Supplementary Material

Experimental

Most data are presented here were previously published in the literature or from new analysis of some of the natural gases (*Table 1*). In the case of the Icelandic DICE samples for which no gas composition was available, we analyzed gases extracted from aliquots of DICE11 and AO12, another glass sampled at the same site, by vacuum crushing, manometric measurement, and mass spectrometry. Between 0.5 and 1 g of fresh glass devoid of alteration features and washed in acetone were loaded in crushing tubes and baked overnight at 100°C. The cm-sized glass fragments were crushed by activating with external solenoids an iron piston (Marty and Zimmermann, 1999). The total pressure of extracted gas was measured using a Baratron@ in a calibrated volume. The gas was then purified and the He amount and $^{3}He/^{4}He$ ratio were measured. The analysis of a gas aliquot by gas chromatography showed CO_2 to be the major species (>90 %). Results are given in Table S1. For the DICE11sample, the amount of ^{4}He (0.5-1.3 x 10^{-10} mol/g for different aliquots) is comparable to the total amount of ^{4}He recovered from sample DICE10 (1.2±0.3 x 10^{-10} mol/g) by Mukhopadhyay (2012), the differences being probably due to vesicle density heterogeneities. Likewise, the measured ^{3}He/^{4}He ratios of DICE11 (17.90±0.35 Ra) and A012 (17.96±0.58 Ra), and the $CO_2/^{3}He$ ratios of the two samples (2.86±0.28 x 10^{9} and 3.17±0.23 x 10^{9}, respectively) are similar even if the amount of gas in AO12 is a factor of 2 higher than in DICE11. This strongly suggests that gases in the respective samples originate from a common magma source. Since we are interested in abundance ratios, heterogeneities in the total gas amounts should not impact our conclusions. The nitrogen content, which could not be analyzed at CRPG at present, was estimated from the Ar-N_2 analysis done previously by Marty and

Dauphas (2003). Repeated analysis of 5 aliquots showed an excellent homogeneity of the $N_2/^{40}Ar^*$ ratio (72.5±4.4; 1 STD) (Marty and Dauphas, 2003). We use the $^4He/^{40}Ar^*$ ratio of 2.26±0.41 determined for DICE10 (Mukhopadhyay, 2012) as representative of the AO12 and DICE samples to reconstruct the gas compositions given in *Table 1*.

Computing the C/N and ^{36}Ar/N of the BSE

In order to compute the C/N and ^{36}Ar/N of the BSE, we carried out Monte Carlo simulations (10^7 iterations) based on the C, N and ^{36}Ar elemental concentrations of the surface and mantle reservoirs summarized in Table 4. Note that all concentrations used here are normalized to the mass of the silicate Earth (4.10^{27} g) for comparison propose.

Basically, at each iteration step of the simulation, the concentrations of carbon ([C]) and nitrogen ([N]) of the surface reservoir are generated randomly from normal distributions corresponding to 1.56 ± 0.06 (ppmw) and 27 ± 5 (ppmw), respectively. Note that negative concentrations are forbidden. The C/N of the surface is hence derived by computing the ratio of [C] and [N]. The ^{36}Ar/N of the surface reservoir is then generated randomly from a normal distribution corresponding to $(1.24\pm0.05).10^{-5}$, therefore allowing the ^{36}Ar concentration [^{36}Ar] of the surface reservoir to be computed as [^{36}Ar] = $^{36}Ar/N$ * [N].

Regarding the mantle, the [N] is generated randomly from normal distributions corresponding to 1.10 ± 0.55 (ppmw). In order to compute [C] and [^{36}Ar], we need to compute the C/N and ^{36}Ar/N of the mantle, which can be defined following three different approaches.

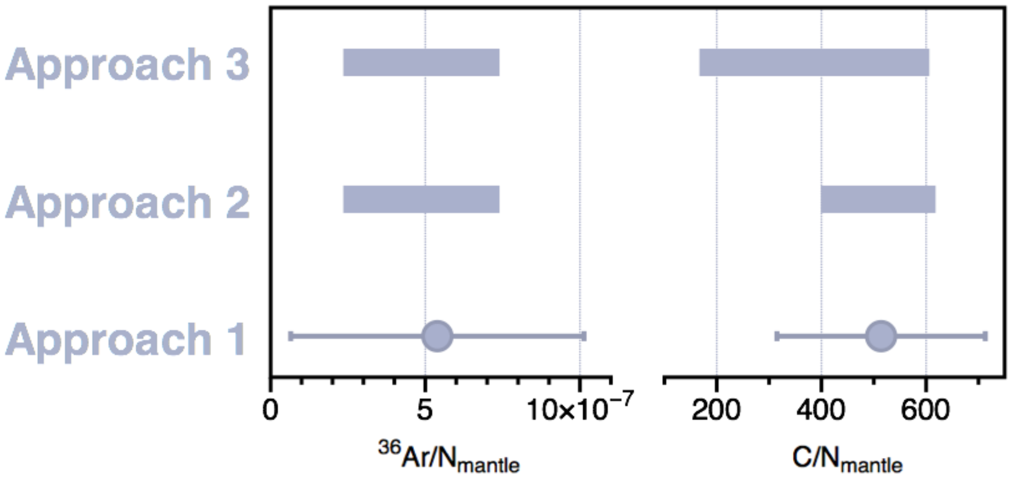

Fig. S1: $^{36}Ar/N$ and C/N values used for Monte Carlo simulations in our three approaches. Approach 1 only considers the magmatic gas data and uses means and standard deviations to compute $^{36}Ar/N$ and C/N. *Approach 2* only considers the magmatic gas data and uses interquartile ranges (IQR). *Approach 3* considers both MORB and magmatic gas data and also uses interquartile ranges (IQR).

First of all, C/N derived from the analysis of magmatic gas yield consistent values that defined a mean composition of 514 ± 199 (*Approach 1; Fig. S1*). The median (474) and interquartile range (IQR=Q3-Q1=618-399=219) corresponding to C/N values from magmatic gas (approach 2; Fig. S1) are broadly consistent with the mean value (Fig. S1). Another way of estimating the C/N of the mantle is to take into account all available values in the literature (including MORB and magmatic gas samples). Given the disparities between C/N values from MORB and magmatic gas samples, the mean C/N of the mantle derived by this method would be 532±599, which cannot be used here for the purpose of our Monte Carlo approach. The corresponding median value is 352, with an IQR of Q3-Q1=606-167=439 (*Approach 3*; *Fig. S1*). Hence, for approach 1, the C/N values of the mantle were generated randomly from normal distributions corresponding to 514 ± 199 (negative values also prohibited). For approaches 2 and 3, the C/N values of the mantle were generated randomly from uniform distributions within the ranges 399-618 and 167-606, respectively. The C concentrations of the mantle were then computed as [C] = C/N * [N]. The ^{36}Ar/N values of the mantle were only computed from magmatic gas data, defining a mean value of $(5.39 \pm 4.74).10^{-7}$ (<u>approach 1</u>), and median and IQR of $2.90.10^{-7}$ and $Q3\text{-}Q1=7.40.10^{-7}-2.35.10^{-7}=5.05.10^{-7}$, respectively (<u>approach 2</u>). For the sake of consistency, ^{36}Ar/N values were also generated randomly from a uniform distribution within the range $2.35.10^{-7}$-$7.40.10^{-7}$ for approach 3. The ^{36}Ar concentration of the mantle was then computed as [^{36}Ar] = ^{36}*Ar/N* * [N].

Lastly, the concentration of C in the BSE was computed by summing [C] of the surface and of the mantle. The concentration of C in the BSE was computed the same way. The C/N of the BSE was finally computed by as $C/N = [C]_{BSE}/[N]_{BSE}$.

<u>Approach 1:</u>

C/N_{BSE}: mean = 218 ± 101

median = 210, 25th percentile (Q1) = 143, 75th percentile (Q3) = 283.

$^{36}Ar/N_{BSE}$: mean = $7.73.10^{-6}$

median = $7.51.10^{-6}$, Q1 = $6.65.10^{-6}$, Q3 = $8.59.10^{-6}$.

$[C]_{mantle}$ (ppmw): median = 453, Q1 = 269, Q3 = 679.

$[Ar]_{mantle}$ (ppmw) : median = $1.54.10^{-6}$, Q1 = $7.44.10^{-7}$, Q3 = $2.66.10^{-6}$.

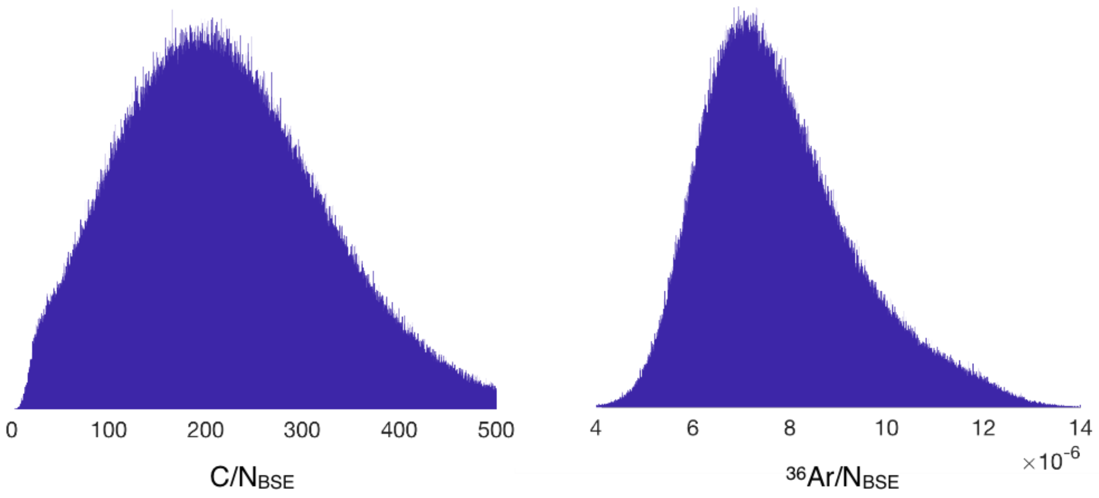

Fig. S2: Probability distributions of the C/N_{BSE} and $^{36}Ar/N_{BSE}$ obtained from approach 1.

<u>Approach 2:</u>

C/N_{BSE} : mean = 214

median = 219, 25th percentile (Q1) = 174, 75th percentile (Q3) = 259

$^{36}Ar/N_{BSE}$: mean = $7.66.10^{-6}$

median = $7.43.10^{-6}$, Q1 = $6.57.10^{-6}$, Q3 = $8.52.10^{-6}$

$[C]_{mantle}$ (ppmw): median = 479, Q1 = 323, Q3 = 646.

$[Ar]_{mantle}$ (ppmw) : median = $1.29.10^{-6}$, Q1 = $8.24.10^{-7}$, Q3 = $1.89.10^{-6}$.

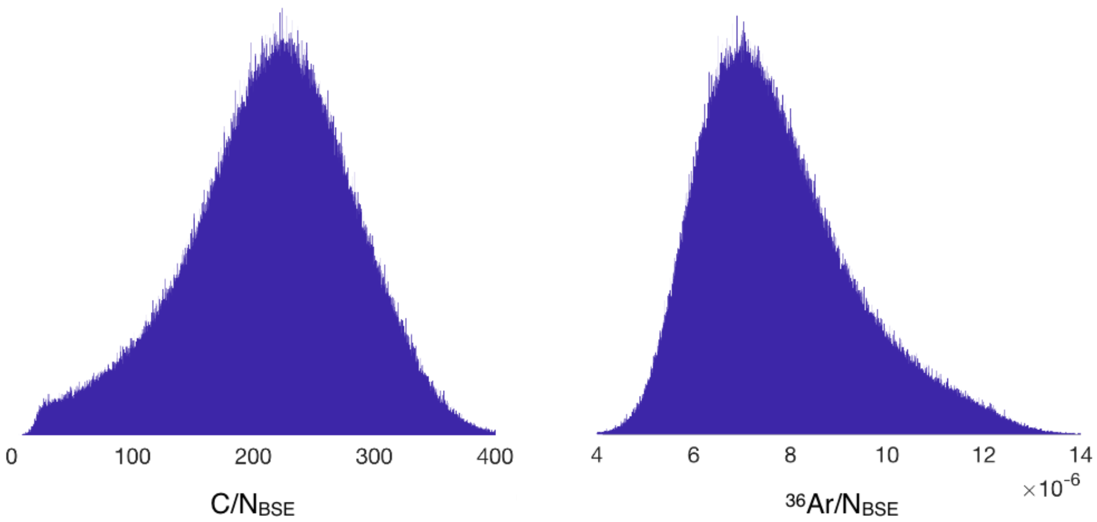

Fig. S3: Probability distributions of the C/N_{BSE} and $^{36}Ar/N_{BSE}$ obtained from approach 2.

<u>Approach 3:</u>

C/N_{BSE} : mean = 166

median = 158, 25th percentile (Q1) = 112, 75th percentile (Q3) = 215

$^{36}Ar/N_{BSE}$: mean = $7.66.10^{-6}$

median = $7.43.10^{-6}$, Q1 = $6.57.10^{-6}$, Q3 = $8.52.10^{-6}$

$[C]_{mantle}$ (ppmw): median = 337, Q1 = 211, Q3 = 504.

$[Ar]_{mantle}$ (ppmw) : median = $1.29.10^{-6}$, Q1 = $8.24.10^{-7}$, Q3 = $1.89.10^{-6}$.

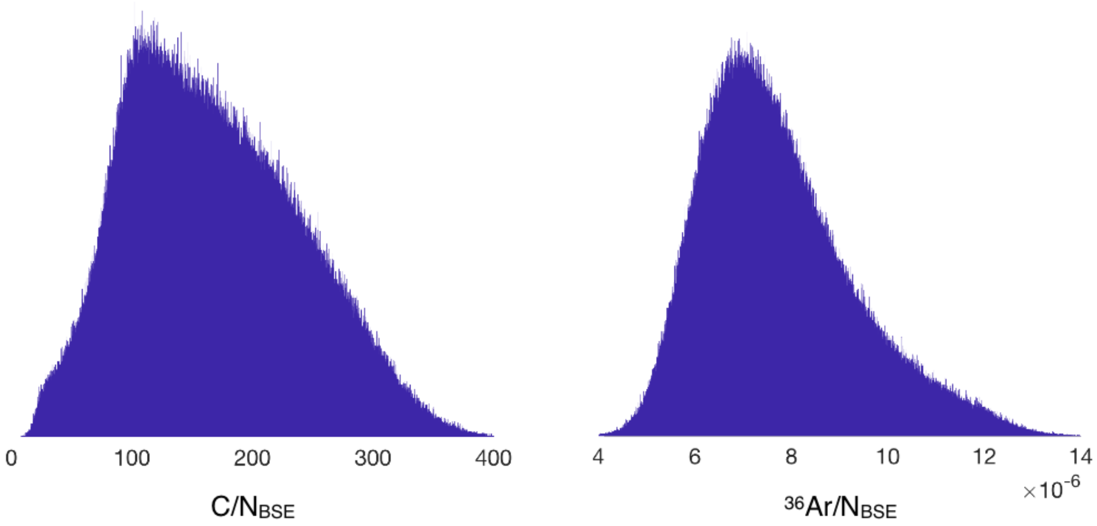

Fig. S4: Probability distributions of the C/N$_{BSE}$ and ^{36}Ar/N$_{BSE}$ obtained from approach 3.

Supplementary Table S1: Compilation of meteorite C, N and ^{36}Ar data (attached).

Supplementary Figure S5 and discussion of meteorite data

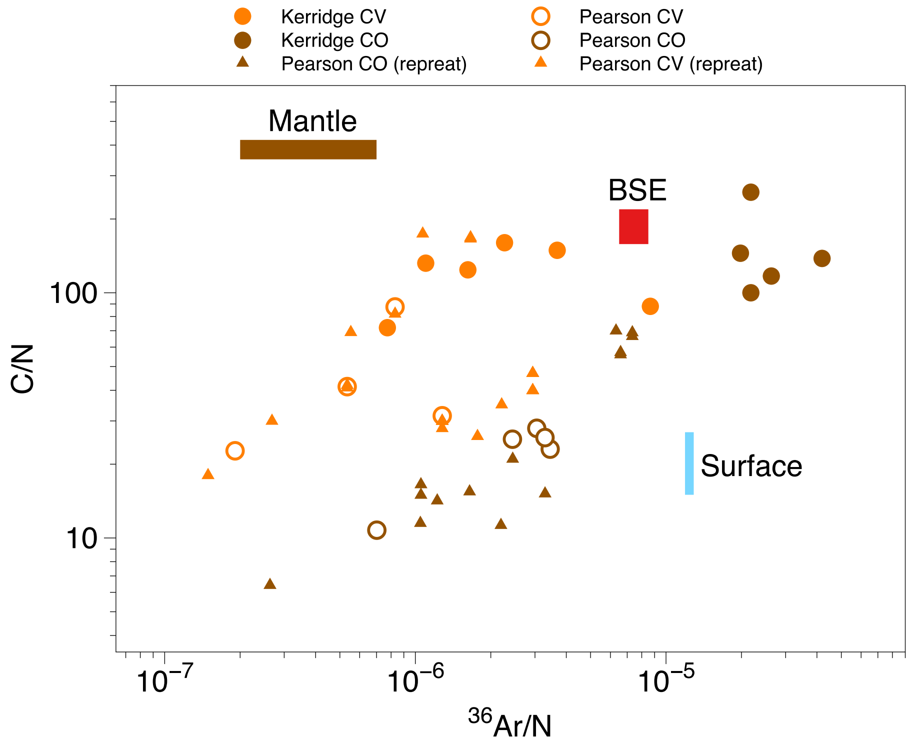

Fig. S5. Nitrogen concentration data for CO and CV chondrites is scant in the literature. In the main text we use the values given by Kerridge, (1985), however we note that Pearson et al., (2006) also analyzed N within CO and CV chondrites. In this figure we plot the bulk C/N and ^{36}Ar/N for all CO and CV chondrites measured in these studies. The mean C/N and ^{36}Ar/N values for Pearson et al., (2006) are consistently lower than those calculated from the C and N concentrations given by Kerridge, (1985). One possibility for the discrepancy between the two studies is that the mass of sample analyzed by Pearson et al., (2006) is 1-2 orders of magnitude lower than that used by Kerridge, (1985). This results in large uncertainties (up to 100%) on the N concentration within the CO and CV chondrites measured by Pearson et al., (2006). Another possibility that may explain the difference between these studies is that Kerridge, (1985) pre-heated the samples to 200°C prior to analysis. This may have reduced atmospheric contamination but could have also resulted in the preferential release of N relative to C and ^{36}Ar. We note however, that the C/N and ^{36}Ar/N of CM chondrites as calculated from Kerridge, (1985) is consistent with that calculated by Pearson et al., (2006), Alexander et al., (2012) and Alexander et al., (2013), who did not pre-heat their samples (Supplementary Table 1). We therefore do not consider the preferential loss of N during preheating to be a significant factor in explaining the difference between these studies. Finally, we highlight that whilst the mean data from Pearson et al., (2006) is consistently lower than that calculated from Kerridge, (1985), the individual analyses span a broader range that approach the values of Kerrdige, (1985). The small sample sizes used by Pearson et al., (2006) may therefore reflect the sample heterogeneity and we therefore consider the data from Kerridge, (1985) to be better representative of the bulk chondrites (Fig. 5, main text).

Sources of data presented in the meteorite compilation (*Table S1*)

The names in the Table are those of the first author and the two following numbers refer to the year of publication.

Alexander, C.M.O., Bowden, R., Fogel, M.L., Howard, K.T., Herd, C.D.K., Nittler, L.R., 2012. The provenances of asteroids, and their contributions to the volatile inventories of the terrestrial planets. Science 337, 721–3. doi:10.1126/science.1223474

Alexander, C.M.O., Howard, K.T., Bowden, R., Fogel, M.L., 2013. The classification of CM and CR chondrites using bulk H, C and N abundances and isotopic compositions. Geochim. Cosmochim. Acta 123, 244–260. doi:10.1016/j.gca.2013.05.019

Bischoff, A., Palme, H., Ash, R.D., Clayton, R.N., Schultz, L., Herpers, U., Stöffler, D., Grady, M.M., Pillinger, C.T., Spettel, B., Weber, H., Grund, T., Endreß, M., Weber, D., 1993. Paired Renazzo-type (CR) carbonaceous chondrites from the Sahara. Geochim. Cosmochim. Acta 57, 1587–1603. doi:10.1016/0016-7037(93)90014-N

Bogard, D.D., Clark, R.S., Keith, J.E., Reynolds, M.A., 1971. Noble gases and radionuclides in Lost City and other recently fallen meteorites. J. Geophys. Res. 76, 4076–4083.

doi:10.1029/jb076i017p04076

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Downes, H., Abernethy, F.A.J., Smith, C.L., Ross, A.J., Verchovsky, A.B., Grady, M.M., Jenniskens, P., Shaddad, M.H., 2015. Isotopic composition of carbon and nitrogen in ureilitic fragments of the Almahata Sitta meteorite. Meteorit. Planet. Sci. 50, 255–272. doi:10.1111/maps.12413

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Grady, M.M., Verchovsky, A.B., Franchi, I.A., Wright, I.P., Pillinger, C.T., 2002. Light element geochemistry of the Tagish Lake CI2 chondrite : Comparison with CI1 and CM2 meteorites. Meteorit. Planet. Sci. 37, 713–735.

Grady, M.M., Wright, I.P., Carr, L.P., Pillinger, C.T., 1986. Compositional differences in enstatite chondrites based on carbon and nitrogen stable isotope measurements. Geochim. Cosmochim. Acta 50, 2799–2813.

Grady, M.M., Wright, I.P., Swart, P.K., Rllinger, C.T., 1985. The carbon and nitrogen isotopic composition of ureilites : implications for their genesis 49.

Kerridge, F., 1985. Carbon, hydrogen and nitrogen in carbonaceous chondrites: abundances and isotopic compositions in bulk samples. Geochim. Cosmochim. Acta 49, 1707–1714.

Mazor, E., Heymann, D., Anders, E., 1970. Noble gases in carbonaceous chondrites. Geochim. Cosmochim. Acta 34, 781–824.

Murty, S.V.S., Mahajan, R.R., Jenniskens, P., Shaddad, M.H., Eldien, B., 2010. Noble gases and nitrogen in the Almahata Sitta ureilite. Meteorit. Planet. Sci. 45, 1751–1764. doi:10.1111/j.1945-5100.2010.01095.x

Nakamura, T., Noguchi, T., Zolensky, M.E., Tanaka, M., 2003. Mineralogy and noble-gas signatures of the carbonate-rich lithology of the Tagish Lake carbonaceous chondrite: evidence for an accretionary breccia. Earth Planet. Sci. Lett. 207, 83–101. doi:10.1016/S0012-821X(02)01127-5

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Ott U., Loehr H.P. and Begemann F. (1985) Trapped noble gases in 5 more ureilites and the possible role of Q. Lunar Planet. Sci. **16**, 639-640.

Patzer, A., Schultz, L., 2001. Noble gases in enstatite chondrites I: Exposure ages, pairing, and weathering effects. Meteorit. Planet. Sci. 36, 947–961. doi:10.1111/j.1945-5100.2001.tb01932.x

Pearson, V.K., Sephton, M.A., Franchi, I.A., Gibson, J.M., Gilmour, I., 2006. Carbon and nitrogen in carbonaceous chondrites: Elemental abundances and stable isotopic compositions. Meteorit. Planet. Sci. 41, 1899–191