Lawrence Livermore National Laboratory

Characterization of Uranium Oxyfluoride Particles for Nuclear Safeguards

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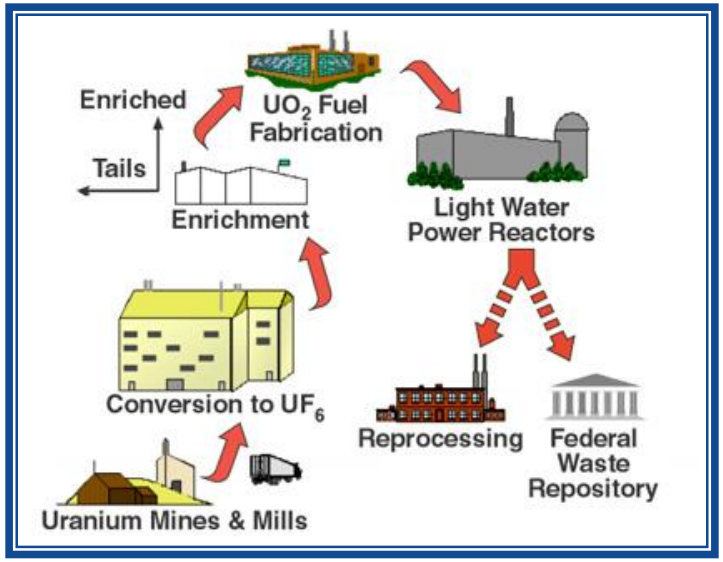
The next 10 min I will talk about…

- What is **particle analysis** for **environmental sampling (ES)**?
- What are the **current trends** in nuclear safeguards?
- The uranium oxyfluoride particle project at LLNL
  - Scope of the work
  - Analytical techniques used
  - Our latest findings
  - Conclusions

Collecting nuclear fingerprints through Environm. Sampling



Whenever nuclear material is processed, small amounts are released to the environment

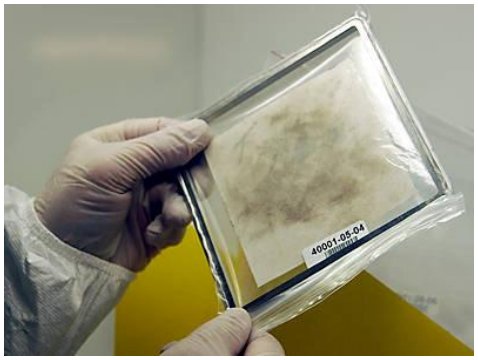
Material released = characteristic of process material

Environmental sampling uses pieces of cotton to collect this material (dust, particles) **SWIPES**

Swipes analyzed in **bulk** and on a **particle basis** at IAEA's Network of Analytical Labs (NWAL)

Uranium isotopic composition is measured

Detection of undeclared nuclear materials for States w/ AP



Swipe sample

Sent to IAEA Safeguards Analytical Lab

Swipe analyzed as a whole and on a particle-by-particle basis

The next generation of safeguards inspectors

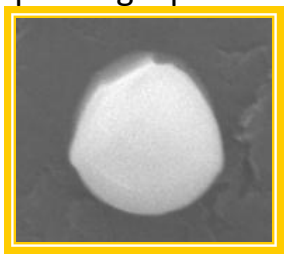
From accountants…

…who verify declarations

…to detectives

…looking at the bigger picture

1 µm single particle



crystallinity

isotopic composition

shape & size

surface structure

elemental composition

molecular information

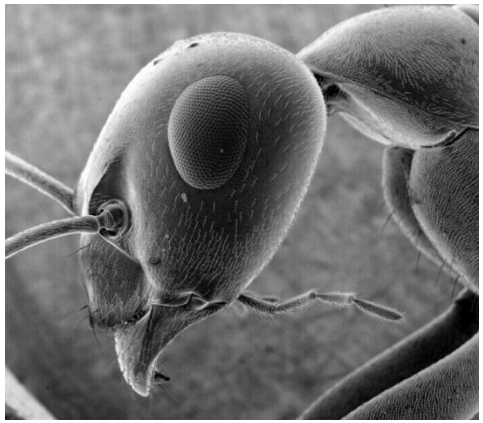
This information could help determine the particle's source and exposure history

As always, there are some challenges…

Advantage analyzing individual particles vs. swipe as a whole
no information lost through averaging

Natural U particles are everywhere
How do we find the non-natural/HEU ones?

The particles are so incredibly small
How do we analyze them?

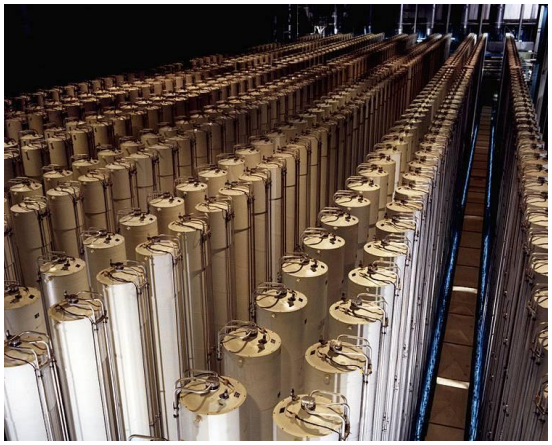


From: Dartmouth Electron Microscope Facility/Dartmouth College

300x smaller than the eye of this ant!

Particle analysis requires very sensitive equipment with ultra-high spatial resolution!

Uranium oxyfluoride particles from enrichment activities

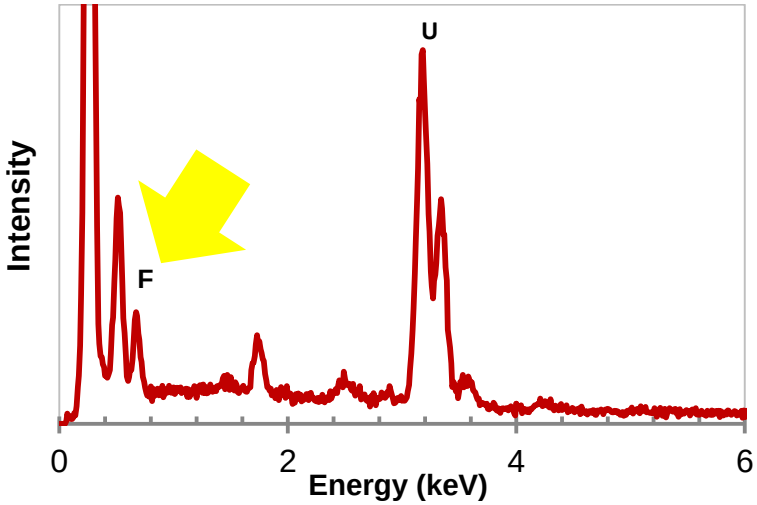


Uranium hexafluoride (UF_6) for the enrichment of uranium

- Used in large quantities
- Very reactive
- Small amounts inadvertently released to the atmosphere

UF_6 reacts with atmospheric moisture

UO_2F_2 particles



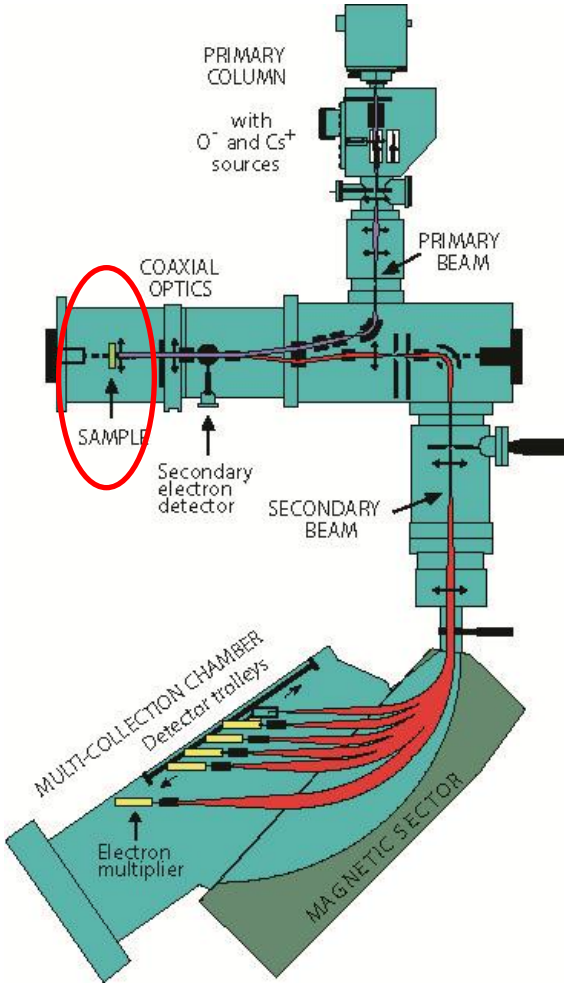
Hypothesis

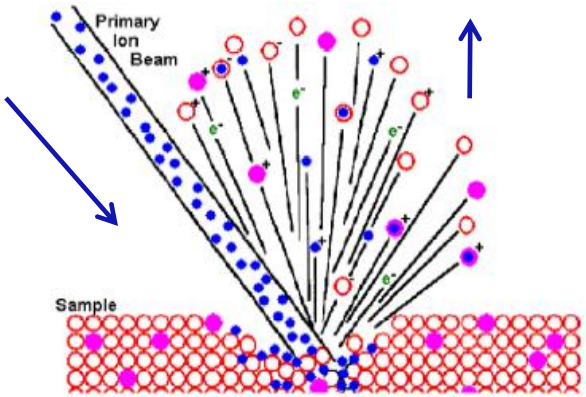
1. *If we can measure the fluorine in these particles…*
   - Indicator of enrichment activities (NPT)
2. *UO_2F_2 is known to be affected by the environment*
   - F-to-U ratio may be indicator of exposure history of collected particle

Particle analysis using NanoSIMS

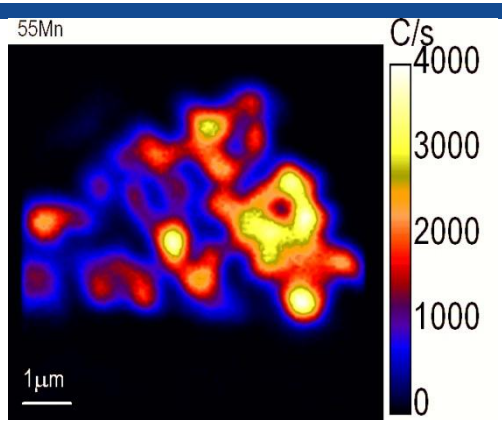
The Cameca NanoSIMS 50 at LLNL

Secondary ion mass spectrometry at ultrahigh spatial resolution

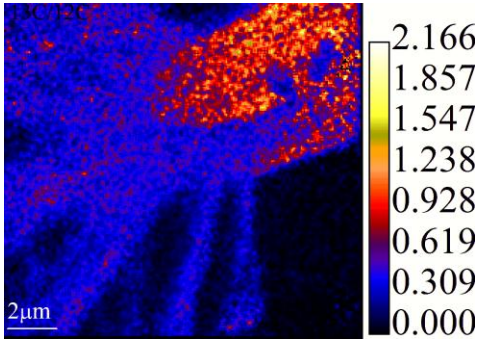




- Surface technique
- Surface sputtered with high energy ion beam
- NanoSIMS spatial resolution down to the nanometer scale
- Ion microscope
- Produces elemental maps
- LLNL NanoSIMS only NanoSIMS used for nuclear applications



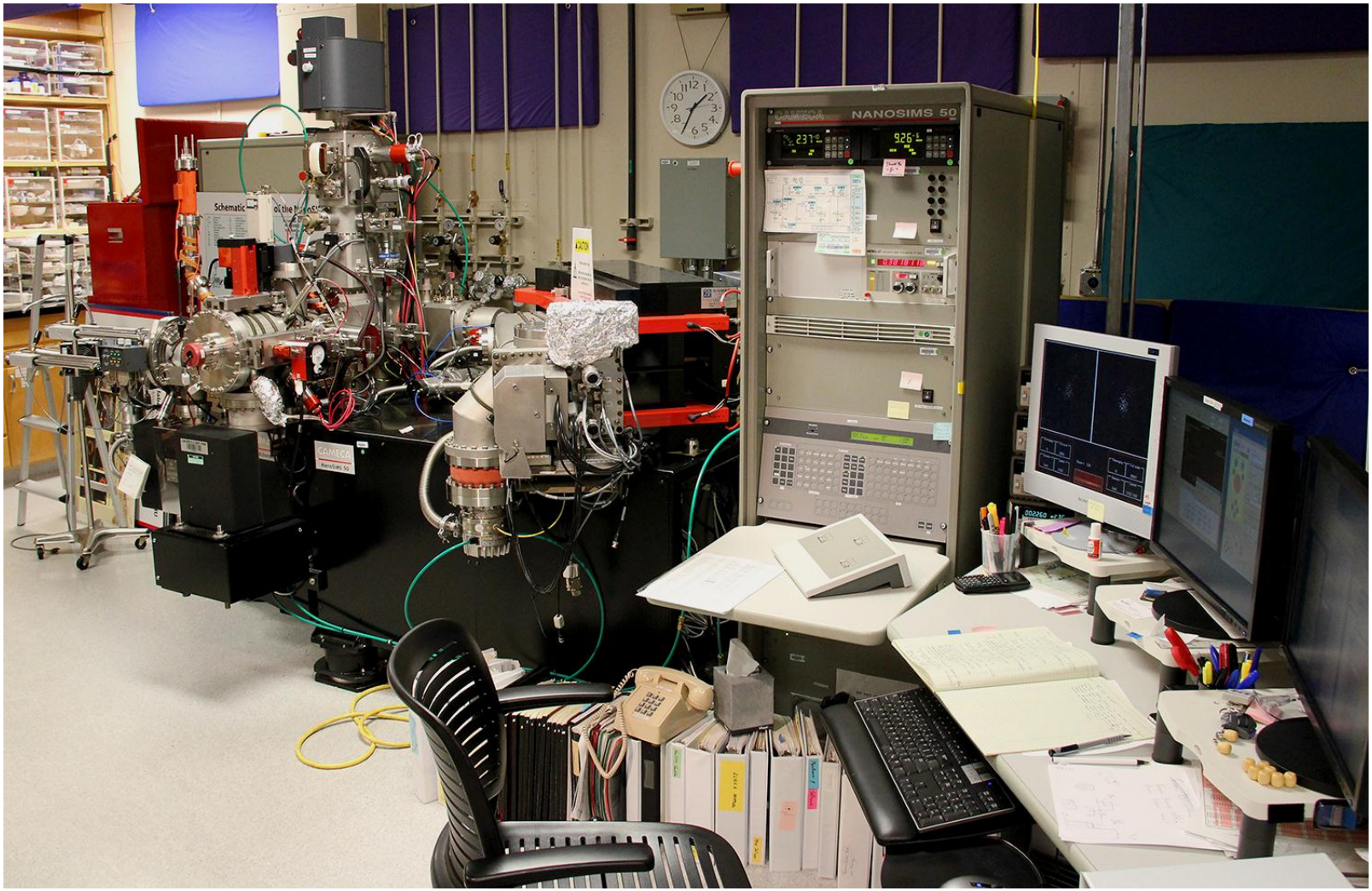
^{55}Mn image of "Ada" Stardust particle - estimated 2.56 billion years old – Courtesy J. Matzel



NanoSIMS image of bacteria and flagella *Hoplonympha*
Courtesy K. Carpenter

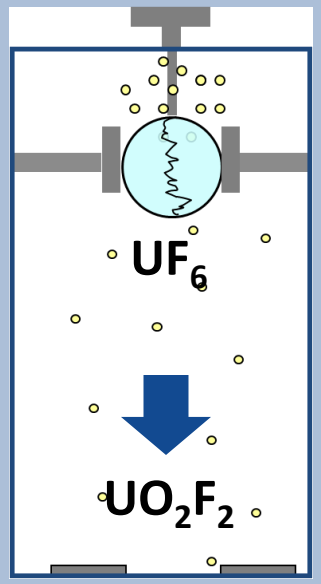
The Cameca NanoSIMS 50 at LLNL

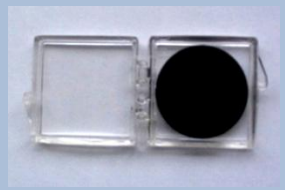
Secondary ion mass spectrometry at ultrahigh spatial resolution

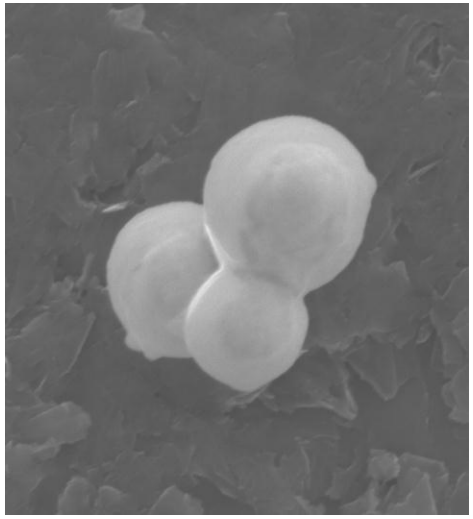


Preparation of UO_2F_2 particles
at the EU's Joint Research Centre, IRMM in Belgium

Aerosol deposition chamber

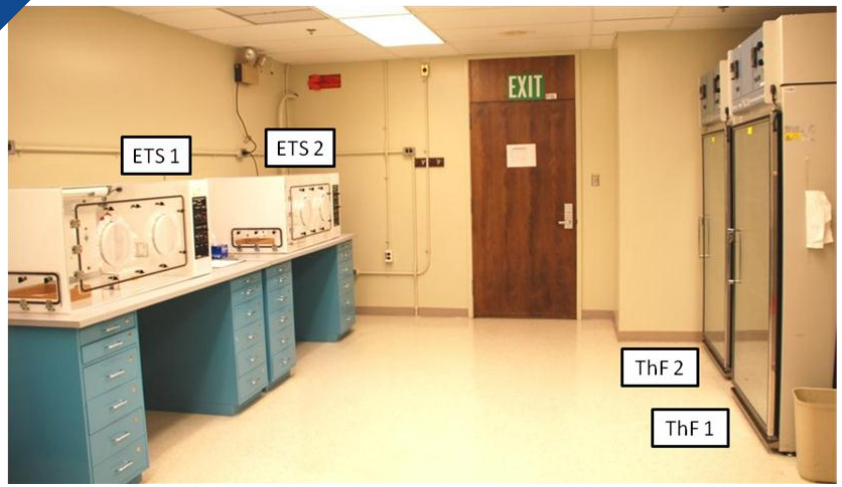




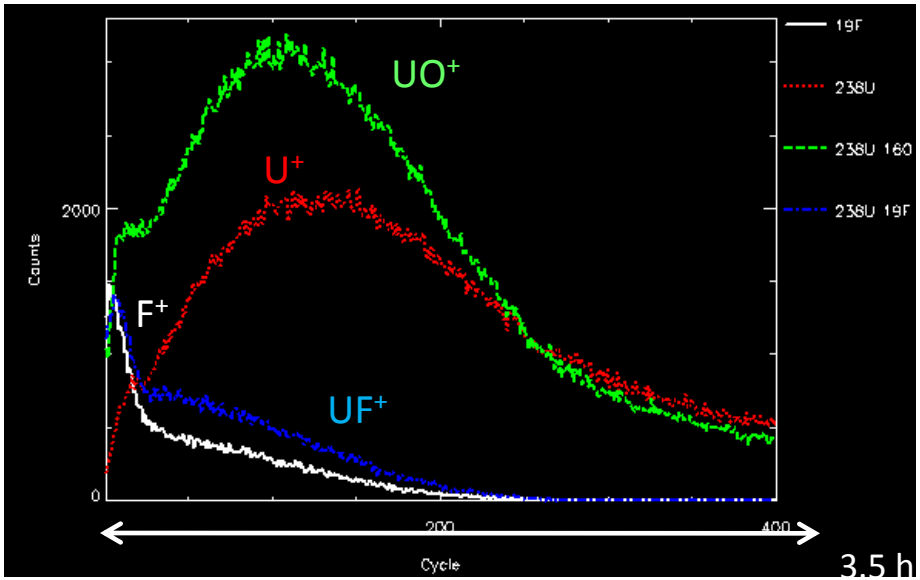


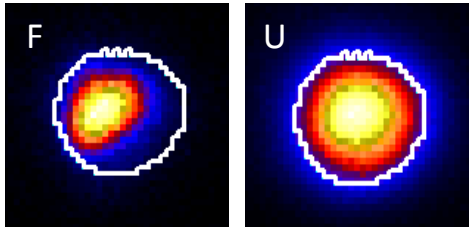
Storage in different environmental conditions at LLNL

	ETS 1	ETS 2	ThF1	ThF2
Temp	25 °C	25 °C	25 °C	25 °C
RH	<15 %	30-40 %	50 %	70-77 %
Light	diffuse	UV-light	diffuse	diffuse



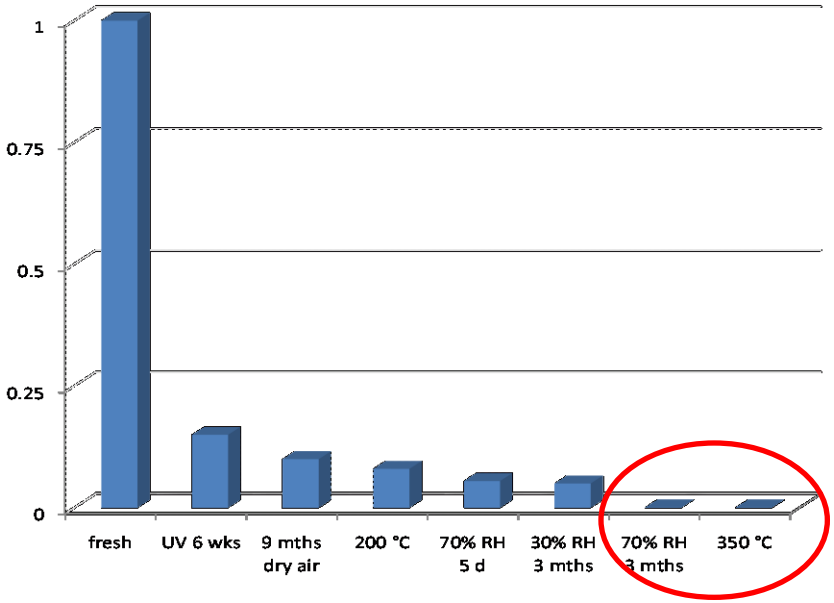
Correlation F-to-U ratio – exposure conditions





8 x 8 micron ion images of F and U

- NanoSIMS depth profiles collected for different exposure conditions
- F-to-U ratio calculated based on difference in ion intensity
- High humidity (70 % RH) and high T (350 C) caused biggest reduction in F-to-U ratio
- F still detected after 5 years at ambient conditions



F-to-U ratio for different exposure conditions

To conclude…

- **Particle analysis for environmental sampling**
  = Powerful tool for nuclear safeguards inspections
  - Focus on uranium isotope analysis
  - Demonstrated advantages of using complementary particle analysis techniques
- **NanoSIMS analysis of uranium oxyfluoride particles**
  - F and U distribution of individual particles
  - Variations in F-to-U ratio for different exposure conditions
  - High humidity and high temperature accelerate F loss
  - F still detected after 5 years for dry/intermediate RH conditions

Thank you!

Acknowledgements

Thanks to J. Zaug and D. Grant for help with the Raman measurements and to the NNSA Office of Nonproliferation and International Security (NA-243) for their generous support

The uranium oxyfluoride particles were prepared at IRMM expressly for the purpose of this study.
We also have no evidence that fugitive emissions from a uranium enrichment facility would, or would not, appear similar to the analyzed samples.

Thank you!