater than the mm-scale, such mm-scale heterogeneities in AA would not be possible, as the fluid and mineral surfaces would have become homogenised with AAs, at least at the mm-scale. The results of this study are compatible with either a scenario where convection did not occur (Bland et al., 2009), or in which the body acted as a convecting mudball (Bland & Travis, 2017). Whereas, models that employ the convective flow of fluid through rock, to redistribute SOM containing compounds (Li et al., 2023), must explain not only the lack of elemental fractionation (Bland et al., 2009; Bland & Travis, 2017), but also the presence of mm-scale SOM heterogeneity, such as that demonstrated here and elsewhere (Nakamura et al., 2022; Potiszil et al., 2020).

In terms of understanding whether CM2 and CI1 chondrites could have originated from the same parent body, the data reported here is not entirely conclusive. However, the distinct trend in the γ-Aba/Gly versus β-Ala/Gly plot (Fig. 6) for Orgueil, compared to Murchison and Aguas Zarcas, may indicate that the CM2 and CI1 chondrites were once part of different progenitor planetesimals, which experienced different aqueous alteration conditions. In such a scenario, the difference in the Dmg/Gly ratios between Murchison and Orgueil could also arise from differences in the components accreted by their progenitor planetesimals. Indeed, while glycine, formic acid and formaldehyde have been reported as products in either the ISM or ISM/PSN irradiated ice analogues (Oba et al., 2016; Zuckerman et al., 1971; 1970), no Dmg has been observed in the ISM or reported for irradiated ice analogue experiments. Instead, Dmg has only been proposed to form during planetesimal aqueous alteration (Potiszil et al., 2023a).

Accordingly, the CM2 chondrites and Orgueil could have originated from different progenitor planetesimals that contained different ice/dust ratios (essentially W/R ratios), as indicated by their distinct Δ^{17}O values (Nakamura et al., 2022; Tanaka et al., 2024). Upon the reaction of the Dmg precursors, the higher levels of aqueous alteration operating on the Orgueil progenitor planetesimal, may have led to higher overall Dmg/Gly ratios for Orgueil compared to Murchison and Aguas Zarcas. Furthermore, the Ca and Cr isotopes measured for CM and CI CC indicate that the material accreted by CI CC was less thermally processed than that accreted by CM CC (Nakamura et al., 2022). As such, the progenitor planetesimal of CI CC, including Orgueil, may have formed further out in the PSN/PPD than that of the CM CC, such as Murchison and Aguas Zarcas.

Moreover, current models, which aim to explain the origin of CM and CI chondrites from the same parent bodies (Li et al., 2023; Palguta et al., 2010), utilise stratigraphically differentiated planetesimals undergoing convection as a way of creating different temperature, oxygen isotope, W/R ratio and electrochemical conditions within separate regions of the body. However, such models fail to take into account the fact that elemental fractionation is not observed within individual CM and CI specimens and that planetesimals would not have accreted as rock, but instead as loosely gravitationally bound objects (Bland et al., 2009; Bland and Travis, 2017). Therefore, the body could have been free to convect as a mudball, which would have prevented water-rock differentiation (Bland & Travis, 2017). In such a scenario, silicate minerals, their associated water and the compounds dissolved within it, would circulate together. As such, heterogeneities in SOM could have been preserved either within the fluid or through adsorption onto newly formed secondary minerals (e.g. phyllosilicates) (Le Guillou & Brearley, 2014; Muneishi & Naraoka, 2021; Potiszil et al., 2020; Yesiltas & Kebukawa, 2016).

Nevertheless, petrographic type-1 and type-2 CC have been suggested to have experienced temperatures less than those that initiate the thermal decomposition of AA (>150 °C) (Chandru et al., 2013; Elmasry et al., 2020; Glavin et al., 2018; Lee et al., 2014; Li et al., 2023). The thermal decomposition of AA has been suggested as necessary to destroy α-AA and increase the relative proportion of β-AA, compared to α-AA (Li et al., 2023). However, the thermal history of type-1 and 2 CC progenitor planetesimals remains unclear. Most temperature estimates are based on mineral formation conditions or oxygen isotope fractionation models calculated for minerals (Glavin et al., 2018; Li et al., 2023). As the formation of secondary minerals could occur during the cooling of a planetesimal, temperatures of formation for such minerals do not necessarily give the peak aqueous alteration temperature. Thus, it is possible that petrographic type-1 and 2 CC could have been heated above 150 °C. Additionally, previous studies that have investigated the thermal decomposition of AA, have not taken into account the potential catalytic effects of the minerals present within the planetesimals during aqueous alteration (Chandru et al., 2013; Elmasry et al., 2020; Lee et al., 2014). Accordingly, at least some AA may decompose at lower temperatures than previously proposed. Future studies should investigate the stability of AAs that are associated with minerals during low-temperature aqueous alteration.

4.3. *Implications for future sample return missions*

The results of the current study indicate that both meteorite and returned asteroid samples contain inherent heterogeneity in their SOM components at the mm scale. As sample return missions (Hayabusa2 and OSIRIS-REx) have now brought C-type asteroidal materials back to Earth, it is important to understand how representative such samples are of the asteroids that they were sampled from. Accordingly, the data reported here, when compared with that reported previously (Ehrenfreund et al., 2001; Glavin et al., 2021; 2010; Potiszil et al., 2023a), indicates that bulk measurements of sample (>10′s of mg) can be representative of the AA in at least some meteorite-sized objects (10′s to 100′s of kg), as is the case for Murchison. For Orgueil, while the Iva/Gly and γ-Aba/Gly abundance ratios from different studies and the average values reported here do seem to differ, the β-Ala/Gly ratios appear to be similar. Thus, for some meteorites how representative a particular sample size is, appears to depend on the AAs that are investigated. However, some meteorites and asteroid samples, such as Aguas Zarcas and Ryugu, display very high levels of heterogeneity above the mm-scale and can yield large-scale variations in their AA abundances even for bulk measurements. As, such there is likely no particular sample size that can be truly representative for all meteorites and asteroids, in terms of AA. Instead, it is likely necessary to take multiple measurements from distinct parts of the meteorite or asteroid to understand the variation in the AA abundances and make average values.

As a result of the potential for both large-scale and small-scale variation in SOM abundance for C-type asteroids and CC meteorites, it is important to devise sample strategies accordingly. For instance, the Hayabusa2 mission visited two touchdown sites on Ryugu, which were separated by ~870 m (Nakamura et al., 2022). The sampling of two locations, separated by a large distance, enabled an investigation of whether large scale variations are present in terms of SOM across the Ryugu asteroid. It was found that the amino acid abundances differed between the samples from each touchdown site, but it was not clear if this related to a systematic asteroid scale difference in the aqueous alteration style or accretion source (Potiszil et al., 2023b).

Nevertheless, for one particle from each touchdown site, the AA abundances and modal mineralogy were determined (Potiszil et al., 2023a). It was found that the β-Ala/Gly ratio and Dmg abundance were higher in the TD1 sample, and this correlated with higher abundances of secondary minerals, including carbonate, as well as evidence for the mobilisation of trace elements beyond the mm-scale (Nakamura et al., 2022). Together this information pointed towards a higher level of aqueous alteration for the TD1 particle and that the Dmg could have formed during planetesimal aqueous alteration via the Eschweiler-Clarke reaction (Potiszil et al., 2023a). The above findings highlighted that it is essential to investigate the differences in the SOM and mineral abundances between small sample-sizes, but also between samples from distinct localities on an asteroid. Furthermore, it is also essential to analyse enough samples to calculate average abundances of particular regions of a given asteroid or meteorite specimen, so that regional variations can be investigated.

In the case of the OSIRIS-REx mission, a much larger sample size as 121.6 g was collected (Lauretta et al., 2022), than for the Hayabusa2 mission (5.4 g). Such a large sample size for Bennu, will allow a detailed investigation of the heterogeneity of SOM between small samples and the generation of an average value for Bennu. Through such an investigation, it will be possible to understand how representative individual particles are of the region from where they were collected and gauge the general heterogeneity of Bennu at the mm-scale or below. However, due to the sampling of only one location on Bennu, it will be impossible for the mission to understand if large-scale variation of SOM exists across the surface of Bennu. Therefore, future sample return missions should aim to combine the best parts of both OSIRIS-REx and Hayabusa2 and collect large quantities of sample from a number of separate locations, ideally even more than two.

5. Conclusions

The Murchison (CM2), Aguas Zarcas (CM2) and Orgueil (CI1) chondrites investigated by the current study, all demonstrated mm-scale heterogeneity in their AA abundances and ratios for the majority of the AA analysed. The heterogeneity is likely the result of a combination of heterogenous accretion of AA or AA precursors and the heterogeneities in aqueous alteration levels throughout the progenitor planetesimals of these carbonaceous chondrites with respect to space.

Furthermore, the differences observed between the CM2 and CI1 meteorites studied here, as well as the Ryugu asteroid samples, likely stem from differences in the accretion position of their progenitor planetesimals. The findings reported here are compatible with a scenario in which the W/R ratio generally increases with distance from the protosun and in which the protosolar nebular/protoplanetary disk is spatially heterogenous. In this scenario, the CM2s and CI1s may have formed relatively close together sourcing similar amino acids or amino acid precursors, but with the CI1s accreting more water ice as they formed further away from the sun. However, Ryugu should have formed in a distinct region due to its highly heterogenous nature and distinct AA composition, ratios and ratio trends to the CM2 and CI1 chondrites.

Meanwhile, differences in the AA abundance and ratio data observed between Murchison and Aguas Zarcas (especially concerning γ-aminobutanoic acid), may stem from where they were sourced in their parent asteroid. Murchison may represent a deeper source, less affected by small impacts than Aguas Zarcas, which may have been sourced from the regolith of its parent asteroid. Here it is proposed that the effects of impacts could locally raise temperatures and melt water ice, which could in turn lead to distinct later episodes of aqueous alteration.

Planetesimal aqueous alteration could not have proceeded via fluid flow through solid rock, as this is not supported by amino acid abundance or ratio heterogeneity. Instead, the findings reported here are compatible with either localised closed system aqueous alteration or a convecting mudball scenario.

Lastly, the findings reported here provide important information concerning future sample return missions. Due to the heterogenous nature of carbonaceous chondrite and asteroid return samples, future sample return mission must collect large sample amounts (such as those collected by NASA's OSIRIS-REx mission), but also from multiple sample sites across a bodies surface (such as was undertaken by JAXA's Hayabusa2 mission). Therefore, combining the best parts of OSIRIS-REx and Hayabusa2 are imperative for the success of future sample return missions by enabling the return of truly representative samples.

Research Data Link

Amino acid concentration data are included within Table 1 of this paper.

CRediT authorship contribution statement

Christian Potiszil: Writing – review & editing, Writing – original draft, Visualization, Resources, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Tsutomu Ota:** Writing – review & editing, Investigation. **Masahiro Yamanaka:** Investigation. **Katsura Kobayashi:** Writing – review & editing, Resources, Formal analysis, Data curation. **Ryoji Tanaka:** Writing – review & editing, Resources. **Eizo Nakamura:** Writing – review & editing, Resources.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Christian Potiszil reports financial support was provided by Japan Society for the Promotion of Science. Christian Potiszil reports financial support was provided by Japan Science Society. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

We are greatly indebted to PML members for their assistance in maintaining the laboratory, especially Kayo Tanaka and Yukari Shimizu whose daily efforts greatly aided the work carried out here. This work was supported by the Ministry of Education, Culture and Sports, Science and Technology (MEXT) of Japan through the JSPS Grant-in-Aid for Scientific Research (B) (assignment number: 24K17112) and the Sasakawa Scientific Research Grant (research number: 2024–2001).

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.epsl.2025.119205.

Data availability

The data is available in the following repository: DOI 10.17605/OSF.IO/WVN2M

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