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Journal Article

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Publication date:

2023-03-15

Permanent link:

<https://doi.org/10.3929/ethz-b-000598217>

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Originally published in:

Geochimica et Cosmochimica Acta 345, <https://doi.org/10.1016/j.gca.2023.01.020>

Funding acknowledgement:

182649 - Understanding the messages from space recorded in the noble gases to understand the formation of the solar system (SNF)

182901 - NCCR PlanetS – Funding of Equipment for the LIFE project (SNF)

193331 - Unlocking the origin of Earth's volatiles (SNF)



Contents lists available at ScienceDirect

Geochimica et Cosmochimica Acta

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

The noble gas and nitrogen relationship between Ryugu and carbonaceous chondrites



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ARTICLE INFO

Article history:

Received 4 October 2022

Accepted 19 January 2023

Available online 26 January 2023

Associate editor: Frederic Moynier

Keywords:

Ryugu

Hayabusa2

ABSTRACT

Carbonaceous chondrites are considered to have originated from C-type asteroids and represent some of the most primitive material in our solar system. Furthermore, since carbonaceous chondrites can contain significant quantities of volatile elements, they may have played a crucial role in supplying volatiles and organic material to Earth and other inner solar system bodies. However, a major challenge of unravelling the volatile composition of chondritic meteorites is distinguishing between which features were inherited from the parent body, and what may be a secondary feature attributable to terrestrial weathering. In December 2020, the Hayabusa2 mission of the Japan Aerospace Exploration Agency (JAXA) successfully returned surface material from the C-type asteroid (162173) Ryugu to Earth. This material has now been classified as closely resembling CI-type chondrites, which are the most chemically pristine meteorites.

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Carbonaceous Chondrites
Volatiles
Noble Gases
Nitrogen

The analysis of material from the surface of Ryugu therefore provides a unique opportunity to analyse the volatile composition of material that originated from a CI-type asteroid without the complications arising from terrestrial contamination. Given their highly volatile nature, the noble gas and nitrogen inventories of chondrites are highly sensitive to different alteration processes on the asteroid parent body, and to terrestrial contamination. Here, we investigate the nitrogen and noble gas signature of two pelletized grains collected from the first and second touchdown sites (Okazaki et al., 2022a), to provide an insight into the formation and alteration history of Ryugu. The concentration of trapped noble gas in the Ryugu samples is greater than the average composition of previously measured CI chondrites and are primarily derived from phase Q, although a significant contribution of presolar nanodiamond Xe-HL is noted. The large noble gas concentrations coupled with a significant contribution of presolar nanodiamonds suggests that the Ryugu samples may represent some of the most primitive unprocessed material from the early solar system. In contrast to the noble gases, the abundance of nitrogen and $\delta^{15}\text{N}$ composition of the two Ryugu pellets are lower than the average CI chondrite value. We attribute the lower nitrogen abundances and $\delta^{15}\text{N}$ measured in this study to the preferential loss of a ^{15}N -rich phase from our samples during aqueous alteration on the parent planetesimal. The analyses of other grains returned from Ryugu have shown large variations in nitrogen concentrations and $\delta^{15}\text{N}$ indicating that alteration fluids heterogeneously interacted with material now present on the surface of Ryugu. Finally, the ratio of trapped noble gases to nitrogen is higher than CI chondrites, and is closer to refractory phase Q and nanodiamonds. This indicates that Ryugu experienced aqueous alteration that led to the significant and variable loss of nitrogen, likely from soluble organic matter, without modification of the noble gas budget, which is primarily hosted in insoluble organic matter and presolar diamonds and is therefore more resistant to aqueous alteration.

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1. Introduction

Carbonaceous chondrites represent some of the most primitive materials found within our solar system. They are generally made up of variable abundances of high temperature phases such as chondrules and calcium-aluminium-rich inclusions (CAIs), together with hydrated silicates set within a fine-grained matrix containing abundant organic material (Scott and Krot, 2003). The bulk chemical composition of carbonaceous chondrites (in particular CI chondrites) approximates the composition of the solar photosphere, with the exception of the volatile elements, which are the focus of this study (Palme et al., 2014). Primitive CI chondrites are therefore invaluable samples for studying the chemical makeup of our solar system.

The high abundance of secondary hydrated minerals such as phyllosilicates (>60 vol%) in certain primitive carbonaceous chondrites (CI, CM and CR) is evidence that different parent bodies underwent periods of low temperature (<150 °C) aqueous alteration (Bunch and Chang, 1980). The fluid driving this alteration likely originated as water ice that was melted by heat generated by radioactive decay (principally ^{26}Al) or impact induced heating. The presence of water ice suggests that chondrite parent planetesimals accreted in the outer solar system, beyond the so-called snow line, where temperatures are low enough that water ice, either from the interstellar medium (Vacher et al., 2016) or condensed from the solar nebula (Lodders, 2003), can be preserved. Carbonaceous chondrites can carry a large concentration of volatile elements, with hydrated minerals containing significant concentrations of hydrogen (up to 1.5 wt%; Alexander et al., 2012), and organic material containing abundant carbon and nitrogen. As such, carbonaceous chondrites may have played a crucial role in delivering volatiles and organic material to Earth and the other terrestrial planets (Marty, 2012; Pizzarello and Shock, 2017; Broadley et al., 2020). The analysis of carbonaceous meteorites has therefore been fundamental to our understanding of the composition of the early solar system and how volatile elements were accreted into growing planetesimals.

A major challenge of unravelling the volatile composition and alteration history of meteorites lies in distinguishing between those features inherent to the parent body, and those which may have been induced by terrestrial weathering. As soon as a

meteorite enters Earth's atmosphere, it immediately starts to interact with the water in the atmosphere and on Earth's surface (Bland et al., 2006; Stephant et al., 2018; Vacher et al., 2020), as well as other terrestrial contaminants. CI chondrites in particular are susceptible to terrestrial contamination given their porous nature and high abundance of volatile bearing phases (King et al., 2020). The extent to which terrestrial weathering has modified the volatile composition of carbonaceous chondrites remains largely unknown. The recent JAXA Hayabusa2 mission successfully returned material from the surface of the C-type asteroid (162173) Ryugu (Yada et al., 2022; Tachibana et al., 2022), which is similar in composition to CI chondrites (Yokoyama et al., 2022). This now presents a unique opportunity to analyse material that originated from a carbonaceous asteroid without the potential obfuscation arising from terrestrial contamination.

Given the highly volatile nature of noble gases and nitrogen, and their high abundance on Earth's surface, the noble gas and nitrogen inventory of chondrites is highly sensitive to alteration processes and terrestrial contamination. Primordial noble gases in carbonaceous chondrites are primarily contained within poorly defined phase(s) associated with insoluble organic matter and a variety of different presolar grains, each of which has a unique elemental and isotopic signature (Huss and Lewis, 1994a; Busemann et al., 2000). As such they can provide a wealth of information on the different sources that contributed to the accretion of different planetary bodies. Nitrogen on the other hand is primarily held within soluble and insoluble macromolecular organic matter and can be used to provide an insight into the origin of organic matter accreted to planetary bodies (Alexander et al., 2012). Given that the different carrier phases of noble gases and nitrogen can be more (soluble organic matter) or less (insoluble organic matter and presolar grains) affected by aqueous alteration, the combined analysis of noble gases and nitrogen can provide not only information on the primordial volatile composition of chondrites but also how they may have been modified by different alteration processes. In this contribution we examine the noble gas and nitrogen composition of Ryugu in the context of known primitive carbonaceous chondrite groups, and we discuss potential new insights into different alteration processes occurring on the carbonaceous chondrite parent bodies.

2. Samples and methodology

Two grains were allocated to the Centre de Recherches Pétrographiques et Géo-chimiques (CRPG), Nancy, France as part of the Hayabusa2 initial analysis volatile team (Okazaki et al., 2022a). Grain A0105-05 was sampled during the first touchdown, whilst grain C0106-06 was sampled during the second touchdown following the creation of a crater by an artificial impactor, and thus could be more representative of the subsurface of Ryugu (Arakawa et al., 2020). Prior to their arrival at CRPG the grains were pressed into pellets in order to obtain a flat surface for in-situ petrographic analysis. The force employed in generating the pellets (0.6 N) was not deemed sufficient to alter the volatile composition of the samples as highlighted by the low temperature release of solar wind implanted noble gases in some of the pellets (Okazaki et al., 2022a).

In this study, two pelletized grains were analysed using a multi mass spectrometer approach without the samples ever being in contact with Earth's atmosphere (Okazaki et al., 2022a,b). The pellets were consistently held within a dry nitrogen atmosphere or within a vacuum during sample preparation, transportation, weighing, and transfer to the mass spectrometer for analysis (Okazaki et al., 2022a,b). The samples arrived at CRPG in a stainless steel capsule that was filled with an ultra pure nitrogen atmosphere. This capsule was opened within a nitrogen glove box and the samples were weighed within the glove box, before being transferred to a ZnSe windowed laser chamber for analysis.

Pellet A0105-05 was analysed for He, Ne, Ar, Kr and Xe as well as N isotopes on the Thermo Scientific Helix MC+, whilst pellet C0106-06 was analysed for Ne, Ar and N isotopes on a Nu Instruments Noblesse HR. A concise outline of the methods used to generate the data used in this study has been previously reported by Okazaki et al. (2022a). We have provided a more detailed description of how the noble gases and nitrogen abundances and isotopic compositions were determined at CRPG's noble gas facility, which can be found in the [supplementary material](#).

3. Overview of noble gas and nitrogen results from Ryugu

The noble gas step heating and bulk nitrogen concentrations and isotope ratios of the pelletized grains (18 in total) given to the Hayabusa2-initial-analysis-volatile team (including pellets A0105-05 and C0106-06) have been presented by Okazaki et al. (2022a), in an initial overview of the volatile composition of Ryugu. Here we present a more detailed description of the combined noble gas and nitrogen elemental and isotopic ratios obtained at CRPG from the analysis of pellets A0105-05 and C0106-06, as well as the individual step heating noble gas and nitrogen results ([Supplementary Material](#)) that were not previously presented by Okazaki et al. (2022a).

In summary, the concentrations of primordial noble gases in Ryugu reported by Okazaki et al. (2022a) were found to be very high, with $^{22}\text{Ne}_{\text{trap}}$ - corrected for cosmogenic contributions - ($2.1\text{--}3.9 \times 10^{-12}$ mol/g), ^{36}Ar ($5.1\text{--}6.5 \times 10^{-11}$ mol/g), ^{84}Kr (6.5×10^{-13} mol/g) and ^{130}Xe (1.2×10^{-13} mol/g) concentrations, within the grains analysed in this study, all being in excess of most CM and CI chondrites values (Table S1). This suggests that Ryugu may represent one of the most primitive volatile-rich examples of carbonaceous asteroidal material available for study. The bulk Ne_{trap} , Ar, Kr and Xe isotope signatures of Ryugu are all very similar to phase Q (Figs. 1, 2; Fig. S4; Okazaki et al., 2022a), a carbonaceous phase and the dominant carrier of Ar, Kr and Xe in chondritic meteorites (Busemann et al., 2000). Despite the bulk composition being dominated by phase Q, there is evidence for significant presolar grain contributions in the Ne and Xe isotopes (Figs. 1 and 2). The

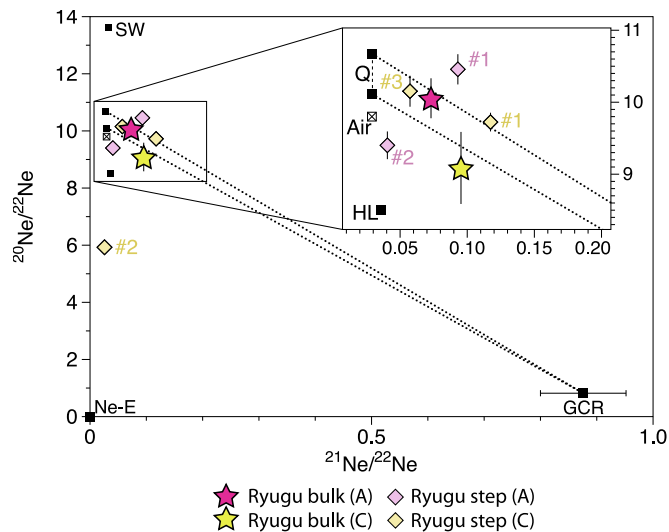


Fig. 1. Neon isotopic composition of the two Ryugu pelletized grains analysed at CRPG in a Ne three isotope plot. The bulk Ne composition of the two samples is consistent with a Q-like trapped component (Busemann et al., 2000) with a contribution of cosmogenic Ne (GCR). Sample A0105-05 (A) shows relatively consistent Ne isotope ratios between the two extractions. The second laser extraction of sample C0106-06 (C) on the other hand shows a significantly different composition than the first and third extraction as well as the bulk value. The low $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ of the second extraction of C0106-06 appears consistent with a contribution of Ne-E, which is primarily contained within presolar graphite, and/or SiC (Lewis et al., 1994, Amari et al., 1995). Uncertainties are to 1 σ .

second laser extraction of sample C0106-06 especially shows a significant contribution from presolar Ne-E component contained in presolar SiC and graphite (Fig. 1). There is limited evidence of a cosmogenic contribution to Ar, Kr and Xe, and so no correction for cosmogenic addition for these elements was necessary.

The noble gas elemental ratios calculated for the pellets analysed at CRPG are shown in Figs. 3, 4 and Table S2. The bulk $^{36}\text{Ar}/^{130}\text{Xe}$ (523 ± 7) and $^{84}\text{Kr}/^{130}\text{Xe}$ (5.1 ± 0.1) of pellet A0105-05 are very similar to gas released from phase Q ($^{36}\text{Ar}/^{130}\text{Xe} = 469 \pm 29$ and $^{84}\text{Kr}/^{130}\text{Xe} = 5.0 \pm 0.3$; Busemann et al., 2000). There is no evidence for the presence of an atmospheric component or implanted solar wind, which is common in CI and certain CM chondrites (Fig. 3). The lack of an atmospheric component highlights the efficient return and handling of the Ryugu samples, avoiding contamination from the terrestrial environment. The $^{36}\text{Ar}/^{22}\text{Ne}_{\text{trap}}$, $^{84}\text{Kr}/^{22}\text{Ne}_{\text{trap}}$ and $^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ of A0105-05 are intermediate between phase Q and HL, a presolar component held within nanodiamonds. This is similar to, although at the lower end of, previously analysed CI chondrites (Fig. 4).

The N concentration measured in pellets A0105-05 and C0106-06 was 885 ± 25 ppm and 858 ± 10 ppm, respectively (Table S3). In contrast to the noble gases, this is lower than the range previously measured in CI chondrites (1600–2300 ppm; Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012), but within the range previously reported for CM chondrites (500–1700 ppm; Alexander et al., 2012). The majority of the nitrogen (>60%) was released from both pellets during the first extraction. However, the $^{36}\text{Ar}/\text{N}$, $^{84}\text{Kr}/\text{N}$ and $^{130}\text{Xe}/\text{N}$ of the samples increased successively as greater laser power settings were used to extract the gas (Fig. 5). Conversely, the $^{22}\text{Ne}_{\text{trap}}/\text{N}$ of all laser extractions and the bulk of pellets A0105-05 and C0106-06 are very consistent (Fig. 5). The ratio of noble gases to nitrogen in Ryugu also appears to be distinct from the majority of previously measured CI and CM chondrites. (Fig. 5). The bulk $^{22}\text{Ne}_{\text{trap}}/\text{N}$ and $^{36}\text{Ar}/\text{N}$ of both A0105-05 and C0106-06 are higher than the range previously determined for CI chondrites and at the higher end of that measured in CM

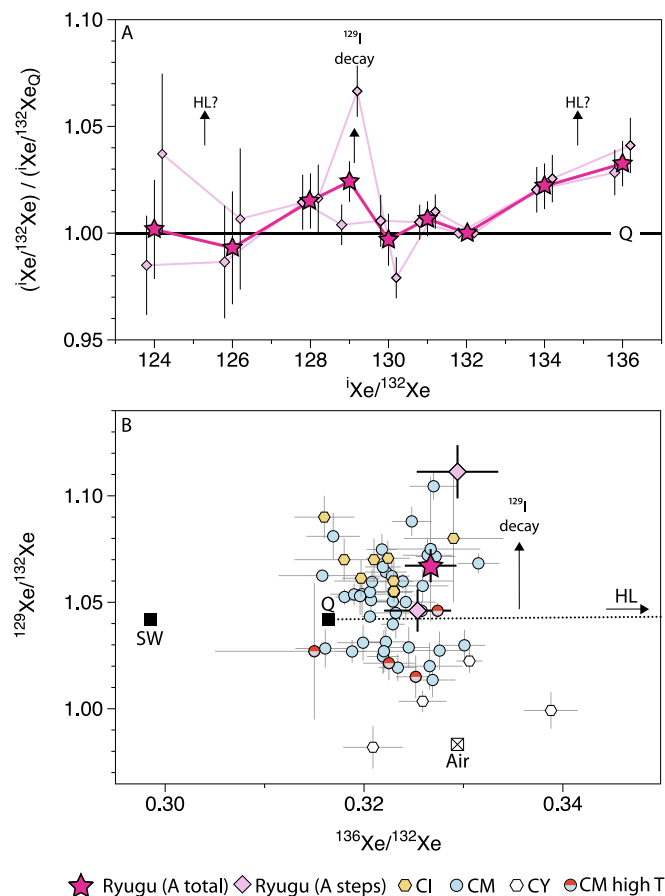


Fig. 2. Xenon isotope spectra of Ryugu normalised to phase Q (A) and a Xe three isotope plot comparing the composition of Ryugu to known endmembers and other carbonaceous chondrites (B). The individual laser extractions (diamonds) and the bulk (star) Xe isotope composition of Ryugu are similar to phase Q (Busemann et al., 2000), with no evidence for an atmospheric contribution. There are notable excesses in $^{129}\text{Xe}/^{132}\text{Xe}$ (first extraction), $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ (both extractions), which are likely due to the decay of extinct ^{129}I and a strong contribution from presolar Xe-HL (as seen in the heavy Xe isotopes), respectively (B). Literature data is from Eugster et al., 1967; Mazor et al., 1970; Bogard et al., 1971; Nagao et al., 1984; Nakamura et al., 1999; Okazaki and Nagao, 2017; King et al., 2019 and Krietsch et al., 2021. None of the samples have been corrected for fission contributions or atmospheric contamination. Uncertainties are presented to 1σ .

chondrites (Fig. 5A). Measured $^{84}\text{Kr}/\text{N}$ and $^{130}\text{Xe}/\text{N}$ of Ryugu appear to be significantly higher than CI and CM (Fig. 5B).

Finally, the bulk $\delta^{15}\text{N}$ composition of pellets A0105-05 and C0106-06 are $+18.14 \pm 0.94\text{‰}$ and $+19.47 \pm 0.89\text{‰}$ (1σ), respectively (Okazaki et al. 2022a), which is once again distinct from bulk CI chondrites (averaging between $+42$ and $+49\text{‰}$; Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012), but similar to bulk CM chondrites (Fig. 6). The $\delta^{15}\text{N}$ shows little variation across the different laser extractions, however the first extraction (low temperature) consistently exhibited the lowest $\delta^{15}\text{N}$ across the two pellets. In the following discussion sections we outline the potential reasons for Ryugu's unique noble gas and nitrogen signature.

4. Discussion

4.1. The noble gas make-up of Ryugu

Noble gases measured in the Ryugu pellets at CRPG represent a mixture of cosmogenic, radiogenic, fissionogenic and trapped components. A cosmogenic contribution is resolvable only for Ne iso-

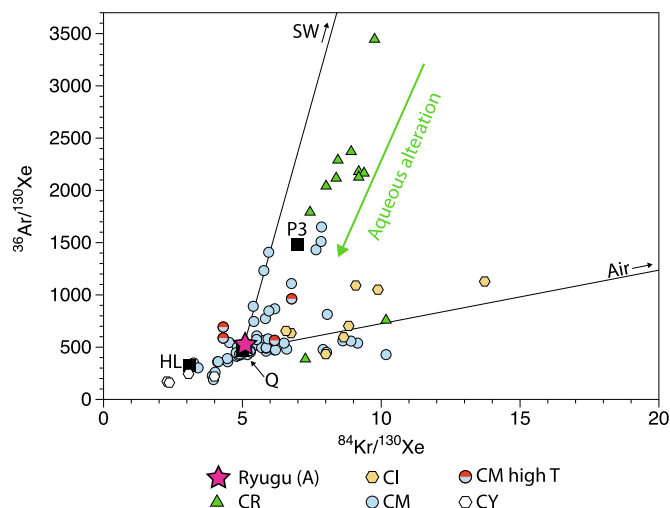


Fig. 3. Argon, krypton and xenon three-element plot of Ryugu and primitive carbonaceous chondrites. The bulk Ryugu composition is very similar to phase Q with no evidence for an atmospheric contribution as is seen in CI chondrites. The most primitive and unaltered CR and CM chondrites have higher $^{36}\text{Ar}/^{130}\text{Xe}$ than Ryugu and CI chondrites, suggesting that they contain an Ar-rich phase that has been lost from the more aqueously altered CI-type chondrites, including Ryugu. CY chondrites, which are considered to represent thermally altered CIs, have $^{36}\text{Ar}/^{130}\text{Xe}$ ratios lower than phase Q suggesting that they may have lost a substantial amount of Ar during thermal alteration. Literature data is from Eugster et al., 1967; Mazor et al., 1970; Bogard et al., 1971; Nagao et al., 1984; Nakamura et al., 1999; Okazaki and Nagao, 2017; King et al., 2019; Krietsch et al., 2021 and Obase et al., 2021. The P3 phase also shown is another presolar component found in nanodiamonds (Huss and Lewis, 1994a; Ott, 2002). Uncertainties are presented to 1σ .

topes, which has been discussed in detail previously by Okazaki et al. (2022a). Here we will primarily concentrate on the trapped, radiogenic and fissionogenic noble gas (Ne, Ar, Kr and Xe) components present within the two CRPG Ryugu pellets.

The bulk Ne composition of both pellets falls along a mixing line between the cosmogenic endmember and phase Q (Fig. 1, Busemann et al., 2000; Wieler et al., 1992; Okazaki et al. 2022a). Whilst neither of the pellets analysed at CRPG show evidence of implanted solar wind, it cannot be fully ruled out that the pellets originally contained a minor implanted solar wind component that could not be resolved from the major release of trapped Ne. The second laser extraction of pellet C0106-06 shows a significantly lower $^{20}\text{Ne}/^{22}\text{Ne}$ (5.93 ± 0.11) than the bulk values, and plots towards the Ne-E endmember (almost or entirely pure ^{22}Ne ; Ott, 2014). This suggests that the Ryugu samples likely contain some presolar SiC and graphite grains, which are the carriers of the Ne-E component (Huss and Lewis, 1994a). However, after correction for the cosmogenic contribution, the bulk $^{20}\text{Ne}/^{22}\text{Ne}$ of sample C0106-06 is still within the range of values of phase Q, suggesting that the contribution of Ne from presolar SiC and graphite in Ryugu is relatively limited.

The heavy noble gas (Ar, Kr and Xe) signature of Ryugu is dominated by the phase Q component (Figs. 2, 3 and Fig. S4). In the case of Ar and Kr isotopes, variations among potential cosmochemical endmembers are much smaller than for Xe, and they will not be discussed further. Xenon isotopes on the other hand show some variations relative to the composition of phase Q (Fig. 2). During the first laser extraction of pellet A0105-05, a significant amount of excess radiogenic ^{129}Xe ($^{129}\text{Xe}^*$), produced from the decay of extinct ^{129}I , was observed. Taking the bulk $^{129}\text{Xe}/^{132}\text{Xe}$ of 1.067 ± 0.008 and assuming a phase Q starting composition, we calculate the amount of excess $^{129}\text{Xe}^*$ to be 1.92×10^{-14} mol/g. If all excess $^{129}\text{Xe}^*$ produced within Ryugu has been retained, and by assuming that Ryugu started with an initial solar system like $^{129}\text{I}/^{127}\text{I}$ ratio

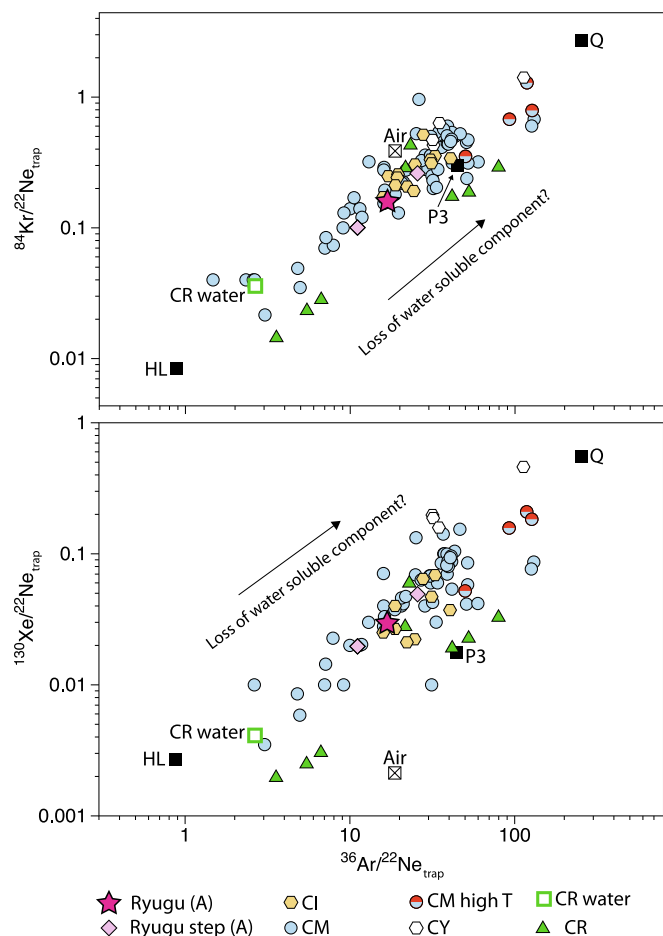


Fig. 4. Noble gas three-element plots of Ryugu and primitive carbonaceous chondrites. All carbonaceous chondrites fall along a trend-line between phase Q and presolar HL. The noble gas phase released from CR chondrites during interaction with water (CR water; Krietsch, 2020) has a similar composition to HL suggesting that this CR water component may be a significant noble gas endmember in carbonaceous chondrites. Ryugu and CI chondrites contain less of this water-soluble component than CM chondrites, consistent with their higher level of aqueous alteration. Compared to CI chondrites Ryugu has lower ^{36}Ar , ^{84}Kr , $^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ suggesting that it may have lost less of the water soluble noble gas phase than similar CI-type chondrites. Finally, thermally altered CY and CM chondrites have higher ^{36}Ar , ^{84}Kr , $^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ ratios that approach phase Q, suggesting that they have lost the majority of the water-soluble noble gas carrier. Literature data is from Eugster et al., 1967; Mazor et al., 1970; Bogard et al., 1971; Nagao et al., 1984; Nakamura et al., 1999; Okazaki and Nagao, 2017; King et al., 2019; Krietsch, 2020; Krietsch et al., 2021 and Obase et al., 2021. Presolar P3 composition taken from Huss and Lewis, 1994a. Uncertainties are 1σ and are smaller than symbol sizes.

of $\sim 10^{-4}$ (Gilmour et al., 2006), we estimate that sample A0105-05 contains around 24 ppb of I. This value is slightly lower than the value previously determined by Clay et al. (2017) for the CI chondrite Orgueil (57 ppb), however it is fully within the range of concentrations for CM chondrites (6–65 ppb), also determined by Clay et al., (2017). One thing to note is that some carbonaceous chondrites (including CR and CV) exhibit $^{129}\text{Xe}^*/\text{I}$ ratios of around 0.3, indicating that they contain uncorrelated I (Gilmour, 2000). If this is also the case for Ryugu then the true concentration of I would be around 3 to 4 times higher than what we calculate here based on the excess $^{129}\text{Xe}^*$. Our estimated I concentrations are therefore likely to be a lower limit; although whether CI chondrites also contain uncorrelated I remains to be seen.

Only one CI chondrite (Orgueil) was measured by Clay et al. (2017), so the range of halogen concentrations in CI chondrites is not well known. Many older studies found I concentrations in CI

chondrites (Orgueil and Ivuna) to be significantly higher than that measured in Ryugu (this study) and by Clay et al. (2017), with values ranging between 230 and 1200 ppb (Reed and Ralph, 1966; Goles et al., 1967). The lower concentration of I within Ryugu, which has never been in contact with the terrestrial atmosphere, compared to some previous studies would seem to confirm that terrestrial contamination can significantly alter the concentration of halogens within meteorites residing on the surface of the Earth. However, the study of Clay et al. (2017) has shown that it is still possible to extract accurate halogen concentrations from meteorites by using a combination of careful sample preparation (avoidance of surface alteration) and new techniques that enable smaller masses of unaltered material to be analysed.

Pellet A0105-05 also presents consistent excesses in $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ relative to Q for both laser extractions (Fig. 2). Excesses in heavy Xe isotopes ($^{134,136}\text{Xe}$) within Ryugu are likely to originate from either the spontaneous fission of ^{238}U and ^{244}Pu , or from the addition of another noble gas component, likely presolar. Yokoyama et al. (2022) determined the U concentration of Ryugu to be 0.009 ppm, which is similar to the average CI chondrite abundance (0.008 ppm; Lodders, 2020). Using the decay constant (λ_{sf}) and the fractional yield of ^{136}Xe ($^{136}\text{Y}_{\text{sf}}$) from Ragettli et al. (1994) and assuming a closure age of around 4.5 Ga, we can estimate the amount of ^{136}Xe produced from the spontaneous fission of ^{238}U . Combining this with the initial solar system $^{244}\text{Pu}/^{238}\text{U}$ and the branching ratio of ^{244}Pu (Fields et al. 1966), we can determine the total contribution of fissionogenic Xe to sample A0105-05. We estimate that fissionogenic Xe contributes around 0.5% of the total ^{136}Xe excess. We consider this an upper limit since a maximum closure age of 4.5 Ga was assumed, although ^{53}Mn - ^{53}Cr ages confirm that Ryugu formed ~ 5 Myr after CAI formation and has not been significantly heated since (Yokoyama et al., 2022). It is therefore clear that the excesses in $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ measured within sample A0105-05 cannot be accounted for by the addition of fissionogenic Xe (Fig. 2) and requires the presence of another Xe component in addition to the predominant phase Q.

The excesses in $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ are therefore likely due to the presence of presolar nanodiamonds in Ryugu (Huss and Lewis, 1994a). Presolar nanodiamonds can contribute significantly to the total noble gas budget of carbonaceous chondrites (Ott, 2002). In particular, the excesses in $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ of Ryugu are likely due to the addition of Xe-HL, which is an endmember component carried within presolar nanodiamonds (Huss and Lewis, 1994a). Xenon-HL shows a near equal enrichment of light ($^{124,126}\text{Xe}$) and heavy ($^{134,136}\text{Xe}$) isotopes compared to the more isotopically “normal” composition of phase Q and solar (Huss and Lewis, 1994a). If Ryugu samples contain Xe-HL-bearing nanodiamonds, then there should be similar, but slightly lower, excesses in $^{124}\text{Xe}/^{132}\text{Xe}$ and $^{126}\text{Xe}/^{132}\text{Xe}$, relative to phase Q, than is seen in $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ respectively. However, the $^{124}\text{Xe}/^{132}\text{Xe}$ and $^{126}\text{Xe}/^{132}\text{Xe}$ of the Ryugu samples are indistinguishable from phase Q, within uncertainty (Fig. 2). The lack of $^{124}\text{Xe}/^{132}\text{Xe}$ and $^{126}\text{Xe}/^{132}\text{Xe}$ excesses could be the result of the lower abundance of the light Xe isotopes, making it difficult to distinguish a clear Xe-HL contribution for $^{124}\text{Xe}/^{132}\text{Xe}$ and $^{126}\text{Xe}/^{132}\text{Xe}$ within the analytical uncertainties (Fig. 2). For example, from the $^{136}\text{Xe}/^{132}\text{Xe}$ excess relative to Q, after correction for fissionogenic contribution, we calculate that Ryugu is best explained as a mixture of 2.7% Xe-HL and 97.3% phase Q. This amount of Xe-HL would equate to a $^{124}\text{Xe}/^{132}\text{Xe}$ of 0.00465, which is close to the uncertainty (1σ) of the measured value (0.00454 ± 0.00009). Alternatively, since Xe-H (heavy) and Xe-L (light) are produced by different nucleosynthetic processes, it may be possible that the Ryugu only contains the Xe-H component (Bekaert et al., 2019). As of yet Xe-H and Xe-L have been consistently found together at

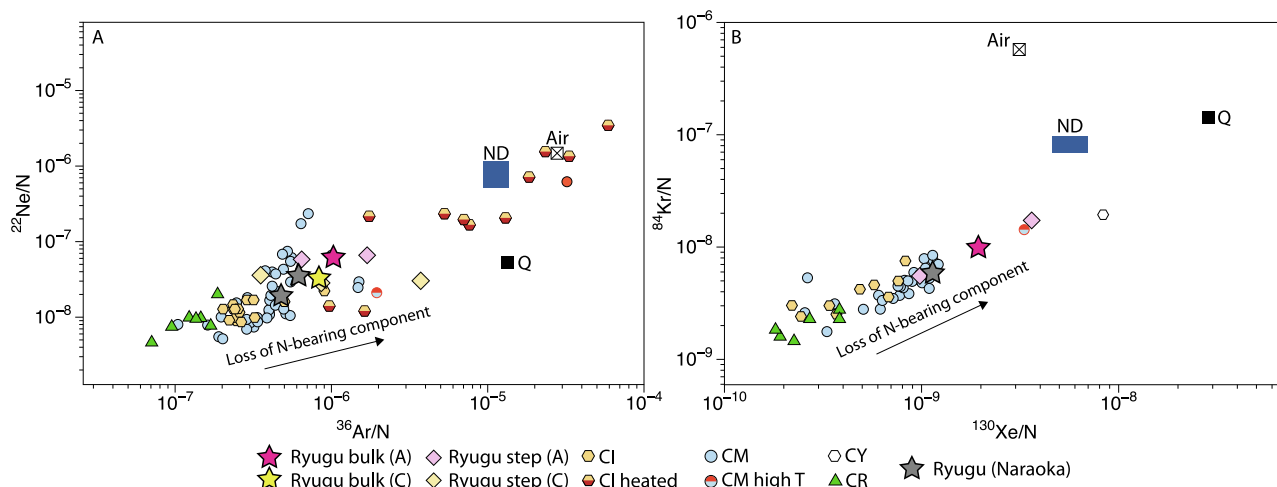


Fig. 5. Three element plots of noble gases and nitrogen in Ryugu and carbonaceous chondrites. The Ryugu samples have elevated noble gas to nitrogen ratios relative to other CI chondrites and closer towards the composition of refractory phase Q (Marty et al., 2010) and nanodiamonds (ND - Russell et al., 1996; Huss and Lewis, 1994b). This is consistent with the Ryugu samples having lost a ^{15}N -rich soluble organic matter component during aqueous alteration. This results in the more refractory phases (phase Q and nanodiamonds) dominating the nitrogen signature of the Ryugu samples. We have also combined our noble gas data with the N data from Naraoka et al. (2022). Even with the higher concentrations of N from Naraoka et al. (2022), Ryugu still appears distinct from the average CI composition, suggesting that it has lost more soluble organic matter during aqueous alteration. A similar loss of organic N-rich material also occurs during heating as shown by thermally altered CY and CM chondrites and lab heating experiments of CIs (Füri et al., 2013). The composition of the nanodiamonds is calculated using data from CI and CM diamond separates (from Russell et al., 1996; Huss and Lewis, 1994b). There are a limited number of CR chondrites that have been analysed for both nitrogen and noble gases; here we show two examples (Al Rais and Renazzo), which have been analysed several times for nitrogen (Pearson et al., 2006) and noble gas abundances (Mazor et al., 1970). Noble gas data for CI and CM chondrites is from Eugster et al., 1967; Mazor et al., 1970; Bogard et al., 1971; Nagao et al., 1984; Nakamura et al., 1999; Okazaki and Nagao, 2017; King et al., 2019; Krietsch et al., 2021 and Obase et al., 2021 with N data coming from Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012; Vacher et al., 2020. Uncertainties are 1σ and are smaller than symbol sizes.

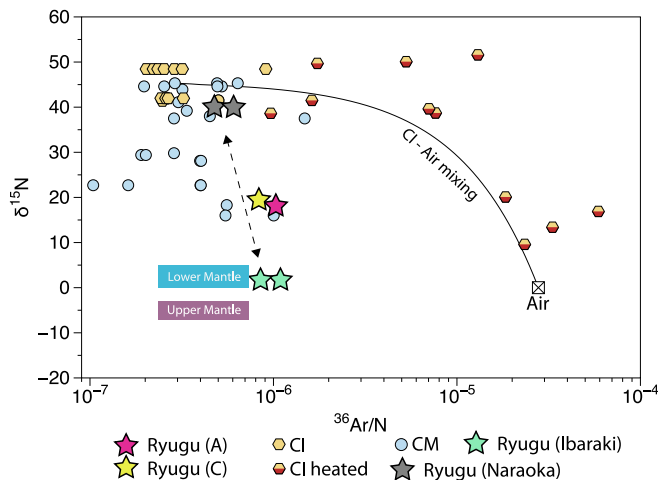


Fig. 6. N isotope vs $^{36}\text{Ar}/\text{N}$ ratio of Ryugu and other carbonaceous chondrites. The $\delta^{15}\text{N}$ and $^{36}\text{Ar}/\text{N}$ of the two Ryugu pelletized grains analysed in this study are lower and higher than that of previously analysed CI chondrites respectively. Combining the N abundance and $\delta^{15}\text{N}$ measured by Naraoka et al. (2022), as presented in Okazaki et al., 2022a, with the noble gases measured in this study results in Ryugu (grey star) being more consistent with CI chondrites. Conversely, if the ^{36}Ar concentrations measured in this study are combined with the N abundances and $\delta^{15}\text{N}$ measured at Ibaraki University (Okazaki et al., 2022a), then the resulting composition of Ryugu (green star) is even more distinct from CIs than the grains analysed in this study. Air contamination and thermal processing of the two samples can be ruled out since they do not fall along the mixing line between CIs and the atmosphere, or fit the data from CIs heated in the laboratory (Füri et al., 2013). The nitrogen composition of the Ryugu pellets analysed at CRPG appears to indicate loss of a ^{15}N -rich component during aqueous alteration. Noble gas literature data is from Eugster et al., 1967; Mazor et al., 1970; Bogard et al., 1971; Nagao et al., 1984; Nakamura et al., 1999; Okazaki and Nagao, 2017; King et al., 2019 and Krietsch et al., 2021, with N data coming from Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012. Uncertainties are 1σ and are smaller than symbol sizes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

a constant abundance ratio in nanodiamonds, so whether Xe-H can be added to chondrites within nanodiamonds without the associated Xe-L remains to be proven. Future analyses of larger samples masses of Ryugu may be able to better distinguish a Xe-HL component for the lighter Xe isotopes.

Presolar noble gases held within nanodiamonds are relatively resistant to aqueous alteration and only significantly affected by thermal alteration (Huss et al., 2003). Of all the different carbonaceous chondrites, CIs and CMs are noted as having a high abundance of presolar noble gas components, since they are the least affected by thermal metamorphism (Huss et al., 2003). The amount of Xe-HL within the bulk sample A0105-05 (2.7%) is higher than nearly all previously measured CI chondrites (Fig. 2). Since the majority of CI chondrites have not been significantly thermally metamorphosed, and nanodiamonds are resistant to aqueous alteration, the variation in Xe-HL contribution between different CI and CM chondrites may suggest that presolar grains were heterogeneously distributed throughout the carbonaceous chondrite-forming region, and likely the entire solar system (Floss and Haenecour 2016; Bekaert et al., 2019). This spatial heterogeneity could directly reflect the variable inheritance of presolar grains into the disk from the interstellar medium (Dauphas et al., 2002), or more likely represent the differential preservation of the grains during thermal processing within the early disk (Huss et al., 2003; Burkhardt et al., 2012).

Observations of Ryugu's rubble pile structure and high abundance of organic matter have been taken as evidence that it may have started life as a comet, before losing its water and CO_2 ice components due to sublimation (Miura et al., 2022). The high abundance of presolar noble gases measured in this Ryugu, suggesting a cold unprocessed outer solar system origin for Ryugu, could therefore further attest to this possibility. However, the phase Q dominated Xe isotope signature of Ryugu is at odds with that previously measured within the coma of comet 67P/Churyumov-Gerasimenko, which exhibited significant deficits in the heavy Xe isotopes relative to solar and phase Q (Marty et al.,

2017). This difference in Xe isotopes suggests that a genetic link between Ryugu and comets may not be well founded. It should be noted however, that the Xe isotopes measured in comet 67P/Churyumov–Gerasimenko, were from the sublimating ice phases and therefore it cannot be completely ruled out that the solid silicate and organic material within comets have chondritic-like noble gas signatures. Measurements of Ne isotopes in material returned from comet 81P/Wild 2 by the NASA Stardust mission were indistinguishable from phase Q (Marty et al., 2008). Based on the noble gas evidence alone, it cannot therefore be concluded whether Ryugu started life as a comet, given that the organic phase Q likely dominates the heavy noble gas composition of the non-ice components of both carbonaceous asteroids and comets. Future analyses of the alteration products formed during interactions with water, may provide information on the noble gas composition of the original ice phase and at the same time uncover any genetic link between Ryugu and comets.

4.1.1. Noble gas evidence for limited thermal alteration on Ryugu

Initial analyses of the materials returned from the surface of Ryugu indicate that it is dominated by hydrous material similar to that found in CI chondrites (Pilorget et al., 2022; Yada et al., 2022; Yokoyama et al., 2022). The release profile of the H₂O suggests that the Ryugu samples have not been heated above ~100 °C since the last period of aqueous alteration took place on the parent planetesimal prior to its break up and re-accumulation in to the current Ryugu asteroid (Yokoyama et al., 2022). However, based on the visible and near-infrared (VNIR) reflectance spectra taken during the Hayabusa2 mission, it had been suggested that the surface of Ryugu may share certain similarities with thermally metamorphosed carbonaceous chondrites (Kitazato et al., 2019; Sugita et al., 2019), such as CY chondrites. These chondrites share similar mineralogical features and elemental compositions with the phyllosilicate-rich CI and CM chondrites (King et al., 2019), but unlike the CI and CM chondrites, CYs experienced peak metamorphic temperatures in excess of 500 °C (King et al., 2019). The similarity in VNIR reflectance spectra between Ryugu and CY chondrites could suggest that Ryugu experienced thermal metamorphism as a result of internal heating from the decay of ²⁶Al or impact induced heating (Sugita et al., 2019). Alternatively, the very surface of Ryugu experienced dehydration caused by micrometeoroid bombardment (Noguchi et al., 2022).

Field Emission Scanning Electron Microscope (FESEM) observations revealed that our Ryugu samples do not contain CY chondrite-like clasts (Supplementary Material), indicating that at least our samples have not been significantly thermally altered. Furthermore, the concentrations of trapped noble gases in CY chondrites are markedly lower than those of the Ryugu samples, sometimes by more than an order of magnitude (King et al., 2019). CY chondrites measured for their noble gas abundance also show a negative correlation between peak metamorphic temperature and trapped noble gas abundances, suggesting that thermal metamorphism played an important role in controlling the trapped noble gas budget of chondrites (King et al., 2019; Krietsch et al., 2021). The high concentration of trapped noble gases within the Ryugu pellets therefore suggests that the Ryugu parent body has not been subjected to significant thermal metamorphism and noble gas loss.

The bulk and individual step laser releases from pellet A0105-05 have ³⁶Ar/¹³⁰Xe, ⁸⁴Kr/¹³⁰Xe and Xe isotopic compositions dominated by phase Q (Figs. 2 and 3). The bulk ³⁶Ar/¹³⁰Xe (529 ± 7) and ⁸⁴Kr/¹³⁰Xe (5.1 ± 0.1) of Ryugu show no evidence for the addition of the atmospheric component (³⁶Ar/¹³⁰Xe = 9602 and ⁸⁴Kr/¹³⁰Xe = 183.5) that is present in several CI chondrites (Fig. 3), indicating that the Ryugu pellets were successfully analysed without being modified by interactions with the terrestrial atmosphere.

Interestingly, the Xe isotope compositions of CI chondrites show no evidence for the addition of atmosphere, despite clear evidence of air contribution from the ³⁶Ar/¹³⁰Xe and ⁸⁴Kr/¹³⁰Xe (Fig. 3). Assuming that CI chondrites contain only phase Q and atmosphere, then using the ³⁶Ar/¹³⁰Xe we calculate that the most air contaminated CI chondrite (³⁶Ar/¹³⁰Xe = 1128; Fig. 3) contains only 5% atmosphere. This addition of 5% atmosphere would not significantly change the Xe isotopic composition of the CI chondrites, and would be difficult to distinguish from pure phase Q within analytical uncertainties. Although the CY and heated CM chondrites do not show significant air contamination from their ³⁶Ar/¹³⁰Xe and ⁸⁴Kr/¹³⁰Xe ratios, the Xe isotopes of the CY and heated CM chondrites have a significant atmospheric contribution, suggesting that the isotope and elemental ratios may be decoupled in thermally metamorphosed carbonaceous chondrites (Figs. 2 & 3).

The lower than phase Q ³⁶Ar/¹³⁰Xe and ⁸⁴Kr/¹³⁰Xe ratio of CY chondrites are more similar to the presolar HL component (Fig. 3; King et al., 2019). This could suggest that the HL phase contained within presolar diamonds is more resistant to gas loss during thermal metamorphism. However, based on step heating experiments, the major release of the heavy noble gases from phase Q and HL occurs at similar temperatures (>1000 °C; Huss and Lewis, 1994a; Huss et al., 1996). It is therefore more likely that CY chondrites have preferentially lost lighter Ar and Kr compared to Xe due to diffusion, lowering the ³⁶Ar/¹³⁰Xe (Fig. 3). Overall, the noble gas composition of Ryugu is distinct from thermally metamorphosed chondrites such as CY and heated CMs, indicating the Ryugu has not undergone significant thermal metamorphism, at least not at temperatures significantly above 500 °C (Krietsch et al., 2021). Furthermore, step heating of some of the Ryugu pellets shows that a significant amount of solar wind and cosmogenic Ne was released at temperatures lower than 200 °C, indicating that Ryugu samples are unlikely to have been heated to temperatures significantly above 200 °C in the last few million years (Okazaki et al., 2022a). This contrasts with reflectance spectra of material excavated from below the surface of Ryugu, which suggest the sub-surface of Ryugu was heated to temperatures in excess of 300 °C (Kitazato et al., 2021). Although it should be noted that the maximum temperature experienced by Ryugu may have occurred on Ryugu's parent body and therefore the noble gases may record the thermal history of Ryugu after its formation.

In conclusion, noble gases measurements suggest that Ryugu samples returned by the Hayabusa2 mission have been largely unaffected by thermal processes and therefore provide a unique sample suite to study the volatile composition of early solar system materials.

4.1.2. Potential effects of aqueous alteration on the noble gas composition of Ryugu

Aqueous alteration of carbonaceous chondrites can have a strong effect on noble gas abundances and elemental, and to a lesser extent isotopic, compositions (Browning et al., 1996; Bekaert et al., 2019; Krietsch et al., 2021). Abundances of Ar, Kr and Xe in CM chondrites are correlated with the petrologic type of the meteorite, with the samples exhibiting the lowest degree of aqueous alteration containing the highest concentration of Ar, Kr and Xe (Browning et al., 1996; Bekaert et al., 2019; Krietsch et al., 2021). The high abundance of hydrated phases, limited number of anhydrous silicates and the photosphere-like elemental abundance pattern of Ryugu suggests that it is composed of material similar to CI chondrites, which have undergone a high degree of aqueous alteration (Pilorget et al., 2022; Yokoyama et al., 2022). As previously stated however, the abundance of Ar, Kr and Xe measured in the Ryugu samples are higher than previously measured in CI chondrites (Mazor et al., 1970; King et al., 2019; Okazaki et al.,

2022a). The high concentrations of Ar, Kr and Xe measured in Ryugu would therefore suggest that it has been less affected by aqueous alteration-driven noble gas loss than other CIs (Table S1).

The apparently greater loss of noble gases in other CI chondrites, in comparison to Ryugu, could have occurred on their respective parent bodies, or it may have occurred within CI chondrites during their interactions with the terrestrial environment. Several mineralogical differences are observed between the Ryugu samples and CI chondrites that suggest that they may have experienced distinct alteration conditions. For example, the high abundance of sulfides and lack of sulfates and ferrihydrite in Ryugu compared with CIs suggests that Ryugu experienced alteration under more reducing conditions (Yokoyama et al., 2022). However, it has not yet been established whether the more oxidising alteration conditions observed in CI chondrites are the result of differing alteration conditions on the parent bodies, or from the alteration of CIs within the terrestrial environment. Although the latter would be more probable considering sulfate formation in the terrestrial environments (Gounelle and Zolensky, 2001), as well as the evidence for the rapid growth of sulfates on polished sections of Ryugu grains that have been exposed to the Earth's atmosphere (Nakamura et al., 2022). Regardless of where this alteration took place, the noble gas concentration of the Ryugu samples again confirms that it represents one of the most primitive examples of CI-like material.

Noble gases are hosted in a variety of different chemical phases that have variable solubilities and can therefore exhibit more or less resistance to aqueous alteration. For example, the majority of heavy noble gases in carbonaceous chondrites are hosted in phase Q, an insoluble organic matter phase that is relatively resistant to aqueous alteration (Yamamoto et al., 2006). However, the process of aqueous alteration appears to result in the preferential loss of Ar relative to Kr and Xe (Krietsch et al., 2021), therefore suggesting that there is another Ar-rich phase that is susceptible to aqueous alteration. Carbonaceous chondrites that have experienced the lowest degrees of aqueous alteration are likely to have higher $^{36}\text{Ar}/^{130}\text{Xe}$ ratios. Primitive CR chondrites, which are generally less affected by aqueous alteration than CI and CM chondrites, have some of the highest $^{36}\text{Ar}/^{130}\text{Xe}$ ratios measured amongst the primitive carbonaceous chondrites (>2000; Fig. 3; Obase et al., 2021). In contrast, the more aqueously altered CR chondrites, together with the majority of CI and CMs, have lower $^{36}\text{Ar}/^{130}\text{Xe}$ ratios (Fig. 3), suggesting that an Ar-rich component has been lost from the chondritic parent bodies during aqueous alteration (Krietsch et al., 2021). The $^{36}\text{Ar}/^{130}\text{Xe}$ of Ryugu sample A0105-05 (529 ± 7) is very similar to phase Q (469 ± 29 ; Busemann et al., 2000), suggesting that, as is the case with other CI chondrites, Ryugu does not contain the Ar-rich component found in primitive CR and CM chondrites (Obase et al., 2021).

The carrier of the Ar-rich component that is lost during aqueous alteration is not well known. Alteration experiments on primitive CR chondrites show that during hydrothermal interaction, a large quantity of He and Ne is released from within the meteorite but surprisingly little Ar (Fig. 3; Krietsch, 2020; Obase et al., 2021). In contrast, during interaction with acetic acid it has been shown that primitive CR chondrites release a considerable concentration of Ar (approaching 60% of the bulk; Krietsch, 2020). The $^{36}\text{Ar}/^{130}\text{Xe}$ of the gas released during interaction with acetic acid can be >3000, suggesting that the Ar-rich phase lost during aqueous alteration is soluble in weak acid solutions (Krietsch, 2020). Interestingly, the $^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ ratios of CI, CM and CR chondrites, as well as the Ryugu samples, fall intermediate between the water-soluble phase found in CR chondrites (Krietsch, 2020) and phase Q (Busemann et al., 2000), with heated CM and CY chondrites being the most similar to phase Q (Fig. 4). This could potentially indicate that all primitive carbonaceous chondrites originally contained a

water-soluble noble gas carrier, which has been subsequently lost from certain meteorites during aqueous alteration, and potentially to an even further degree during extensive heating (see heated CM and CY in Fig. 4). This is substantiated by the fact that the $^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ measured during the multiple laser extractions of the Ryugu pellets increased as successively more laser power was applied to the sample (Fig. 4). Noble gases contained within the water-soluble phase therefore appear to be released at lower temperatures than the heavy noble gas bearing phase Q. The bulk $^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ of Ryugu is similar to other CI chondrites but falls slightly closer to the CR water component (the composition of gas released from CR chondrites during hydrothermal experiments in the laboratory, Krietsch, 2020) than the average CI chondrite composition, suggesting that Ryugu may have lost less of this water-soluble noble gas component than other CI chondrites. Together with the high noble gas abundances measured in the Ryugu pellets, this could suggest that noble gas composition of Ryugu has been less significantly affected by aqueous alteration than other CI-type material.

The Ryugu samples, as well as the majority of CI chondrites, show little evidence for a high Ar/Xe component found in the most primitive unaltered CR and CM chondrites (Krietsch, 2020; Krietsch et al., 2021; Obase et al., 2021). It therefore remains to be seen whether this Ar-rich phase was originally present in all carbonaceous chondrites and was lost from the severely aqueously altered CI chondrites and Ryugu, or whether this phase was never accreted to the CI parent bodies in the first place. The potential identification of a similar Ar-rich phase in some ordinary chondrites, which formed in a distinct reservoir from the carbonaceous chondrites (Warren, 2011), suggests that the former scenario is the most likely (Obase et al., 2021). Interestingly, several studies have identified fragments of anhydrous olivine and pyroxene within different Ryugu grains (Liu et al., 2022; Nakamura et al., 2022). The high Mg content of the grains suggests that they have experienced limited interaction with water (Liu et al., 2022; Kawasaki et al., 2022). Furthermore, in situ measurements of the olivine grains show that they have similar oxygen isotope compositions to amoeboid olivine aggregates and chondrule phenocrysts, suggesting that chondrule precursors and chondrules were accreted to the Ryugu parent body. The preservation of anhydrous minerals in some of the Ryugu grains indicates that sections of the Ryugu protolith have likely experienced less extensive aqueous alteration. Future targeted analysis of material containing anhydrous phases may reveal whether these phases have retained this Ar-rich component.

4.2. Nitrogen in Ryugu - close but no CI: evidence for the heterogeneous alteration of nitrogen bearing components on the surface of Ryugu

The abundance and isotopic composition of nitrogen within the Ryugu pellets measured at CRPG are significantly different from those previously measured in CI chondrites (Figs. 5 & 6; Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012). Bulk nitrogen abundances and isotopes have also been measured in other Ryugu samples by other laboratories (Okazaki et al., 2022a; Naraoka et al., 2022). The nitrogen abundances and $\delta^{15}\text{N}$ signature of the samples range from 700 to 1500 ppm and 0‰ to +43‰ respectively (Okazaki et al., 2022a; Naraoka et al., 2022), with the pellets analysed at CRPG intermediate between these extreme values. We also note that when the different laboratories measured N within multiple Ryugu samples, they found little internal variation (Okazaki et al., 2022a; Naraoka et al., 2022), which is surprising given that some of the laboratories analysed samples from both touchdown sites. This could suggest there is some preparation/analytical issue, which results in similar nitrogen abundances and $\delta^{15}\text{N}$ being measured across several samples. Terrestrial contamination of the

Ryugu samples can likely be ruled out, given that this would simultaneously raise the N concentration (abundant in atmosphere) and lower the $\delta^{15}\text{N}$ value towards more atmospheric-like values ($\delta^{15}\text{N} = 0\text{‰}$), which is not the case (Fig. 6). In fact Okazaki et al. (2022a) showed that all the Ryugu samples fall along a linear correlation on a plot of $\delta^{15}\text{N}$ vs $1/\text{N}$ suggesting that N in Ryugu can be best accounted for by a mix between a nitrogen-rich high $\delta^{15}\text{N}$ component and a nitrogen-poor low $\delta^{15}\text{N}$ component. Furthermore, at CRPG the two pellets were analysed using two different mass spectrometers and purification lines (Supplementary Material), so a consistent analytical issue across different analytical setups would seem unlikely. Finally, the pellets analysed at CRPG and Ibaraki University ($\delta^{15}\text{N} \approx 0\text{‰}$) were prepared using the same protocol so the chances that the preparation of the samples could account for the interlaboratory variation seen for N abundances and isotope compositions seem remote. We therefore consider the differences in abundance and isotope signature in the Ryugu samples measured across the different laboratories to be representative of the variable nitrogen composition of the surface material of Ryugu.

In terms of the samples measured at CRPG, the difference in nitrogen composition compared to CI chondrites is likely due to: (i) an inherent difference in the N-bearing components accreted to the respective Ryugu and CI parent bodies, (ii) some process(es) occurring on the parent bodies that modified the original constant N signature, or (iii) a combination of the two. In terms of different accretionary histories, a higher contribution of presolar nanodiamonds with a $\delta^{15}\text{N}$ of $\sim -350\text{‰}$ (Russell et al., 1996) to Ryugu could account for its lower $\delta^{15}\text{N}$. The noble gas to nitrogen ratios shown in Fig. 5, indicate that the Ryugu pellets contain a higher contribution of nitrogen and noble gas from presolar nanodiamonds than the majority of primitive (unheated) CI and CM chondrites. Taken together with the high Xe-HL contribution measured in the Ryugu samples, this suggests that Ryugu contains a higher proportion of presolar N than other CI and CM chondrites. However, it has been suggested that all carbonaceous chondrites likely originated with a relatively constant ratio of presolar nanodiamonds to organic macromolecular material (Alexander et al., 1998). Variations in the ratio of nanodiamond to organic N in carbonaceous chondrites are therefore normally attributed instead to the preferential loss of organic matter from chondrite parent bodies during aqueous and thermal alteration (Alexander et al., 1998). Furthermore, nanodiamonds are a minor N component ($\sim 10\%$ of the total N), so to lower the $\delta^{15}\text{N}$ of CI chondrites towards the values measured in the Ryugu pellets would require Ryugu to have accreted a substantially greater amount of nanodiamonds than other carbonaceous chondrites. Whilst a higher contribution of presolar N to Ryugu cannot be ruled out, it appears unlikely that variable contributions of presolar material can explain the difference between Ryugu and CI chondrites (Alexander et al., 1998).

The higher noble gas to nitrogen ratio of Ryugu is closer to those of nanodiamonds and phase Q than the majority of CI and CM chondrites (Fig. 5). Since nanodiamonds and phase Q are both resistant to aqueous and low temperature alteration, this suggests that the composition of the Ryugu samples may have been modified towards a more refractory N composition by the loss of some other phase rich in ^{15}N (Sephton et al., 2003). The $^{36}\text{Ar}/\text{N}$, $^{84}\text{Kr}/\text{N}$ and $^{130}\text{Xe}/\text{N}$ also successively increase towards the value of nanodiamonds and phase Q as the laser power was increased with each gas extraction. This suggests that there are at least two nitrogen and noble gas bearing phases in Ryugu, one enriched in nitrogen, which is released at low temperatures, and one which is enriched in Ar, Kr and Xe likely contained in nanodiamonds and insoluble organic material (phase Q) that is released at higher temperatures (Fig. 5). Similar noble gas to nitrogen ratios (Fig. 5) are seen for Ryugu and the thermally altered CY and heated CM chon-

drites, suggesting that they have all lost this ^{15}N -rich organic phase to varying degrees. In the case of the CY and heated CM chondrites this may have occurred during thermal alteration, as is highlighted by the evolution of the nitrogen and noble gas composition of CI chondrites during heating in the laboratory (Figs. 5 & 6; Füre et al., 2013). However, in the case of Ryugu there is little evidence for thermal alteration (Okazaki et al., 2022a; Yokoyama et al., 2022) suggesting that the ^{15}N -rich phase was primarily lost during aqueous alteration.

There is a general relationship between the degree of hydration (aqueous alteration state) and $\delta^{15}\text{N}$ composition of carbonaceous chondrites, with the most aqueously altered CI and CM chondrites having lower $\delta^{15}\text{N}$ signatures (Alexander et al., 2013). This suggests that aqueous alteration on the parent body can play an important role in controlling the nitrogen composition of carbonaceous chondrites (Sephton et al., 2003; Alexander et al., 2013; van Kooten et al., 2018). Laboratory experiments, which replicate aqueous alteration conditions on carbonaceous chondrite parent bodies, have shown that free organic matter (FOM) and labile organic matter (LOM) are preferentially lost from chondritic macromolecular material (Sephton et al., 2003). The free and labile organic matter in these carbonaceous chondrites has $\delta^{15}\text{N}$ compositions of $\sim +88\text{‰}$ and $\leq +85\text{‰}$, respectively (Sephton et al., 2003), therefore the preferential loss or remobilisation of these components could lower $\delta^{15}\text{N}$ towards the values measured in this study. Soluble organic matter is a significant carrier of N in carbonaceous chondrites (Sephton et al., 2003), the loss of this component from Ryugu samples could also account for its lower N concentration relative to other CI chondrites. Since soluble organic matter phases are not a major source of noble gases in carbonaceous chondrites (Ott, 2002), the loss of this component from Ryugu would have a limited impact on the noble gas budget.

CR chondrites, which contain water soluble Ne and Ar rich phase(s) (Fig. 3), have some of the highest $\delta^{15}\text{N}$ values measured for primitive carbonaceous chondrites suggesting that they may also contain more ^{15}N -rich organic matter (Alexander et al., 2013). This appears to be confirmed by the lower noble gas to nitrogen ratio of CR chondrites in comparison to other carbonaceous chondrites (Fig. 5), even though the number of CR chondrites measured for both nitrogen and noble gases is rather limited. Interestingly, although the $^{36}\text{Ar}/\text{N}$, $^{84}\text{Kr}/\text{N}$ and $^{130}\text{Xe}/\text{N}$ of the Ryugu samples increased with increasing temperature of extraction, the $^{22}\text{Ne}_{\text{trap}}/\text{N}$ ratios are consistent across all temperature extractions (Fig. 5). This suggests that N and trapped Ne may be hosted together in the same water-soluble organic phase. However, in contrast to the lower N concentration and $\delta^{15}\text{N}$ of Ryugu compared to CI chondrites, the ^{36}Ar , ^{84}Kr , $^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ measured in Ryugu and CI chondrites are similar, and therefore even if the N and trapped Ne are currently contained together within a similar low temperature phase, the preferential loss of this phase cannot solely account for the N and noble gas signature of the Ryugu samples analysed at CRPG. The loss of another ^{15}N -rich component is therefore required to fully account for the nitrogen and noble gas composition of Ryugu.

If our conclusion is correct that the nitrogen composition of Ryugu is primarily controlled by aqueous alteration, then the large range of nitrogen concentrations and $\delta^{15}\text{N}$ measured in the different Ryugu samples across different laboratories likely suggests that aqueous fluids were heterogeneously distributed throughout the surface of Ryugu. Evidence for small-scale variations in aqueous alteration has been reinforced by the discovery of (i) grains rich in anhydrous silicate material, which are considered to have experienced a lesser degree of aqueous alteration (Nakamura et al., 2022; Liu et al., 2022), (ii) grains containing magnetite-apatite veins, which provide direct evidence for the presence of aqueous alteration fluids on the near-surface of Ryugu (Yokoyama et al.,

2022; Bazi et al., 2022), (iii) the heterogeneous distribution of organic molecules across different Ryugu grains (Ito et al., 2022), and (iv) large variations (factor of 2) in Ca abundances between different Ryugu grains from the dissolution of primary Ca-bearing phases during aqueous alteration (Moynier et al., 2022). The identification of lithologies that have undergone variable degrees of aqueous alteration may therefore account for the variation in N isotopes analysed across the different laboratories. Indeed, when the nitrogen isotopic composition of the bulk Ryugu matrix is investigated at a fine scale using NanoSIMS, it shows a large variation (+28 to +556‰; Ito et al., 2022), highlighting the potential for differential preservation of different nitrogen bearing components.

Variations in aqueous alteration may also explain why the samples analysed at CRPG and Ibaraki University ($\delta^{15}\text{N} \approx 0\text{‰}$) show a larger difference from the average CI chondrite composition compared to that analysed by Naraoka et al. (2022) (+43.0 ± 9.0‰). This is because the pellets analysed at CRPG and Ibaraki University were an order of magnitude smaller than those analysed by Naraoka et al. (2022) (~1 mg) and are therefore more likely to have captured some of the small-scale variations that exist within the surface of Ryugu. In contrast, the larger mass of samples analysed by Naraoka et al. (2022) may be more representative of the bulk N concentration and $\delta^{15}\text{N}$ of Ryugu, which is closer to previously analysed CI chondrites. However, even if we take the N abundance and $\delta^{15}\text{N}$ of Naraoka et al. (2022) and combine it with the noble gas data from CRPG, the $^{22}\text{Ne}_{\text{trap}}, ^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/\text{N}$ ratios of Ryugu (Fig. 5) still remain higher than the average CI chondrite values and closer to the refractory nitrogen components (nanodiamonds and refractory organic matter). Furthermore, the trapped noble gas concentrations measured at CRPG are towards the lower end of concentrations measured in Ryugu pellets (Okazaki et al., 2022a), so there remains potential for the bulk $^{22}\text{Ne}_{\text{trap}}, ^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/\text{N}$ of Ryugu to be even higher. This once again suggests that the Ryugu samples analysed at CRPG, and potentially all Ryugu samples, have lost more ^{15}N -rich soluble phases than other CI chondrites, or simply never contained them in the first place. A higher degree of N loss driven by aqueous alteration would be consistent with the higher C/N ratio of the Ryugu pellets analysed at Ibaraki University (96–117; Okazaki et al., 2022a), compared to CI chondrites (Kung and Clayton, 1978; Kerridge, 1985; Pearson et al., 2006; Alexander et al., 2012), as N is lost preferentially to C during aqueous alteration (van Kooten et al., 2018). Certain Ryugu grains therefore appear to have undergone a higher degree of aqueous alteration driven loss of FOM and LOM, than the parent bodies of CI chondrites, or accreted a slightly different mix of N-bearing components (higher refractory to soluble organic ratio). The heterogeneous degree of aqueous alteration currently found on the surface of Ryugu may therefore be the result of mixing between altered and less altered clasts during the catastrophic break-up of the original parent body and accumulation to form Ryugu.

5. Conclusion

In this work we present a comprehensive overview of the noble gas and nitrogen composition of two pelletized grains returned from the surface of the asteroid Ryugu. We find that the trapped noble gas concentrations of the Ryugu samples are amongst some of the highest measured in carbonaceous chondrite-like material. The trapped noble gases are predominantly derived from the phase Q endmember, yet there is also evidence for a considerable contribution of Xe-HL from presolar nanodiamonds. The high concentration of trapped noble gases and presolar nanodiamonds in comparison to other volatile-rich CI chondrites suggests that

Ryugu represents one of the least processed evidence of carbonaceous asteroidal material. These new data from Ryugu may have important implications for the amount of carbonaceous chondrite-like material that is required to account for the volatile budget of Earth and the other terrestrial planets.

The Ar/Xe and Kr/Xe of Ryugu are similar to phase Q, with no evidence of the atmospheric contamination which is often observed in CI chondrites. The Q-like Ar/Xe and Kr/Xe of Ryugu further indicates the absence of any Ar-rich phase such as those which have been found in the least aqueously altered CR and CM chondrites. This suggests either that Ryugu has lost this Ar-rich phase during periods of aqueous alteration, or simply that this phase was never accreted to Ryugu. The Ryugu samples also exhibit $^{36}\text{Ar}, ^{84}\text{Kr}, ^{130}\text{Xe}/^{22}\text{Ne}_{\text{trap}}$ ratios lower than most CI chondrites, again suggesting that it contains proportionally more of a Ne-rich water soluble phase than CI chondrites. This difference may be ascribed to different levels of aqueous alteration on the respective parent bodies, or on the surface of the Earth in the case of CI chondrites.

For nitrogen, the Ryugu samples analysed at CRPG show lower values than typical CI chondrites for both their N abundance and $\delta^{15}\text{N}$. Noble gas to nitrogen ratios indicates that the Ryugu samples contain a relatively higher contribution of refractory nitrogen bearing components (IOM and nanodiamonds) than other CI chondrites. The larger contribution of nitrogen-bearing refractory phases in Ryugu, coupled with the lower nitrogen abundance and $\delta^{15}\text{N}$ value, suggest that Ryugu has lost a ^{15}N -rich organic phase, potentially during aqueous alteration on Ryugu itself. Interestingly, comparisons with other nitrogen measurements of Ryugu conducted at other laboratories show a large degree of heterogeneity, suggesting that individual grains on the surface of Ryugu have been altered by aqueous fluids to variable degrees.

Higher noble gas abundance coupled with variable nitrogen compositions suggests that Ryugu has experienced a rather unique history of alteration. Aqueous alteration of Ryugu appears to have played a significant role in heterogeneously modifying the nitrogen signature of different grains, whilst having limited impact on the noble gas budget. This is likely due to the loss of a N-rich soluble organic component, as this would not significantly alter the noble gas budget. Ryugu may therefore represent a unique sample archive for investigating the variable composition of alteration fluids on chondritic bodies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The Hayabusa2 mission was developed under the leadership of the Japan Aerospace Exploration Agency (JAXA), with contributions from the German Aerospace Center (DLR) and the Centre National d'Études Spatiales (CNES), and in collaboration with NASA, and other universities, and institutes. This work was primarily funded by the European Research Council (PHOTONIS Advanced Grant # 695618 awarded to BM and VOLATILIS Starting Grant 715028 awarded to EF) and by the Centre National d'Études Spatiales (CNES). RO was supported by JSPS KAKENHI grants JP19H01959 and JP20H05846. Work by D.K., M.R. and H.B. was carried out within the framework of the NCCR PlanetS and the Ambizione program supported by the Swiss NSF (grants 200020_182649, 51NF40-182901, PZ00P2_193331). JG, SC, TL and LF were supported by STFC grants numbers ST/R000751/1 and ST/V000675/1. PLC was supported by UKRI FLF grant number MR/S03465X/1. KH and AI were supported by JSPS KAKENHI Grant Number

JP20H00190. We are thankful to the Associate Editor, Ashley King and an anonymous reviewer for their helpful and constructive feedback on this manuscript. This is CRPG contribution 2833.

Appendix A. Supplementary material

The following document includes a more detailed outline of the analytical methods used in this study, as well as supplementary figures and data tables. Supplementary material to this article can be found online at <https://doi.org/10.1016/j.gca.2023.01.020>.

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