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Origin of radiogenic ^{129}Xe variations in carbonaceous chondrites

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Abstract

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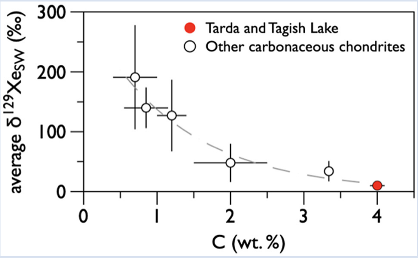

Carbonaceous chondrites are pristine witnesses of the formation of the solar system. Among them, the carbon-rich Tarda and Tagish Lake meteorites are thought to have sampled very distant regions of the outer circumsolar disk (Hiroi *et al.*, 2001). Here, we show that their noble gas isotopic compositions (especially ^{129}Xe excesses) are similar, implying their formation in comparable environments. Combined with literature data, we show that the radiogenic excesses of ^{129}Xe relative to solar wind in carbonaceous chondrites define anti-correlations with their respective iodine and carbon contents. These trends do not result from the heterogeneous distribution of ^{129}I in the disk but rather evidence a xenon dilution effect; the radiogenic ^{129}Xe excesses being dominated by trapped xenon in the most carbon-rich carbonaceous chondrites. Our data also suggest that both Tarda and Tagish Lake accreted beyond 10 astronomical units, in regions of the disk that were cold enough for CO_2 to condense.

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Introduction

Noble gases trapped in primitive meteorites (chondrites) allow quantification of the physical processes that operated during the evolution of the protoplanetary disk (*e.g.*, Kuga *et al.*, 2015). Although these elements are present in different carriers contained in meteorites (including presolar SiC, diamonds, graphite; Ott, 2014), they are mainly hosted in a phase – referred to as phase Q – whose nature is still poorly characterised (Busemann *et al.*, 2000). Notwithstanding this uncertainty, it has been shown that phase Q dominates the heavy noble gas budget of chondrites and is closely associated with carbonaceous material that survives HF/HCl attack of bulk meteorites (Lewis *et al.*, 1975). Thanks to its extreme sensitivity to oxidation, the xenon isotopic composition of phase Q has been precisely determined, revealing a mass dependent isotopic fractionation relative to solar wind (SW-Xe) in favour of the heavy isotopes relative to the light ones (Wieler *et al.*, 1991; Busemann *et al.*, 2000; Gilmour, 2010). However, the commonly used Xe-Q isotopic composition hinges on the average of measurements of several carbonaceous chondrites (CCs) showing distinct Xe isotopic compositions between and within each group, especially for ^{129}Xe (Busemann *et al.*, 2000). Such ^{129}Xe excesses result from the decay of extinct ^{129}I ($t_{1/2} = 16$ Myr), which was producing radiogenic ^{129}Xe* during the first $\sim$100 million years of the solar system (Jeffery and Reynolds, 1961). The measurement of xenon isotopes in the coma of comet 67P/Churyumov-Gerasimenko revealed extreme ^{129}Xe enrichment relative to ^{132}Xe and the solar composition (Marty *et al.*, 2017). As this large monoisotopic excess would require unlikely ^{129}I enrichment, it has been interpreted as originating from a specific nucleosynthetic process producing ^{129}Xe that was sampled by icy bodies formed in the outer solar system (Marty *et al.*, 2017). Interestingly, the carbon-rich primitive chondrites Tagish Lake and Tarda are thought to originate from D-type asteroids (Hiroi *et al.*, 2001; Marrocchi *et al.*, 2021) considered to have formed at large heliocentric distances beyond the current orbit of Saturn, and potentially as far as the Kuiper Belt (*i.e.* 30–50 astronomical units = au; Levison *et al.*, 2009). Here we report the results of a comprehensive study of the isotopic compositions of noble gases contained in Tagish Lake and Tarda to evaluate if material accreted in the outer solar system presents specific signatures. We compare our data to other CCs and discuss the origin of the variable radiogenic ^{129}Xe excesses between and within each CC groups.

Material and Methods

Noble gases were extracted from bulk fragments of Tarda, Tagish Lake and Orgueil meteorites by a laser step-heating method and measured with a noble gas mass spectrometer. Uncertainties on isotope ratios include internal uncertainties, external uncertainties assessed by measurements of standard aliquots, and uncertainties on the blank contribution. Details on the analytical procedure are in the Supplementary Information.

Results of Noble Gas Measurements and Cosmic-ray Exposure Ages

Abundances and isotopic compositions of Ne, Ar, Kr and Xe extracted from bulk Tarda, Tagish Lake and Orgueil samples are reported in Table S-1. Elemental abundances of Ne, Ar,

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Kr and Xe in Tarda, Tagish Lake and Orgueil are similar to those reported for other volatile-rich carbonaceous chondrites (Table S-1; Mazor *et al.*, 1970). Most heating steps show a similar $^{20}Ne/^{132}Xe$ ratio (average value of 22 ± 4), slightly lower than the range reported for the HL component (50 ± 20; Huss and Lewis, 1994). For all samples, $^{36}Ar/^{132}Xe$ and $^{84}Kr/^{132}Xe$ ratios plot close to the Q component although the first, low temperature, heating steps are systematically plotting toward higher $^{84}Kr/^{132}Xe$ ratios, which are compatible with a contribution from weakly bound atmospheric gases (Fig. S-1). For all heating steps, the isotopic composition of neon indicates the presence of abundant trapped neon in the different meteorite samples (Fig. 1). Data points of heating steps of Tarda and Tagish Lake samples plot slightly below a mixing line defined by Ne-HL (Huss and Lewis, 1994) and cosmogenic neon (Supplementary Information). The two first heating steps of Orgueil samples plot on the Ne-Q/cosmogenic mixing line while the high temperature extraction steps show lower $^{21}Ne/^{22}Ne$ ratios and plot close to the Ne-Q/Ne-HL mixing line. For argon, $^{38}Ar/^{36}Ar$ ratios are compatible with either the atmospheric $^{38}Ar/^{36}Ar$ ratio (≈0.188; Ozima and Podosek, 2002) or the $^{38}Ar/^{36}Ar$ ratio of argon in phase Q (≈0.187; Ott, 2002). However, the $^{40}Ar/^{36}Ar$ ratios range from 3 to 43, well below the atmospheric value (≈300, Ozima and Podosek, 2002), but typical for trapped argon contained in carbonaceous chondrites (Krietsch *et al.*, 2021). The isotopic ratios of Kr and Xe are distinct from those of air, as well, and are similar again, to those measured for bulk carbonaceous chondrites (*e.g.*, Krietsch *et al.*, 2021). The first heating steps of Tarda and Tagish Lake samples reveal the presence of weakly bound atmospheric gases (Fig. S-2). For the $^{129}Xe/^{130}Xe$ ratio, high temperature heating steps of Tarda and Tagish Lake samples gave reproducible results with an average value of 6.37 ± 0.01 (1σ s.d.). This value is 8.3 ± 3.4 ‰ lower than the $^{129}Xe/^{130}Xe$ measured for Q-Xe (Busemann *et al.*, 2000). High temperature heating steps of Orgueil samples reveal the presence of excess radiogenic ^{129}Xe compared to Q-Xe.

The presence of abundant trapped Ne in both Tarda and Tagish Lake prevents us from determining precisely the cosmogenic $^{22}Ne/^{21}Ne$ ratio and thus cosmogenic ^{21}Ne production rates (Supplementary Information). Tarda has a cosmic ray exposure (CRE) age within 5–12 Ma, very similar to Tagish Lake (5–8 Ma), while for Orgueil, the possible CRE age ranges from 6 to 11 Ma. The nominal (K-Ar) radiogenic gas retention ages are 2.4–2.7 Ga for Tarda, 2.0–2.8 Ga for Tagish Lake, and 2.2–2.7 Ga for Orgueil.

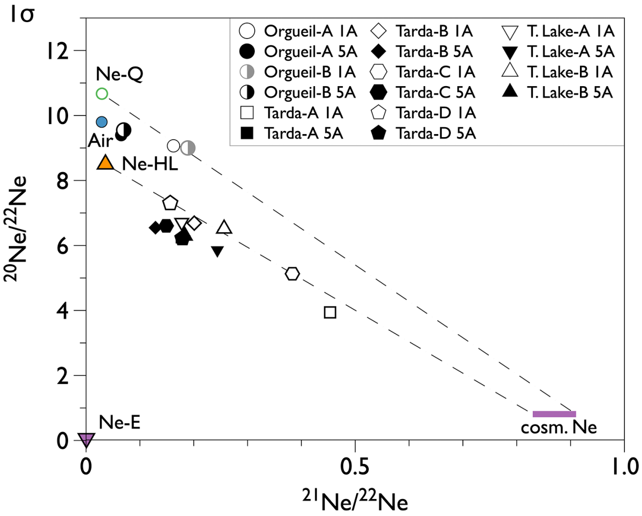

Figure 1 Neon three isotope plot for bulk samples of Tarda, Tagish Lake and Orgueil. The compositions of Ne-Q, Air, Ne-HL, Ne-E and cosmogenic (purple range) are also shown for comparison (see Ott, 2014 and Krietsch *et al.*, 2021 and refs. therein). The two dashed lines represent mixing arrays between Ne-Q and cosmogenic neon and Ne-HL and cosmogenic neon. Error bars (1σ) are smaller than the symbols.

Discussion

Based on multiple isotopic systems (*i.e.* H, C, N and O), it has recently been proposed that Tarda and Tagish Lake could be genetically related (Marrocchi *et al.*, 2021). This hypothesis can be tested in the light of noble gas measurements reported here. In the three isotope diagram, the neon isotopic compositions of bulk chondrites plot within a space defined by cosmogenic Ne, Ne-Q and a pole with ($^{20}Ne/^{22}Ne$) slightly below that of Ne-HL carried by presolar nanodiamonds (Fig. 1; Huss and Lewis, 1994; Krietsch *et al.*, 2021). The latter is likely due to the presence of Ne-E from presolar SiC or graphite (Riebe *et al.*, 2020). The data points from Tarda and Tagish Lake plot on the lower part of this isotopic space with similar Ne isotopic compositions, which are clearly resolved from that of the CI chondrite Orgueil (Fig. 1). Our results show that both Tarda and Tagish Lake have similar bulk Xe spectra and $^{129}Xe^*$ excesses (Fig. 2), with $\delta^{129}Xe_{SW} = 10 \pm 3$ ‰ (Fig. 3a). In addition, both chondrites show similar cosmic-ray exposure and radiogenic retention ages: 5–10 Ma and 2.4–2.7 Ga for Tarda, and 5–8 Ma and 2.0–2.8 Ga for Tagish Lake. Altogether, our results thus reinforce the genetic link between Tarda and Tagish Lake, which share similar isotopic signatures for elements having drastically different geochemical behaviour (Marrocchi *et al.*, 2021).

Xenon in the Jupiter-family comet 67P/Churyumov-Gerasimenko (67P/C-G) presents a ^{129}Xe excess and important, tens of percent $^{134-136}Xe$ deficits relative to SW-Xe (Marty *et al.*, 2017). The former has been attributed as resulting from the contribution of parentless ^{129}Xe and the latter of a mixture of two nucleosynthetic processes (*i.e.* s- and r-process; Marty *et al.*, 2017) different from the one measured for most inner solar system material (Avice *et al.*, 2020). This is however not observed in Orgueil, Tarda and Tagish Lake (Fig. 2) whereas they are generally thought to have formed in the outer solar system, at large heliocentric distances >10 au (Desch *et al.*, 2018; Fujiya *et al.*, 2019; Marrocchi *et al.*, 2021). These meteorites are even showing among the weakest $^{129}Xe^*$ excesses measured in CCs (Fig. 3a;

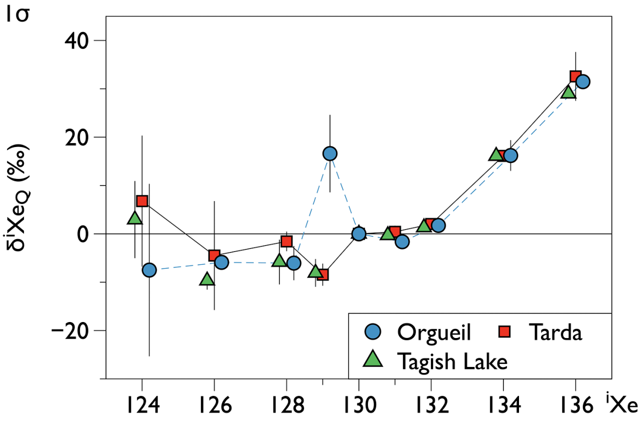

Figure 2 Isotopic composition of total xenon extracted from bulk Tarda, Tagish Lake and Orgueil samples. Isotopic ratios are normalised to Q-Xe (Busemann *et al.*, 2000) and expressed with the delta notation ($\delta^iXe_Q = ((^iXe/^{130}Xe)_{sample}/(^iXe/^{130}Xe)_Q - 1) \times 1000$). Errors are at 1σ.

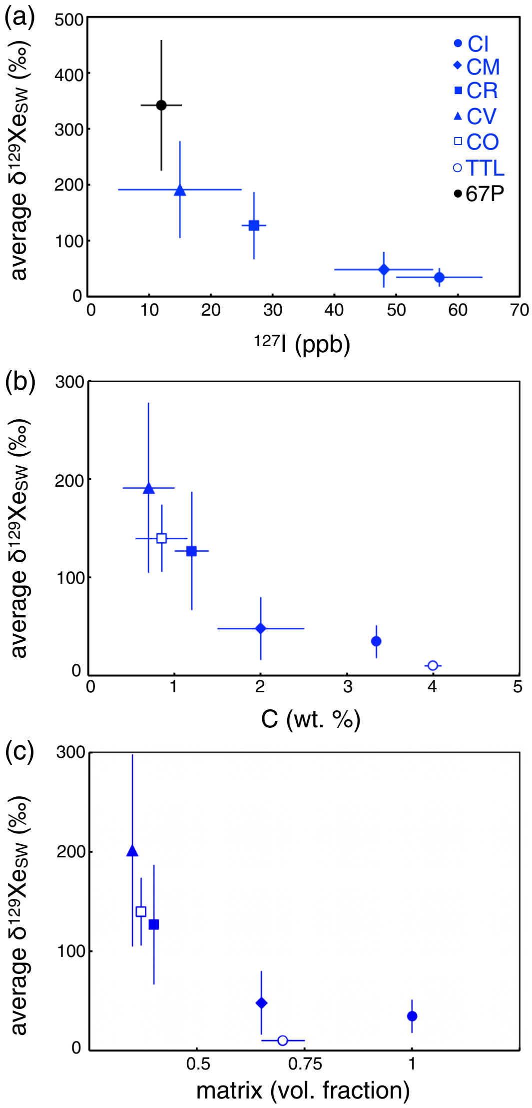

Figure 3 Average $^{129}\mathrm{Xe}^*$ excesses relative to SW-Xe (expressed in δ notation with $\delta^{129}\mathrm{Xe}_{SW} = (^{129}\mathrm{Xe}/^{132}\mathrm{Xe})_{bulk}/(^{129}\mathrm{Xe}/^{132}\mathrm{Xe})_{SW} - 1 \times 1000$) for the different types of chondrites and the comet 67P/C-G (data from Mazor *et al.*, 1970; Marty *et al.*, 2017 and this study). The average $\delta^{129}\mathrm{Xe}_{SW}$ is plotted as a function of **(a)** the ^{127}I content (data from Clay *et al.*, 2017), **(b)** the carbon content (data from Vacher *et al.*, 2020 and Marrocchi *et al.*, 2021), and **(c)** the matrix abundance (data from Alexander *et al.*, 2018). TTL = Tarda and Tagish Lake.

Mazor *et al.*, 1970). Some rare CMs show similar ^{129}Xe/^{132}Xe but suffered from significant heating (Alexander *et al.*, 2012; Krietsch *et al.*, 2021). In addition, when combining data from all CCs and the comet 67P (Mazor *et al.*, 1970; Clay *et al.*, 2017; Marty *et al.*, 2017), we observe an anti-correlation of their ^{127}I content and the ^{129}Xe excess (Fig. 3a), regardless of the available iodine dataset used (Fig. S-4). As previously noticed (Mazor *et al.*, 1970; Gilmour *et al.*, 2001), such an inverse correlation could not result from the heterogeneous ^{129}I distribution in the early solar system as the absolute concentrations of radiogenic $^{129}\mathrm{Xe}^*$ vary by only a factor of ~4 among CCs, while relative $^{129}\mathrm{Xe}^*$ enrichments differ by a factor of 400.

CCs contains variable amounts of carbon with Tarda, Tagish Lake, CI and CM chondrites showing the highest concentrations (Fig. 3b; Kerridge, 1985; Vacher *et al.*, 2020; Marrocchi *et al.*, 2021). With the exception of Tarda and Tagish Lake, the carbon content of CCs is directly related to the abundance of fine grained matrix (see Fig. 3 in Alexander *et al.*, 2018). Interestingly, the $^{129}\mathrm{Xe}^*$ excess is also anti-correlated with the bulk C content of CCs (Fig. 3b), thus implying that the (i) ^{129}I carrier was located in the CC matrices, and (ii) variations of $^{129}\mathrm{Xe}^*$ excesses observed in CCs result from a dilution effect by trapped Xe located in phase Q. Such an effect can be summarised as follows: the less carbon, the less phase Q, the less trapped ^{129}Xe, the more the effect of ^{129}I decay is visible (and *vice versa*). This also indicates that the initial Xe budget (and likely that of other noble gases) in CCs is directly controlled by the abundance of matrix (Fig. 3c). Of note, similar $^{129}\mathrm{Xe}^*$-C anti-correlations are also observed within several CC groups (see Fig. S-5).

Although both Tarda and Tagish Lake are depleted in fine grained matrix relative to CI chondrites (65–80 *vs.* 100 vol. %, Fig. 3c; Alexander *et al.*, 2018), they appear enriched in C (*i.e.* ~4 *vs.* 3.3 wt. %, Fig. 3b; Vacher *et al.*, 2020; Marrocchi *et al.*, 2021). This excess has been attributed to the unusual abundance of carbonates in some highly altered lithologies of Tagish Lake (Alexander *et al.*, 2018). However, the bulk C content of Tagish Lake is relatively homogenous (Marrocchi *et al.*, 2021) regardless of the abundance of carbonates (*i.e.* 4.1 ± 0.1 wt. %). In addition, Tarda shows a carbon content similar to Tagish Lake whereas no specific carbonate-rich lithology has been described so far (Marrocchi *et al.*, 2021 and references therein). This thus requires an additional source of carbon for accounting for the diluted $^{129}\mathrm{Xe}^*$ excesses observed in both Tarda and Tagish Lake (Figs. 2, 3a). It has been recently proposed that peculiar carbon isotopic compositions of carbonates in Tagish Lake (*i.e.* $\delta^{13}\mathrm{C} \approx 70$ ‰; Fujiya *et al.*, 2019) cannot be explained without invoking the accretion of large amounts of ^{13}C-rich CO_2 cometary ices. This implies that the parent body of Tagish Lake (and Tarda) formed beyond 10 au, in regions of the protoplanetary disk that were cold enough for CO_2 to condense.

Conclusions

Results obtained in this study demonstrate that Tarda and Tagish Lake, in addition to C, H, N isotope systematics (Marrocchi *et al.*, 2021), present very similar compositions for noble gases. This implies that those meteorites are genetically related and may have sampled similar environments of the accretion disk. A key feature of xenon present in these two meteorites is a very low excess of radiogenic ^{129}Xe*. When compared to literature data of carbonaceous chondrites, these carbon-rich meteorite samples present inverse correlations between ^{129}Xe* and carbon or iodine content. We interpret these anti-correlations as evidence for a dilution effect of radiogenic ^{129}Xe* by primordial xenon trapped in organic matter.

Author Contributions

GA and YM designed the study. GA performed the measurements. MM reconstructed the cosmic histories. All authors worked on the data and on the manuscript.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2228.

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