NWA 8114 Data Reduction

These notes refer to reduction of xenon isotope data for irradiated NWA 8114 samples.

Data are reduced following the steps listed below. Data are reduced in the spreadsheets listed below, and those spreadsheet are available on Figshare, doi: 10.48420/14229269

1. Irradiated Samples

Two irradiates samples of NWA 8114 were analysed:

- NWA_8114_0.12, mass 0.12 mg,
- NWA_8114_0.23, mass 0.23 mg.

Samples were included in irradiation MN14a.

Samples were analysed in a single campaign, between 10/03/2015 and 29/04/2015.

All files are in nq20 directories.

A sample of Shallowater, the irradiation monitor, and two samples for a different meteorite were loaded in the mass spectrometer laser port alongside these samples, and were analysed at the same time. Data for that other meteorite are not included in any of this work, i.e files nq20_1\File1204 to nq20_1\File1309. In this case, air cals that were run in conjunction with those samples are not included here, i.e. files nq20_ac1\File1169 to nq20_ac1\File1217. Analysis of the Shallowater sample, and reduction of that data are discussed below.

Samples were heated using a CW Nd:YAG laser (λ = 1064 nm).

All files for Air Cals, Blanks and Samples were reduced by SAC using **PulsespecXenon_14.xls**.

Data are reduced in the spreadsheet **NWA8114_Xe_Irradiated_Final.xlsm**. Raw data are listed in worksheets labelled with the appropriate directory names. The calculations for air cals, blanks and each individual samples are set out on separate worksheets labelled accordingly.

Similar to the unirradiated samples, any air cals and blanks noted in lab book as having problems were excluded – e.g. low air cals because mass spec not tuned up properly, problems with triggering, blanks noted as high or as having high 128 hydrocarbon peak, leaks, build up tests, etc. On further examination of the data, some more air cals and blanks were excluded as outliers, often with high ^{128}Xe peaks due either to the 128 hydrocarbon, or having been run immediately after a sample had released a large amount of gas. The **FileSummary** worksheet lists all data files and indicates which were used and which were excluded, with reasons for excluding individual files.

1.1 The summed peak areas, from the first 5 summed spectra (first 50 s) were used in the following calculation.

1.2 From these peak areas, uncorrected ratios relative to ^{132}Xe were calculated for the air cals. The mean ratios of all the separate analyses was also calculated.

1.3 Air cals and blanks were (normally) acquired on the 5mV sensitivity scale, but sample analyses used a range of different scales according to how much gas was extracted from the samples. Therefore the peak areas of all analyses were multiplied by the respective sensitivity scale, so that all analyses could be directly compared regardless of the sensitivity scale used during acquisition.

1.4 The mean ^{132}Xe peak area (as the reference isotope) of the air cals was calculated.

1.5 The numbers of atoms of each isotope were calculated, assuming that an air cal contained 1.2×10^5 atoms ^{132}Xe and had the isotopic composition of atmospheric xenon.

1.5.1 The numbers of atoms in the air cals were calculated simply by multiplying the uncorrected ratios to ^{132}Xe by the number of ^{132}Xe atoms.

1.5.2 The numbers of atoms in blank and sample analyses were calculated by reference to the mean air cal, using the following equation for each isotope, *i*

$$Atoms\ of\ \ i = \frac{peak\ area\ of\ i}{air\ cal\ mean\ ^{132}Xe\ peak\ area} \times 1.2 \times 10^5$$

1.6 The mean number of atoms of each isotope was calculated for the blank analyses, along with the standard deviation of the mean.

1.7 Blank corrected numbers of atoms of each isotope were calculated for the air cals and samples, by subtracting the mean blank values calculated in Step 1.6 from the numbers of atoms calculated in Step 1.5.

1.8 Blank corrected ratios relative to ^{132}Xe were calculated for the air cals. The mean of all the separate analyses was also calculated for each isotope, along with the standard deviation and the standard error of the mean $\left(= standard\ deviation/\sqrt{number\ of\ analyses}\right)$. When the standard deviation of the mean is comparable to (or smaller than) the average error of the analyses contributing to the mean, the standard error can be used as the uncertainty of the mean ratios.

1.9 Delta values were calculated for the mean uncorrected and blank corrected ratios for each isotope *i* of the air cals, using

$$\delta_i = \left(\frac{\left(\frac{^{i}Xe}{^{132}Xe}\right)_{Measured}}{\left(\frac{^{i}Xe}{^{132}Xe}\right)_{Air}} - 1 \right) \times 1000$$

Comparison of the uncorrected and blank corrected ratios illustrated that the blank correction was necessary.

1.10 The ratio of "actual" air to the measured values was also calculated – this is used to apply a correction for instrument dependent mass fractionation to the sample analyses.

1.10.1 A value of 1 is adopted for the ratio of "actual" to measured air for ^{124}Xe, ^{126}Xe and ^{128}Xe. A hydrocarbon interferes with ^{128}Xe, and the $^{128}Xe/^{132}Xe$ ratios can be sensitive to the correction.

1.11 Air corrected numbers of atoms of each isotope were calculated for the samples, by multiplying the blank corrected numbers of atoms from Step 1.7 by the correction factor determined in Step 1.10.

1.12 Corrected ratios to ^{132}Xe and ^{128}Xe were calculated for the sample analyses from the air corrected number of atoms. If ratios to other isotopes are required they should also be calculated at this point from the corrected numbers of atoms.

1.12.1 In some heating steps the number of ^{132}Xe atoms are small, such that after the blank correction has been applied the remaining number of atoms is within error of zero, or is even a negative value. This is due to a combination of the ^{128}Xe and ^{131}Xe peaks being up to four orders of magnitude larger than the ^{132}Xe peaks, meaning that the sensitivity scale used on the data acquisition card to accommodate the ^{128}Xe and ^{131}Xe peaks does not have the resolution to properly resolve ^{132}Xe and the other smaller isotopes, and broadening of the ^{131}Xe peaks means that the signal has not returned to the baseline by the ^{132}Xe peak.

1.13 Some analyses of samples *NWA_8114_0.23* were run using a sensitivity scale on the data acquisition card that was appropriate for the smaller isotopes (^{132}Xe, ^{134}Xe and ^{136}Xe), with the knowledge that the larger isotopes (^{128}Xe and ^{131}Xe) would be saturated in those analyses. The calculated ratios relative to ^{132}Xe for these larger isotopes are therefore meaningless in these analyses. Similarly, ratios relative to ^{128}Xe cannot be calculated for heating steps where data were acquired using an appropriate sensitivity scale for these analyses. Therefore the total number of atoms of each isotope cannot be calculated for this sample.

1.14 The volumes of gas released in some heating steps for sample *NWA_8114_0.12* were so large that only ~1/3 of the gas was admitted into the mass spectrometer for analysis. The numbers of atoms calculated for these analyses are multiplied by 3 prior to calculating the total amounts of each isotope, to give a more realistic estimate of the total number of atoms.

1.15 The calculated ratios for each sample, and the total number of atoms of each isotope for sample *NWA_8114_0.12* are in the individually labelled workbooks, **NWA8114_0.12_132** and **NWA8114_0.23_132**.

2. Shallowater

Aliquots of Shallowater are irradiated alongside the samples of interest, to monitor the conversion of ^{127}I to ^{128}Xe in the reactor. Several different suites of samples can be included in a single irradiation, along with multiple aliquots of Shallowater. The data from Shallowater samples analysed in conjunction with one suite of samples may also be used with the other suites of samples included in the same irradiation, therefore the Shallowater data were reduced separately from the samples.

One aliquot of Shallowater from irradiation MN14a was analysed in conjunction with these samples:

- Shallowater SW9, mass 0.42 mg.

This sample was analysed between 31/3/2015 and 10/04/2015.

All files are in nq20 directories.

The irradiated samples of NWA 8114, which were analysed alongside this Shallowater sample, released huge amounts of ^{128}Xe that could affect the ratios measured in air cals and blank analyses. Those air cal and blank analyses are therefore not considered in conjunction with the Shallowater analyses. Another meteorite was analysed immediately before the Shallowater sample, as part of the same analysis campaign. That sample did not release such large amounts of ^{128}Xe, therefore the air cal and blank analyses measured in conjunction with that sample can be considered alongside those analysed in conjunction with the Shallowater sample, to allow calculation of more accurate average

blank and air cal values. The data used here include files nq20_ac1\File1169 to nq20_ac1\File1217, and files nq20_1\File1204 to nq20_1\File1309 (blanks and Shallowater analyses only).

Samples were heated using a CW Nd:YAG laser (λ = 1064 nm).

All files for Air Cals, Blanks and Samples were reduced by SAC using **PulsespecXenon_14.xls**.

Data are reduced in the spreadsheet **MN14a_Shallowater9_Final_NWA8114.xlsm**. Raw data are listed in worksheets labelled with the appropriate directory names. The calculations for air cals, blanks and the Shallowater sample are set out on separate worksheets labelled accordingly.

Any air cals and blanks noted in lab book as having problems were excluded, in the same way as for the other samples – e.g. low air cals because mass spec not tuned up properly, problems with triggering, blanks noted as high or as having high 128 hydrocarbon peak, leaks, build up tests, etc. On further examination of the data, some more air cals and blanks were excluded as outliers, often with high ^{128}Xe peaks due either to the 128 hydrocarbon, or having been run immediately after a sample had released a large amount of gas. The **FileSummary** worksheet lists all data files and indicates which were used and which were excluded, with reasons for excluding individual files.

2.1 Steps 1.1 to 1.12 that were applied to the irradiated samples were also applied to this data set, except ^{129}Xe was used as the reference isotope, rather than ^{132}Xe.

2.1.1 The numbers of atoms of each isotope were calculated assuming that an air cal contained 1.2×10^5 atoms ^{132}Xe and had the isotopic composition of atmospheric xenon. Taking the atmospheric ratio of ^{129}Xe/^{132}Xe to be 0.9823, this equates to 1.18×10^5 atoms ^{129}Xe in an air cal.

2.1.2 Corrected ratios to ^{129}Xe and ^{132}Xe were calculated for the Shallowater analyses from the air corrected number of atoms.

2.2 The ^{134}Xe/^{132}Xe and ^{136}Xe/^{132}Xe ratios were used to determine whether the Shallowater analyses included a contribution from fission. Ratios higher than Q-Xe, and lying on a mixing line indicate a fission contribution which is assumed to be from neutron induced fission of ^{235}U. A fission correction is made by applying the following steps:

2.2.1 The measured ^{134}Xe/^{132}Xe and ^{136}Xe/^{132}Xe ratios are assumed to be a binary mixture of Q-Xe and fission xenon. The ^{134}Xe/^{132}Xe ratio was used in the calculations because the ^{136}Xe/^{132}Xe ratio is modified by production of additional ^{136}Xe due to neutron capture on ^{135}Xe during the irradiation.

2.2.2 The fraction of the measured ^{132}Xe that is from Q-Xe was calculated using

$$\frac{^{132}Xe_Q}{^{132}Xe_M} = \frac{\left(\frac{^{134}Xe}{^{132}Xe}\right)_F - \left(\frac{^{134}Xe}{^{132}Xe}\right)_M}{\left(\frac{^{134}Xe}{^{132}Xe}\right)_F - \left(\frac{^{134}Xe}{^{132}Xe}\right)_Q}$$

where the subscript *Q* refers to Q-Xe, *F* to the ^{235}U neutron induced fission component, and *M* to measured values.

2.2.3 Neutron induced fission of ^{235}U produces only the heavier Xe isotopes, ^{131}Xe, ^{132}Xe, ^{134}Xe and ^{136}Xe. For these isotopes a ratio of the fission corrected component of that isotope (*C*) to the measured ^{132}Xe was calculated using

$$\frac{^{i}Xe_C}{^{132}Xe_M} = \left(\frac{^{i}Xe}{^{132}Xe}\right)_M - \left(\frac{^{i}Xe}{^{132}Xe}\right)_F \left(1 - \frac{^{132}Xe_Q}{^{132}Xe_M}\right)$$

For the other isotopes which do not have a fission component, the ratio of the corrected component to the measured ^{132}Xe is simply the same as the measured ratio.

2.2.4 Fission corrected ratios to both ^{129}Xe and ^{132}Xe were then calculated from the values determined in Step 2.2.3:

$$\frac{{}^{i}Xe_C}{{}^{j}Xe_C} = \frac{\frac{{}^{i}Xe_C}{{}^{132}Xe_M}}{\frac{{}^{j}Xe_C}{{}^{132}Xe_M}}$$

2.2.5 As neutron induced fission of ^{235}U does not produce ^{124}Xe, ^{126}Xe, ^{128}Xe, ^{129}Xe or ^{130}Xe, there is actually no correction to apply to the ratios of these isotopes to ^{129}Xe and the ratios calculated in Step 2.1.2 are used. But step 2.2.4 must be performed for ratios of all other isotopes to ^{129}Xe, and for ratios of all isotopes to ^{132}Xe.

2.3 After the correction detailed in Step 2.2, the corrected ^{132}Xe/^{129}Xe ratio includes only a contribution to ^{132}Xe from Q-Xe. This is used with the known ^{128}Xe/^{132}Xe ratio for Q-Xe to subtract the ^{128}Xe Q-Xe contribution from the ^{128}Xe/^{129}Xe ratio, to determine the ^{128}Xe*/^{129}Xe ratio (where the * denotes an excess).

2.4 A correlation between ^{128}Xe*/^{129}Xe and ^{132}Xe/^{129}Xe is sought, and the ^{128}Xe*/^{129}Xe* ratio determined using a Mahalanobis fitting routine.