The end of the isotopic evolution of atmospheric xenon


Supplementary Information

The Supplementary Information includes:

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Geological Context

Hydrothermal quartz samples were selected from two different cratons based on their ages of formation, between 2.3 and 2.5 Ga, encompassing the GOE (Holland, 2006; Lyons et al., 2014; Gumsley et al., 2017). The Ongeluk Formation is a succession of pillowed and sheeted andesitic basalts that has undergone alteration with temperatures not exceeding 200 °C and metamorphism up to prehnite-pumpellyite facies (Cornell et al., 1996; Gutzmer et al., 2001, 2003). This formation is intercalated with the Makganyene glaciation deposits formed during one of the Paleoproterozoic snowball earth events (Evans et al., 1997). Pods of quartz formed within the open spaces of the pillows soon after the eruption as indicated by the lack of deformation in the cavities (Gutzmer et al., 2003; Saito et al., 2016). The pillow-lavas are considered to have formed 2425.6 ± 2.6 Ma (2σ; Gumsley et al., 2017) and give an upper bound to the sample. Saito et al., (2018) performed Ar-Ar dating on three distinct quartz pods to estimate when the quartz crystals precipitated. They
found a maximum age of 2701 ± 43 Ma (1σ) and a minimum age of 2114 ± 312 (1σ) Ma. The difference is likely due to the difficulties in estimating the initial amount of K in the fluid. The age of ≈ 2701 Ma is unlikely given the age of the host rock. The petrographic description of the fluid inclusion indicates a predominantly primary origin (Saito et al., 2018). Thus, the minimum age of the trapped atmospheric noble gases within the inclusions is relative to the quartz pods formed at 2114 ± 312 Ma, consistent with the upper bound of the pillow lava formation.

The Seidorechka (sample FD1A) and Polisarka (sample FD3A) sedimentary formations are part of the Imandra-Varzuga Greenstone Belt. These formations were targeted by the Fennoscandian Arctic Russia – Drilling Early Earth Project (FAR-DEEP; Melezhik et al., 2013) of the International Continental Scientific Drilling Program. The Imandra-Varzuga Greenstone Belt consist of several successions of alternating thin sedimentary and thick volcanic formations overlying the Archean basement (Melezhik and Sturt, 1994; Warke et al., 2020). Hydrothermal quartz was collected from veins cross-cutting solely these sedimentary formations, in the shale member for FD1A and in the diamictite-greywacke member for FD3A. Depositional ages of Seidorechka and Polisarka sedimentary formations have been constrained between 2501.5 ± 1.7 Ma and 2441 ± 1.6 Ma (Amelin et al., 1995), and 2441 ± 1.6 Ma and 2434 ± 6.6 Ma (Brasier et al., 2013), respectively. We assumed that minerals contained in quartz veins formed contemporaneously with the overlying volcanic formations that define the minimum age of these sedimentary formations and thus yield ages of 2441 ± 1.6 Ma for the quartz veins crossing Seidorechka Sedimentary Formation (FD1A ; Amelin et al., 1995) and at 2434 ± 6.6 Ma for the quartz veins crossing Polisarka Sedimentary Formation (FD3A ; Brasier et al., 2013).

Materials and Methods

For noble gas analyses, quartz grains of 2 to 4 mm in size were handpicked under a binocular microscope and loaded into hand-activated crushers under vacuum. Gases were extracted using a 3.5 Nm torque wrench to activate each crusher. Purified Ar and Xe gas fractions were measured using a Thermofisher Helix MC Plus© mass spectrometer at CRPG. A total of 9 Ar and Xe analyses were performed on the Ongeluk quartz using the method of Avice et al. (2018) that consisted of replicated analyses on distinct crushes of 1 to 3 g of quartz. The Ar isotopic ratios were systematically analysed after each crush to check for the absence of atmospheric contamination. A new technique was used to study samples from the Kola Craton (Péron and Moreira, 2018). It consists of accumulating the extracted gases after crushing steps in an empty and previously evacuated steel bottle immersed in liquid nitrogen (77 K) during 10 minutes before purification and analysis. This method allowed the crushers to be recharged multiple times with fresh quartz grains and therefore enables the Ar and Xe gas fractions to be concentrated within the bottle. By using this method, we prepared 3 bottles for analyses: two bottles by crushing 16.70 g (FD1A-B1) and 10.08 g (FD1A-B2) of sample FD1A in 4 and 3 crusher loads respectively, and one bottle by crushing 18.16 g of sample FD3A in 4 separate crusher loads. The two
different bottles of FD1A extracted gases (FD1A – B1 and FD1A – B2) permitted the analysis of 4 and 5 aliquots of gas, respectively, to ensure reproducibility of the measurements. One bottle of extracted gases from FD3A permitted the analyses of 4 aliquots (Table S-3). The Ar abundance and isotopes were analysed only in the first gas aliquot for each bottle by multicollection. Isotopic ratios of Xe presented in Table 1 are weighted averages of the replicated analyses for each sample. For each sample, we estimated the potential mass-dependent fractionation of Xe (MDF-X, in permille per atomic mass unit ‰.u⁻¹, where a value of 0 indicates no MDF-Xe relative to modern atmospheric Xe) by computing linear regressions on \(^{124,126,128}\text{Xe}/^{130}\text{Xe}\) isotope ratios using the IsoplotR software (Vermeesch, 2018). Data are given with uncertainties at 2σ.

**Fluid inclusions Study**

**Petrography**

Rocks of Seidorechka and Polisarka sedimentary formations are respectively crossed by 3 pluri-centimetric (5 to 10 cm) hydrothermal quartz veins. These veins are restricted to each sedimentary formation and formed after sedimentation. In these volcano-sedimentary deposits (Melezhik et al., 2013), it is likely that the main fluid circulations are linked to the deposit of volcanic units overlying the sedimentary units. Thus, it is assumed that hydrothermal circulations are linked to the accumulation of Seidorechka and Polisarka volcanic formations overlying both sedimentary formations. Fluid inclusions were studied using 200-μm doubly polished thick sections.

Sample FD1A from Seidorechka sedimentary formations does not present obvious quartz recrystallisation but shows a general pattern of parallel trails of fluid inclusions confined within individual grains. At the grain scale, inclusions are either isolated or organised within parallel trails that do not cross grain boundaries (Fig. S-1). The boundaries between grains are decorated by fluid inclusions of less than 5 μm. The interior of the grains contains bigger inclusions, up to 20 μm, that allowed microthermometry and Raman analyses. The inclusions are thus considered primary in origin if they are isolated or pseudo-secondary when organised in trails within the grain structure, in any case trapped during quartz growth (Roedder, 1984). All the inclusions in FD1A are biphasic with the fraction of the bubble in the range 10–20 % of the total volume. Between 5 and 10 % of the inclusions are characterised by the typical CO₂ double bubble feature at room temperature (e.g., Diamond, 2003).

Petrography of fluid inclusions in FD3A from Polisarka sedimentary formation is complex due to quartz deformation and recrystallisation, which resulted in (i) undulose extinction under microscope and (ii) formation of newly recrystallised grains, over ~15 % of the studied thin-section, depleted in fluid inclusions (Fig S-1; Kerrich, 1976). Given the scarcity of 1 to 5 μm fluid inclusions within recrystallised quartz, the recrystallisation has a negligible effect on noble gas analyses compared to non-recrystallised quartz grains where most of fluid inclusions are observed. In the
latter, inclusions can be isolated, organised as trails confined within grains or as decorating quartz sub-grain boundaries. These observations are coherent with a continuous fluid entrapment contemporaneous with formation and deformation of host quartz (Eglinger et al., 2014). No evidence of secondary fluid inclusions was observed. Liquid monophasic and biphasic inclusions about 10 to 20 μm may coexist within the same grain (Fig. S-1). The volume of the vapour bubbles in biphasic inclusions varies from 80 to 20% of the volume.

**Methods**

Microthermometry was performed on fluid inclusions from samples FD1A and FD3A using a Linkam MDS600 heating-cooling stage at GeoRessources laboratory (Nancy, France). The following phase transition temperatures were measured when possible: final melting of the carbonic phase (T_{m \text{car}}), apparent first melting of the aqueous phase (T_{fm}), ice melting (T_{m \text{ice}}), clathrate dissociation (T_{m \text{cla}}), partial homogenisation of the carbon-dominated phase (T_{h \text{car}}) and bulk homogenisation of the entire inclusion (T_{h}). Microthermometry experiments were made between −150 °C and not above 200 °C to avoid decrepitation. The microthermometric stage was calibrated daily using natural pure CO_{2} (−56.6 °C) and synthetic pure H_{2}O fluid inclusions with known transition temperatures. Precisions of the phase transition temperature measurements are about ± 2 °C for T_{fm}, ± 0.1 °C for T_{m \text{car}}, T_{m \text{ice}}, T_{fm}, and T_{h \text{car}}, and ± 1 °C for T_{h}.

Raman spectra were acquired at room temperature using a LabRAM HR spectrometer (Horiba Jobin Yvon) equipped with an 1,800 gr.mm^{-1} grating and an edge filter at GeoRessources laboratory (Nancy, France). Excitation was provided by an Ar+ laser operating at a wavelength of 514.53 nm focussed through a ×50 objective (Olympus). The aperture of the focal hole and the slit were 500 μm and 100 μm respectively. A total of 3 acquisitions were accumulated for each spectrum and acquisition time was variable between 60 and 10 s depending on the quality of the signal and the mobility of the bubble (shorter acquisition time for moving bubble). Spectrum acquisition was systematically performed in the quartz matrix nearby the inclusion of interest at the same depth and same analytical conditions in order to distinguish between N_{2} from air and N_{2} from the inclusion. Biphasic fluid inclusions often exhibited moving gas bubbles which prevented the acquisition of the Raman spectra. Spectra of CH_{4}, H_{2}S, N_{2} and CO_{2} in the gas phase were acquired separately in two different spectral windows. The absence of H_{2} was monitored in a third spectral window. Peak positions and areas were determined using the integration tool of LabSpec software (Horiba Jobin Yvon) at 4156 cm^{-1} for H_{2}; 2917 cm^{-1} for CH_{4}; 2580 cm^{-1} for H_{2}S liquid; 2331 cm^{-1} for N_{2} and the Fermi doublet at 1285 and 1388 cm^{-1} for CO_{2} (Burke, 2001). The composition of biphasic inclusions in FD1A was acquired for 4 inclusions. For FD3A, 5 compositions were determined for biphasic inclusion and 8 compositions were determined for monophasic inclusions (Table S-2). The salinity of the fluid inclusions was calculated in single-salt NaCl-CO_{2}-CH_{4}-N_{2}-H_{2}O system using Clathrates computer package (Bakker, 1997), by using either (i) the program ICE following the equations of Duan et al., (1996) associated with T_{m \text{ice}} and T_{m \text{cla}} when clathrate dissociation occurred in presence of ice, L_{aq} and CO_{2} vap,
or (ii) Q2 program when clathrate dissociation occurred via a Q2 transition (coexistence of L_{aq}, Cla, CO_{2} liq and CO_{2} vap), associated with T_{m} cla and T_{h} car and on the equations of Thiéry et al. (1994).

**Results**

For FD1A, the observed phase transitions were T_{fm}, T_{m} ice, T_{m} cla, and T_{h} for 17 inclusions (Table S-4). The T_{fm} were difficult to observe because of the small size of the fluid inclusions and the disappearance of the vapour bubble on cooling, leading to a metastable state of the inclusions until the sudden reappearance of the vapour bubble. Seven of the measured inclusions presented the typical double bubble of CO_{2} permitting the observation of T_{m} car and T_{h} car. The average molar composition of the vapour phase in inclusions is 91.9 ± 3.4 mol % CO_{2}, 3.2 to 10.7 mol % N_{2} and < 1 mol % CH_{4}. No significant variations were observed between biphasic and triphasic inclusion. The T_{fm} were observed from −21 to −41 °C and the T_{h} from 118 to over 200 °C. The inclusions with a T_{fm} ≈ −21 °C presented the lowest T_{h}, between 118 and 187 °C, compared to the inclusions with a lower T_{fm} which generally presented a T_{h} over 200 °C. These observations point to the presence of two fluids: one where the salts present in the aqueous phase are made of pure NaCl and one where they are composed of a mixing between NaCl and another salt species. The total salinity was nonetheless calculated considering pure NaCl and varies uniformly between 0.8 and 25.4 weight % in the aqueous solution independently of the salt system. It is moreover important to note that there is no apparent correlation between salinity and volatiles species concentration.

The T_{m} ice, T_{m} cla and T_{h} were observed for 11 biphasic inclusions in FD3A (Table S-4). Vapour and sometimes solid phases in monophasic inclusions appeared at low temperatures (< −120 °C; n = 14). Biphasic fluid inclusion vapours in sample FD3A contained CH_{4} from 9.8 up to 50.4 mole percent (Table S-2). H_{2}S was systematically present from 0.2 to 1.1 mole percent. N_{2} could not be distinguished from atmospheric N_{2} for half of the inclusions and reached a maximum of 2.4 mole percent. T_{fm} was difficult to observe in this sample and we assumed pure NaCl to calculate the salinity. It is on average 3.3 ± 1.9 weight percent. The T_{h} were higher than 174 and generally over 200 °C. Monophasic gaseous liquid inclusions are mostly composed of CH_{4} (79.5 to 98.9 mole percent with one outlier at 26.2 mole percent), with minor amounts of H_{2}S of 0.8 ± 0.3 mole percent. N_{2} is only present as a trace > 0.6 mol %. The CO_{2} content varied between 4.8 and 19.5 mole percent with an outlier at 73.2 mole percent.

**Discussion and interpretation**

The dominant orientation of fluid inclusions along parallel pseudo-secondary trails in FD1A sample suggests a strong tectonic influence coherent with the long-term pulsed tectonomagmatic event that affected the Imandra-Varzuga rift zone (Chashchin et al., 2008). However, the tectonically influenced-fluid inclusion planes rarely cross the grains boundaries. So, these fluid inclusions are representative of the fluid, including primary noble-gas composition, present
during quartz growth. Our observations attest the preservation of two fluids within the sample. The first one characterised by pure NaCl in the aqueous phase could be assimilated to a seawater-derived fluid. The second one, where the aqueous phase contains a mixture of NaCl and another salt species, could point to a magmatic signature (Heinrich, 2005) and the analysed fluid inclusions represent a mixture between high-salinity high-temperature magmatic fluids and the low-salinity low-temperature seawater. This is in accordance with the xenon isotopic composition, which indicate a mixing between an atmospheric endmember, derived from the seawater-fluid, and a minor mantle contribution, derived from the magmatic fluid.

Sample FD3A presents heterogeneous entrapment with the coexistence of monophasic and biphasic inclusions with a continuum of size of the vapour bubbles, from 20 to 90 % of the total volume. This observation linked with the co-variation of CH$_4$/CO$_2$ content, the monophasic liquid inclusions being enriched in the most volatiles species, are solids arguments for boiling and entrapment below the solvus within the H$_2$O-CH$_4$-CO$_2$-H$_2$S-N$_2$ system of a single fluid (Diamond, 2003). Contemporaneous recrystallisation induced the loss of some primary and pseudo-secondary fluid inclusions (Kerrich, 1976). No subsequent fluid circulation was noticed by secondary inclusions. The bulk noble gases composition issued from these fluid inclusions thus likely to reflect the composition of the late Palaeoproterozoic as this sample appears to have precipitated from one unique fluid.

**Fissiogenic and mantle contribution**

**Fissiogenic contribution**

Production of heavy Xe isotopes can be attributed to the spontaneous fission of $^{238}$U (half-life of 4.47 Ga or $^{244}$Pu (half-life of 82 Ma; Ozima and Podosek, 2002). We calculated the theoretical production of the isotopes: $^{131}$, $^{132}$, $^{134}$Xe normalised to $^{136}$Xe by fission of $^{238}$U and $^{244}$Pu (rate of production from Alexander et al., 1971; Ragettli et al., 1994). The Xe isotope excess was calculated as Equation S-1:

$$
\left( \frac{i\text{Xe}}{^{136}\text{Xe}} \right)_\text{excess} = \left( \frac{^{i\text{Xe}}}{^{136}\text{Xe}} \right)_\text{sample} - \left( \frac{^{i\text{Xe}}}{^{136}\text{Xe}} \right)_\text{atm}
$$

(Eq. S-1)

Associated uncertainties are as Equation S-2:

$$
\sigma \left( \frac{i\text{Xe}}{^{136}\text{Xe}} \right)_\text{sample} = \left( \frac{^{i\text{Xe}}}{^{136}\text{Xe}} \right)_\text{sample} \times \left[ \left( \frac{\sigma \left( ^{i\text{Xe}}/^{136}\text{Xe} \right)_\text{sample}}{^{i\text{Xe}}/^{136}\text{Xe}_\text{sample}} \right)^2 + \left( \frac{\sigma \left( ^{136}\text{Xe}/^{130}\text{Xe} \right)_\text{sample}}{^{136}\text{Xe}/^{130}\text{Xe}_\text{sample}} \right)^2 \right]^{1/2}
$$

(Eq. S-2)

By plotting $^{238}$U and $^{244}$Pu fission spectrums in perspective with sample ratios, a $^{238}$U origins of the heavy isotopes can be suggested for all our samples (Fig. 1; Fig. S-2)
Mantle Contribution

The excess of $^{129}$Xe is thought to show a binary mixing between Archean atmosphere and mantle-derived xenon in our samples such as Equation S-3:

$$
\left( \frac{^{129}}{^{130}} \right)_{\text{Sample}} = \alpha \left( \frac{^{129}}{^{130}} \right)_{\text{Mantle}} + (1 - \alpha) \times \left( \frac{^{129}}{^{130}} \right)_{\text{Archean atmosphere}}
$$

(Eq. S-3)

with $\alpha$ as the fraction of the mantellic $^{130}$Xe. The value used for $(129/130)_{\text{Mantle}}$ is dependent on whether significant regassing of atmospheric Xe into the deep Earth is assumed to have occurred. Without regassing, we took the value $(129/130)_{\text{Mantle}} = 14 \pm 1$ (Marty et al., 2019). Considering regassing, we assume a MORB-type composition of $(129/130)_{\text{Mantle}} = 6.92 \pm 0.07$ (Pető et al., 2013), that is the lowest estimation in the literature (Holland and Ballentine, 2006; Mukhopadhyay, 2012; Parai and Mukhopadhyay, 2021). The calculated mantellic $^{130}$Xe contribution is thus an upper limit. From $\alpha$ we calculated the associated $(128/130)$ Expected for FD1A assuming a $(128/130)_{\text{Mantle}} = 0.51 \pm 0.01$ (primordial Xe, Ozima and Podosek, 2002). Results are available in Table S-1. Despite the heavy isotope discrepancies between the two bottles of FD1A, the excess of $^{129}$Xe is identical for the two bottles (Table S-3) so we considered an equal mantellic contribution. The difference between the excess of $^{131}, ^{132}, ^{134}, ^{136}$Xe is then due to a difference of crustal fissiogenic $^{238}$U contribution.

There is potentially a weak positive correlation among individual data in Figure S-4 with all our measurements parallel to the Rayleigh fractionation trend. This imply a similar contribution from mantle gas to the hydrothermal fluids and it does not appear realistic for samples from different locations. Within the uncertainties, the youngest sample from the Ongeluk formation is atmospheric. The excesses of $^{129}$Xe in the samples from the Kola Craton (Polisarka and Seidorechka formations) are consistent with the same mantle contribution to the hydrothermal fluids in this location. However, the oldest sample from Seidorechka formation presents higher $^{128}, ^{129}$Xe compare to the modern-air and to the Polisarka sample. This stands for a slightly fractionated atmosphere at the time that disappeared within the $\approx 7$ million years separating the formation of the 2 samples. Without $^{129}$Xe excesses (this study) or deficits (Avice et al., 2017; Marty et al., 2019), the measurements are expected to fall on the Rayleigh fractionation trend through modern air (Fig. S-4).
Supplementary Tables

Table S-1  Estimation of the $^{130}$Xe mantellic contribution from two different endmembers composition. The Archean mantle $^{129}$Xe/$^{130}$Xe ratio is used to calculate the % mantle without crustal regassing; The MORB $^{129}$Xe/$^{130}$Xe ratio is used to calculate the % mantle with atmospheric regassing. 1 - Marty et al. (2019); Pepin (2003); 3 - Pető et al. (2013); 4 - Ozima and Podosek (2002).

<table>
<thead>
<tr>
<th></th>
<th>$^{129}$Xe/$^{130}$Xe (2σ)</th>
<th>$^{128}$Xe/$^{130}$Xe (2σ)</th>
<th>Without atm regassing</th>
<th>With atm regassing</th>
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<tr>
<td></td>
<td>% Mantle</td>
<td>Expected</td>
<td>% Mantle</td>
<td>Expected</td>
</tr>
<tr>
<td></td>
<td>$^{128}$Xe/$^{130}$Xe (2σ)</td>
<td></td>
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</tr>
<tr>
<td>FD1A</td>
<td>6.58 (0.04)</td>
<td>0.475 (0.003)</td>
<td>0.9 $^{+0.3}_{-0.4}$</td>
<td>0.474 (0.001)</td>
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<tr>
<td>Archean Mantle$^{1,2}$</td>
<td>14 (1)</td>
<td>0.507 (0.004)</td>
<td></td>
<td>16.8 $^{+7.1}_{-5.0}$</td>
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<tr>
<td>MORB$^3$</td>
<td>6.92 (0.07)</td>
<td></td>
<td></td>
<td>0.478 $^{+0.001}_{-0.003}$</td>
</tr>
<tr>
<td>Modern air$^4$</td>
<td>6.50</td>
<td></td>
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Table S-2  Molar percentage of volatiles species in some fluid inclusions determined by RAMAN spectrometry.

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<tr>
<th>N° inclusion</th>
<th>CO₂</th>
<th>N₂</th>
<th>% molar</th>
<th>H₂S</th>
<th>CH₄</th>
<th>Total</th>
</tr>
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<tbody>
<tr>
<td>FD1A - Biphasic</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>88.8 %</td>
<td>10.5 %</td>
<td>0.0 %</td>
<td>0.7 %</td>
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<tr>
<td>28</td>
<td>93.4 %</td>
<td>6.5 %</td>
<td>0.0 %</td>
<td>0.1 %</td>
<td>100.0 %</td>
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<tr>
<td>30</td>
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<tr>
<td>31</td>
<td>89.3 %</td>
<td>10.7 %</td>
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<td>0.0 %</td>
<td>100.0 %</td>
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<tr>
<td>FD3A - Biphasic</td>
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</tr>
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<td>21</td>
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<td>0.3 %</td>
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</tr>
<tr>
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<tr>
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<td>0.0 %</td>
<td>1.1 %</td>
<td>98.9 %</td>
<td>100.0 %</td>
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<tr>
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<td>1.4 %</td>
<td>98.6 %</td>
<td>100.0 %</td>
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<td>3</td>
<td>73.2 %</td>
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<td>0.4 %</td>
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<tr>
<td>5</td>
<td>4.8 %</td>
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<td>0.8 %</td>
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<td>8</td>
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<td>14</td>
<td>19.5 %</td>
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<td>15</td>
<td>15.9 %</td>
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<td>0.7 %</td>
<td>82.9 %</td>
<td>100.0 %</td>
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</tbody>
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Table S-3  Isotopic ratio of the noble gases of each replicate. Values in red are outliers according to the Chauvenet criterion (Anscombe, 1960) and were not taken into account in the calculation of the weighted average.

Table S-4  Observed phase transitions for FD1A biphasic fluid inclusion; FD3A biphasic and monophasic fluid inclusion and calculated NaCl weight percent associated. Value n. o. corresponds to a non-observed phase change.

Tables S-3 and S-4 can be downloaded (Excel) from the online version of the article at https://doi.org/10.7185/geochemlet.2207.

Supplementary Figures

Figure S-1  (a) Thin section photographs highlighting quartz grain boundaries (solid lines) and trails of pseudosecondary fluid inclusions (dashed lines) in FD1A. (b) Coexisting monophasic and biphasic fluid inclusions in FD3A. The red square shows a biphasic inclusion with a small rim of liquid. (c) Heterogeneous entrapment in a fluid inclusion assemblage showing the coexistence of monophasic and biphasic inclusions with variable % of vapour in FD3A characteristic of boiling process. The grain on the left is devoid of fluid inclusions as a result of recrystallisation.
**Figure S-2** Isotopic spectra of Xe released from fluid inclusions in samples from the Ongeluk fm. Points represent the weighted average of 9 crushes of different quartz samples from the Ongeluk fm with error at 1σ. The grey line corresponds to the isotopic trend of the $^{124, 126, 128, 130}$Xe showing no mass-dependant fractionation. Error envelope is at 2σ. The mantellic contribution is calculated considering a starting atmospheric composition as no isotopic fractionation is observed. The associated fission spectrum shows the major influence of $^{238}$U spontaneous fission with uncertainties at 1σ.

**Figure S-3** Records of atmospheric oxygen (O$_2$, blue; Lyons et al., 2014), mass-independent fractionation of sulfur isotopes (MIF-S, orange; data from Killingsworth et al., 2019) and mass dependent fractionation of xenon isotopes over the GOE period (previous data from Avice et al., 2018; Almayrac et al., 2021), errors are at 2σ.
Figure S-4  Isotopic ratios $^{129}\text{Xe}/^{130}\text{Xe}$ vs. $^{128}\text{Xe}/^{130}\text{Xe}$ in this study. The black (a) and grey (b) dashed lines show a mixing between a non-regassed mantle and: (a) the modern air (Ozima and Podosek, 2002); (b) the Archean atmospheric ratio calculated from FD1A. Mantle $^{129}\text{Xe}/^{130}\text{Xe}$ composition comes from Marty et al. (2019) and AVCC-like $^{128}\text{Xe}/^{130}\text{Xe}$ (Pepin, 2003). The blue dashed line shows a Rayleigh fractionation trend through modern air. Uncertainties at 2σ.
Supplementary Information References


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