Correspondence between [James McKamey Guthrie [GuthrieJM@missouri.edu](mailto:GuthrieJM@missouri.edu)] and [TK [tkbhar@neo.tamu.edu](mailto:tkbhar@neo.tamu.edu)] with [J. David Robertson [robertsonjo@missouri.edu](mailto:robertsonjo@missouri.edu)]

**Wednesday, July 9 (Missouri)**

Dr. Robertson asked me to contact you regarding the sample of Pu-239 that you sent. As I understand it, this sample is a solution of ~300 ppb Pu-239 in 8 mL of 0.024M ferrous sulfamate solution. Are there any other known components (acid, base, etc.) in this solution?

Could you please confirm the elements/isotopes we are determining in the solution? I believe that you want to measure Pu-239, but am unsure what other analytes you requested.

As a standard, we are planning to use a Pu-239 standard from Eckert & Ziegler with certified activity as of a specific date. We will calculate Pu concentration from the activity as of the date of analysis, and will report your results in units of concentration (ppb).

**Thursday, August 07 (Missouri)**

Yes, I was planning to do so anyway, although now that you request it specifically, I am going to make up a separate standard series with natural U (natural ratio verified by MC-ICP-MS) to do the analysis. Any idea of the U concentration?  
  
  
Is it only 239Pu that is causing all of the sample's frisker response? Or what other radionuclides do you suspect the sample may contain? Seems a bit hot for just 2400 ng of alpha-emitting 239Pu.

**August 8, 2014 (Missouri)**

I have analyzed the sample by ICP-MS. Results will be on their way to you within a few days. In what units do you want me to report the concentrations of 235U, 238U, and 239Pu (ng/mL, ng/g, moles/mL, total U plus approx. isotope ratio)? 235U appears to be depleted.  
  
Are you interested in the minor Pu isotopes? Anything I can report about mass 241 will be approximate, since it will reflect a mixture of 241Pu and 241Am.  
  
Similarly, are you interested in the minor U isotopes? I'm seeing signals at 234, 236, and 237. Do you think this last one is 237Np?  
  
Since your solution is 0.024M ferrous sulfamate, I will assume a density of 1.006 g/mL in the calculations.

**August 8, 2014 (TK)**

Response

**August 13, 2014**

1. Standards were prepared from an Eckert & Ziegler standard solution of 239Pu which also contained assayed amounts of 238Pu, 240Pu, 241Pu, 241Am, and 242Pu. The levels of these nuclides on the day of analysis were calculated by appropriate decay-corrections. 241Pu and 241Am could not be resolved and so the result for mass 241 represents the sum of the concentrations of these nuclides.  
     
   2. A second series of standards was prepared from a High Purity Standards natural uranium solution. The isotope ratios in this solution were previously verified as natural using multicollector-ICP-MS.  
     
   3. Each standard series was prepared at approximately 0, 1, 2, and 5 ppb (in 239Pu and in 238U) to create a linear 4-point calibration curve. Standards were prepared using gravimetric dilutions so that exact concentrations could be calculated. The calibrations for 234U, 236U, and 237Np were estimated based on the instrument response for 235U or 238U.  
     
   4. The internal standard Th was added to all standards at a level of 2 ppb. Solutions of standards with no Th were also prepared to verify the absence of any signal at mass 232.  
     
   5. Duplicate aliquots of sample were weighed and diluted gravimetrically by a factor of approximately 100. The exact dilution factors were calculated and used in the results spreadsheet. To one sample dilution, no Th internal standard was added, in order to correct for the small amount of Th in the sample (measured to be 0.3 ng/mL).  
     
   6. A serial dilution of the sample was performed (dilution factor approx. 9000) in order to quantify the relatively high level of 238U.  
     
   7. All dilutions have already been accounted for in the results. All results are relative to the original solution you provided.  
     
   8. Limits of detection relative to the original sample are given in blue font below the results. These LODs are calculated based on the instrument backgrounds (three times the standard deviation of concentration of each isotope measured in multiple runs of blank solution). The 238U limit of detection is significantly higher than the others, due to the fact that it is calculated relative to the serially-diluted sample. Limits of detection for 234U, 235U, and 237Np are estimated since the signals in the blanks for these masses were zero.  
     
   9. As stated above, the 241 signal is taken as the sum of the individual signals from the 241Pu and 241Am nuclides. Since Am and Pu will have slightly different instrument response factors, the result for mass 241 is approximate.  
     
   10. The species at mass 237 is assumed to be 237Np. Since its response was estimated based on that of 238U, the concentration should be taken as approximate.  
     
   11. The rate of uranium hydride (UH) formation in the ICP plasma was very low. The signal at mass 239 in a blank was 3.1 cps; in a 5 ppb natural U solution run soon afterward, the signal was 6.7 cps (with 436,000 cps at mass 238). These signals yield a UH ratio of 8.26e-6; this constant was used to mathematically correct the signals at 236 and 239. The corrections to the sample’s signals at masses 236 and 239 were 0.02% and 0.05% of signal, respectively.  
     
   12. Results and limits of detection are given in units of (ng nuclide/mL solution). The estimated sample density of 1.006 g/mL was used in the calculation of results.  
     
   13. For the purpose of any decay corrections, the ICP-MS run occurred on 08 Aug 2014.

**Corrections from TK:**

200 μg of Pu total

Dr. Chirayath calculation involved 13 mg DUO2 dissolved into 5 ml solution

We had 12.9 ml DUO2 in 5.167 ml of solution

We took 0.5 ml (of the 5.167 solution) and diluted it to (4.5 ml of 2.67 M or 5 ml of 4 M)

Rest in AGN, and some might have been left in vials and flasks.