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► To cite this version:

J. Gamblin, E. Füri, B. Marty, L. Zimmermann, D.V. Bekaert. Dissecting the complex Ne-Ar-N signature of asteroid Ryugu by step-heating analysis. Geochemical Perspectives Letters, 2024, 31, pp.44-48. 10.7185/geochemlet.2431. hal-04677606

HAL Id: hal-04677606 https://hal.science/hal-04677606v1

Submitted on 27 Aug 2024

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Geochemical C) EAG Perspectives Letters

 $$\ensuremath{\mathbb{C}}\xspace$ 2024 The Authors Published by the European Association of Geochemistry

Dissecting the complex Ne-Ar-N signature of asteroid Ryugu by step-heating analysis

J. Gamblin^{1*}, E. Füri¹, B. Marty¹, L. Zimmermann¹, D.V. Bekaert¹

Abstract





Samples returned from the carbonaceous asteroid (162173) Ryugu show mineralogical, chemical, and isotopic similarities with Ivuna-type (CI) carbonaceous chondrites, which likely contributed to Earth's volatile inventory. To better understand the complex Ne-Ar-N signature of CI-type material, we analysed a single, mg-sized Ryugu particle by multi-step (n = 85) heating. Noble gases (Ne, Ar) are a mixture between implanted Solar Wind (SW), presolar component(s), and the carbonaceous phase Q, with negligible cosmogenic contributions. The δ^{15} N variations observed during progressive heating reflect the presence of various N-bearing phases. The large number of heating steps provide key insights into the effect of thermal processing on the N abundance and isotopic ratio, and indicate that low temperatures can result in exten-

sive N loss from CI-type material, without significantly affecting the bulk N isotopic composition. Nitrogen isotopes, therefore, remain a reliable and powerful tool for tracing volatile sources in the Solar System.

Received 10 April 2024 | Accepted 25 June 2024 | Published 1 August 2024

Introduction

The Hayabusa2 mission of the Japan Aerospace Exploration Agency (JAXA) collected 5.4 g of regolith on the Cb-type asteroid (162173) Ryugu during two touchdowns in 2019 and returned the samples to Earth on December 6th, 2020 (Tsuda et al., 2020). The mineralogy and the chemical and isotopic compositions of the samples revealed a close relationship between Ryugu and Ivuna-type (CI) carbonaceous chondrites (Nakamura et al., 2022; Yokoyama et al., 2023). However, unlike CI chondrites collected on Earth, material from Ryugu has not been affected by heating processes during atmospheric entry or by terrestrial weathering. Since carbonaceous chondrites may have played an important role in supplying volatile elements to Earth (e.g., Alexander et al., 2012; Marty, 2012; Alexander, 2022), Ryugu samples are key for better understanding the origin of terrestrial volatiles. The Hayabusa2-initial-analysis volatile team and the Phase-2 curation team carried out the first noble gas and N analyses, thus providing insights into the volatile composition, formation, and alteration history of Ryugu (Nakamura et al., 2022; Okazaki et al., 2023). The noble gases were found to be mainly primordial (i.e. carried by the so called phase Q and a variety of presolar components), with variable contributions from solar wind (SW) and cosmogenic isotopes. Okazaki et al. (2023) concluded that the heterogeneous N contents and isotopic compositions of Ryugu samples, including four small pelletised samples and three splits of a large aggregate sample (Naraoka et al., 2023), indicate the presence of at least two carrier phases: a N-rich phase with $\delta^{15}N$ up to +70 ‰ and a N-depleted phase with $\delta^{15}N$ near 0 ‰. These results were subsequently discussed in more detail by Broadley et al. (2023) and Hashizume et al. (2024).

Here, we aim to better understand the nature and behaviour of Ne-Ar-N carriers in Ryugu material by performing a large number (n = 85) of extraction steps at incrementally increasing temperature. This is the first time, to our knowledge, that a single asteroidal particle has been heated in so many steps for coupled noble gas and N analyses, thereby providing unprecedented insight into the complex Ne-Ar-N makeup of primitive extraterrestrial matter.

Material and Methods

Particle C0015 (1.8 ± 0.2 mg), collected during the second touchdown on Ryugu within a crater created by the spacecraft's small carry-on impactor, was targeted for step-heating Ne-Ar-N analysis at the Centre de Recherches Pétrographiques et Géochimiques (CRPG) noble gas facility. The particle was never in contact with Earth's atmosphere and was constantly kept within a dry N₂ atmosphere or under vacuum during sample preparation, storage, and analysis. The particle was heated under static vacuum using a CO_2 laser at increasing laser power, and the fraction of noble gases (Ne, Ar) and N (in the form of N₂) extracted at each step was analysed using a Noblesse–HR noble gas mass spectrometer in multicollection. A total of 85 heating steps were performed to study the progressive release of different noble gas and N components. Details on the analytical procedure and data treatment are provided in the Supplementary Information.

Ne-Ar-N Release Patterns of Particle C0015

Figure 1 shows the release patterns of Ne, Ar, and N (analysed in the form of N_2) and the corresponding isotope ratios measured

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Figure 1 (a) ²⁰Ne, (b) ³⁶Ar, and (c) N₂ abundances (filled bars) and isotope ratios (filled circles) of particle C0015 for 85 extraction steps. Abundances are weighted by the extraction duration (*i.e.* 4 min for the first 26 steps and 12 min for the following 59 steps). Isotope ratios are shown for heating steps with <30 % blank contributions. The different colours represent the laser power used for the extraction (*i.e.* 1 to 45 % of the total power of 32 W). The dashed lines indicate the calculated "bulk" ²⁰Ne/²²Ne, ³⁸Ar/³⁶Ar, and $\delta^{15}N$ values. Isotope ratios of the solar wind (SW), phase Q, and Ne-HL are shown for comparison (see Ott, 2014 and references therein). Uncertainties are 1 σ and error bars are, in most cases, smaller than symbol sizes.

at each heating step. Most of the Ne was released at very low laser power (\leq 3 %) at which the camera view showed no visible heating (indicated by an orange glow) of the particle. The 20 Ne/ 22 Ne ratio was nearly constant (12.92 ± 0.47) during the first 38 extraction steps (Fig. 1a), only slightly below the Ne isotope ratio of SW $(13.74 \pm 0.02 \text{ to } 14.00 \pm 0.04; \text{ Ott, } 2014)$ and close to that of the protosolar nebula (~13.36; Heber et al., 2012). This plateau likely results from the extraction of a pure Ne component. While trapping of nebular gas in CI-type material cannot be ruled out, the ³He/⁴He ratio of solar-gas-rich Ryugu samples analysed previously is consistent with implanted SW-derived gas (Meshik et al., 2023; Okazaki et al., 2023). Neon released at low temperatures must, therefore, be predominantly derived from the SW, fractionated to an isotopically lighter value upon implantation and grain surface sputtering (Grimberg et al., 2006). At higher temperatures, the ²⁰Ne/²²Ne ratio first decreased significantly to 9.39 ± 0.12 , before increasing to 12.03 ± 0.18 . Since the ²¹Ne/²²Ne ratio varied only between ~0.026 and 0.049 (Fig. 2), which implies a small proportion of cosmogenic Ne (i.e. <0.1 % ²¹Ne_{cosm}) in particle C0015, the ²⁰Ne/²²Ne variations are inferred to predominantly reflect the release of the primordial components Ne-Q and Ne-HL. The Ne amounts released during the last 15 steps were too low for reliable isotope ratio measurements. The results indicate the presence of at least three Ne components in Ryugu particle C0015: SW-derived Ne implanted at the grain surface, Ne-HL carried by presolar nanodiamonds, and Ne-Q (or P1) carried by phase Q (Fig. 2b). Overall, the Ne composition of particle

C0015 is dominated by the SW component (Fig. 2a), similar to two pellets studied by the Hayabusa2-initial-analysis volatile team (Okazaki *et al.*, 2023) and several particles analysed by the Phase-2 curation team (Nakamura *et al.*, 2022).

The ⁴⁰Ar signal was small for steps #2 and #5, and comparable to the blank value throughout the rest of the heating procedure, demonstrating the absence of any adsorbed atmospheric Ar. The ³⁸Ar/³⁶Ar ratio of the Ar fraction released at low laser power first oscillated between the Ar-SW (0.1818 ± 0.0005 to $0.1828 \pm$ 0.0010; Ott, 2014) and Ar-Q values (0.1872 ± 0.0007; Wieler et al., 1992), then plateaued around Ar-Q (Fig. 1b). Ar-HL (0.227 ± 0.003; Ott, 2014), expected to be released from presolar diamonds at relatively low temperatures (Supplementary Information), likely also contributed to the observed variations. Unlike Ne, a large amount of Ar was released at higher temperatures, implying that a significant proportion of Ar in particle C0015 was carried by refractory phases. The elevated ³⁸Ar/³⁶Ar ratios observed during sample melting (*i.e.* after step #80, at \geq 24 % laser power) can be explained by a very small contribution of cosmogenic $^{38}\mathrm{Ar}$ (($^{38}\mathrm{Ar}/^{36}\mathrm{Ar})_\mathrm{cosm}$ ~1.54; Wieler, 2002). The calculated bulk 38 Ar/ 36 Ar ratio (0.1865 ± 0.0001; Table S-1) is comparable to that of Ar-Q and similar to the values previously reported for Ryugu $(0.186 \pm 0.001 \text{ to } 0.194 \pm 0.007; \text{ Okazaki et al., 2023}).$

The amount of N released and the corresponding δ^{15} N varied significantly for the different extraction steps, reflecting the presence of several, isotopically distinct N components in Ryugu material. The blank contribution exceeded 30 % of the





Figure 2 (a) Neon isotopic composition of Ryugu particle C0015 (bulk represented by the yellow star; individual extraction steps represented by the small circles) compared to pelletised and fragment Ryugu samples analysed by the Hayabusa2-initial-analysis volatile team and particles analysed by the Phase-2 curation team (Institute for Planetary Materials, Okayama University) (pink diamonds), as well as CI chondrites (grey squares) (Nakamura *et al.*, 2022; Broadley *et al.*, 2023; Meshik *et al.*, 2023; Okazaki *et al.*, 2023 and references therein). Different Ne end members are represented by grey crosses (Ott, 2014). Ne-E is mainly carried by presolar graphite and silicon carbide. **(b)** Close-up (corresponding to the dashed rectangle in panel (a) of the Ne isotopic variations observed for the 85 extraction steps of particle C0015. The different colours represent the laser power used for each extraction. Uncertainties are 1σ.

measured N₂ signal at step #1, and a δ^{15} N of +18.4 ± 1.0 ‰ was measured at step #2. This indicates that the contribution of any dry N₂ adsorbed onto the sample surface during sample storage and handling must be negligible, provided that this component is characterised by an atmospheric isotope signature. Whereas the δ^{15} N ranged from +18.1 ± 1.0 up to +44.5 ± 1.1 ‰ during the first 52 steps (*i.e.* at a low laser power, ≤ 4 %), δ^{15} N increased to +65.8 ± 1.1 ‰ at step #69 and then suddenly dropped to +1.0 ± 1.0 ‰ at step #70. No clear SW contribution (δ^{15} N_{SW} = -407 ± 7 ‰; Marty *et al.*, 2011) could be identified, even for low temperature extraction steps. Thus, the N release pattern is clearly decoupled from that of Ne. Assuming an unfractionated SW ²⁰Ne/¹⁴N elemental abundance ratio of 1.14 (Marty *et al.*, 2010), any SW-derived N is indeed expected to be undetectable in the ²⁰Ne-rich particle C0015.

The production of cosmogenic ¹⁵N during exposure to cosmic rays can potentially modify the N isotope ratio of



Figure 3 Nitrogen isotopic composition (δ^{15} N) as a function of the inverse of the N concentration of Ryugu samples compared to CI chondrites and Y-980115, a CI chondrite recovered in Antarctica that has recently been recommended to be re-classified as a Yamato-type (CY) carbonaceous chondrite (King *et al.*, 2019). Adapted from Okazaki *et al.* (2023) and Hashizume *et al.* (2024), including data reported in Table S-2. A simple two-component mixture, between a N-rich phase with δ^{15} N up to +70 ‰ and a N-depleted phase with δ^{15} N near 0 ‰, fails to explain the N signature of Ryugu particle C0015.

extraterrestrial samples. Combined analyses of noble gases and N make it possible to quantify the amount of ¹⁵N_{cosm} and to identify the isotopic composition of primordial N. Previous Ne analyses revealed that Ryugu samples record short cosmic ray exposure (CRE) ages of ~100 kyr to 8 Myr (Nakamura et al., 2022; Okazaki et al., 2023). The Ne isotopic composition of particle C0015 studied here closely resembles that of pellets A0105-15 and A0105-06, which are dominated by Ne-SW and contain a negligible amount of cosmogenic Ne. By using the same 21 Ne_{cosm} production rate as Okazaki *et al.* (2023) (*i.e.* $P_{21} = 1.34 \times 10^{-13}$ mol g⁻¹ Myr⁻¹), together with a simplified one-stage irradiation model, the CRE age of particle C0015 is estimated at ~30,000 years. This exposure duration could result in the production of $\sim 4 \times 10^{-17}$ mol ^{15}N (when using the maximum $({\rm ^{15}N/^{21}Ne})_{\rm cosm}$ = 5.5 from Mathew and Murty, 1993, estimated for H/L chondrites), which is negligible compared to the bulk N content of the sample. Since particle C0015 has neither been measurably affected by the production of cosmogenic ¹⁵N nor atmospheric contamination and N-SW implantation, it preserves key information on N components and carrier phases in Ryugu and CI-type material. However, distinguishing between different labile and more refractory N components in particle C0015 is challenging because, in contrast to the noble gases, the nature and N isotope compositions of potential chondritic N carrier phases are highly complex, as detailed in the Supplementary Information. While the different N carrier phases cannot be deciphered, the observed N release pattern (Fig. 1c) and the summed bulk N signature (Fig. 3) require the presence of more than just two N components, in contrast to previous findings of Okazaki et al. (2023) and Hashizume et al. (2024).

Discussion

The mineralogical, chemical, and isotopic characteristics of Ryugu revealed a close relationship with CI-chondrites



(Nakamura *et al.*, 2022; Yokoyama *et al.*, 2023). The CI group nominally comprises the five falls Orgueil, Alais, Ivuna Tonk, and Revelstoke, whose bulk N isotopic compositions vary between +31 and +61 ‰ (based on analyses of Alais, Ivuna, and Orgueil; Table S-2 and references therein) (Fig. 3). It is noteworthy that the Meteoritical Bulletin lists an additional four CIs that were recovered in the Yamato Mountains in Antarctica (Y-86029, Y-86737, Y-980115, and Y-980134), two of which (Y-86029 and Y-980115) have been recommended to be reclassified as Yamato-type (CY) chondrites (King *et al.*, 2019). The bulk δ^{15} N of Y-980115 was reported to vary from -2.8 ‰ to +4.0 (Chan *et al.*, 2016; Hashizume *et al.*, 2024) (Fig. 3), distinct from the CI signature.

The N abundances and isotopic compositions of several particles collected at Ryugu's surface (Ryugu-A samples) or within an artificial crater (Ryugu-C samples) fall within the CI range; however, some samples show lower N contents and/or δ^{15} N values (Fig. 3; Nakamura *et al.*, 2022; Broadley *et al.*, 2023; Naraoka et al., 2023; Oba et al., 2023; Hashizume et al., 2024). The four pelletised samples analysed by the Havabusa2-initial-analysis volatile team have particularly low N contents (Table S-2). The bulk δ^{15} N of $+24.43 \pm 0.17$ ‰ of particle C0015 (Table S-1) is comparable to the values previously obtained for the pellets A0105-05 and C0106-06 by noble gas mass spectrometry at CRPG $(+18.14 \pm 0.94 \text{ }\% \text{ and } +19.47 \pm$ 0.89 ‰; Broadley et al., 2023; Okazaki et al., 2023), whereas its summed bulk N abundance (1760 \pm 195 ppm; Table S-1) is consistent with the range observed in most CI chondrites (1400 to 2400 ppm; Table S-2 and references therein). Notably, particle C0015 contains more Ne and Ar than CI chondrites $({}^{36}\text{Ar} = 4.33 \pm 2.78 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm 0.03 \times 10^{-11} \text{ mol/g and } {}^{20}\text{Ne} = 1.53 \pm$ 10⁻¹¹ mol/g, on average, in CIs; Broadley et al., 2023 and references therein), but the large noble gas abundance predominantly results from SW irradiation.

Broadley et al. (2023) suggested that preferential loss of ¹⁵N-rich soluble organic matter during aqueous alteration on Ryugu's parent body may have resulted in a lower δ^{15} N and lower N concentrations in Ryugu samples than in CIs, without significantly affecting the noble gas budget. According to this scenario, the N heterogeneities between the various small, (sub-)milligram-sized Ryugu samples could be due to variable degrees of aqueous alteration on the initial parent body, prior to the catastrophic break-up that turned Ryugu into a rubble-pile asteroid and led to mixing between more and less altered clasts (Broadley et al., 2023). However, this process alone can neither explain the association of "intermediate" $\delta^{15}N$ values with high N abundances in particles C0015 (this study) and A0033 (Nakamura *et al.*, 2022), nor the very low δ^{15} N value and high N content of particle C0082 (Nakamura et al., 2022) (Fig. 3). Significant heterogeneities of the N distribution must, therefore, exist at the scale of (sub-)milligram-sized Ryugu regolith particles, as also demonstrated by in situ secondary ion mass spectrometry analyses of N (Nakamura et al., 2022).

CI-type material is inferred to have played an important role in supplying volatile elements (*e.g.*, H, C, N) to Earth's surface (Piani *et al.*, 2020). However, since thermal metamorphism and accretionary heating could have resulted in extensive devolatilisation and loss of ¹⁵N-rich components (Alexander *et al.*, 1998, Alexander *et al.*, 2007; Pearson *et al.*, 2006; Grewal, 2022), assessing the evolution of the N content and isotopic composition during progressive heating is key for understanding the contribution of CIs to Earth's volatile budget. The numerous heating steps performed on Ryugu particle C0015 provide key information on N abundance and isotopic variations induced by thermal processing, especially since Ryugu never experienced temperatures >150 °C (based on analyses of soluble organic



Letter

Figure 4 Evolution of the residual N isotopic composition $(\delta^{15}N_{residual})$ of particle C0015 as a function of the fraction of N extracted by progressive heating.

matter in Ryugu; Naraoka *et al.*, 2023). Figure 4 illustrates that the "bulk" δ^{15} N value of particle C0015 was not significantly modified during the first 60 heating steps (*i.e.* δ^{15} N = 22.05 ± 0.22 ‰ to 24.47 ± 0.17 ‰ for 1 to 7 % of the maximum laser power), although up to ~73 % of the total initial N content was lost. As heating proceeded and particle C0015 started to melt at step #80, the δ^{15} N decreased to 4.47 ± 0.47 ‰ (with concomitant loss of ~99 % N). This observation confirms that Ryugu samples contain a refractory N component with an isotopic composition similar to Earth's atmosphere (Hashizume *et al.*, 2024) whose proportion of the total amount of N, however, is very small.

Overall, N-rich material collected at Ryugu's surface is predominantly characterised by an isotopic signature that is comparable to, or only slightly lighter than, that of CI chondrites ($\delta^{15}N \ge +18$ ‰; Fig. 3; Table S-2). Furthermore, our new data demonstrate that low temperatures can result in extensive N loss from CI-type material, without significantly affecting the bulk N isotope composition. Only high temperatures result in loss of a ¹⁵N-rich component and a notable decrease of the bulk $\delta^{15}N$ value by ~20 ‰. Consequently, and despite potential losses of thermally labile N-bearing phases, N isotopes remain a reliable and powerful tool for tracing contributions from inner and outer Solar System sources, and they imply that CI- or Ryugu-type material can be ruled out as the major source of N in Earth's mantle.

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 Union (ERC, IRONIS, 101087562). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Research Council. Neither the European Union nor the granting authority can be held responsible for them. Support from the Centre National d'Études Spatiales (CNES) is also acknowledged. We thank Michael W. Broadley for his help with handling the Hayabusa2 samples as well as Bouchaib Tibari and Allan



Bauer for their assistance with the noble gas mass spectrometry analyses at CRPG. We are also grateful to R. Wieler, C.M.O'D. Alexander, and an anonymous reviewer for their constructive comments, and to A. Shahar for editorial handling. This is CRPG contribution 2858.

Editor: Anat Shahar

Additional Information

Supplementary Information accompanies this letter at https:// www.geochemicalperspectivesletters.org/article2431.



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Cite this letter as: Gamblin, J., Füri, E., Marty, B., Zimmermann, L., Bekaert, D.V. (2024) Dissecting the complex Ne-Ar-N signature of asteroid Ryugu by step-heating analysis. *Geochem. Persp. Let.* 31, 44–48. https://doi.org/10.7185/ geochemlet.2431

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