Actinide Measurements at the TANDY AMS Facility

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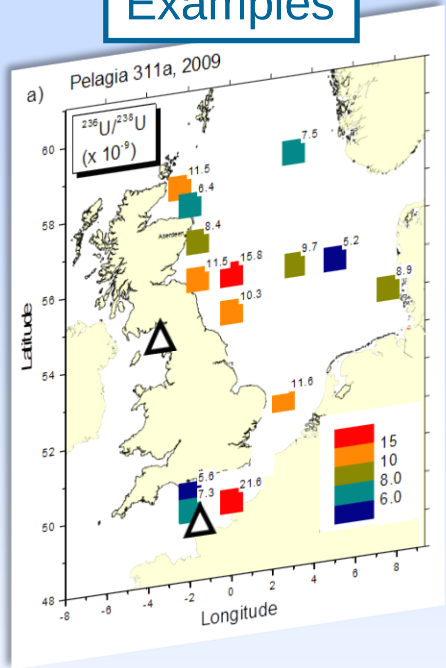
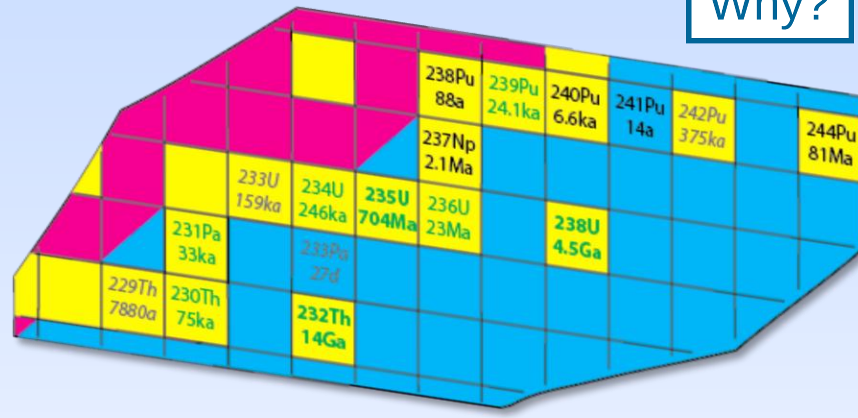
Examples

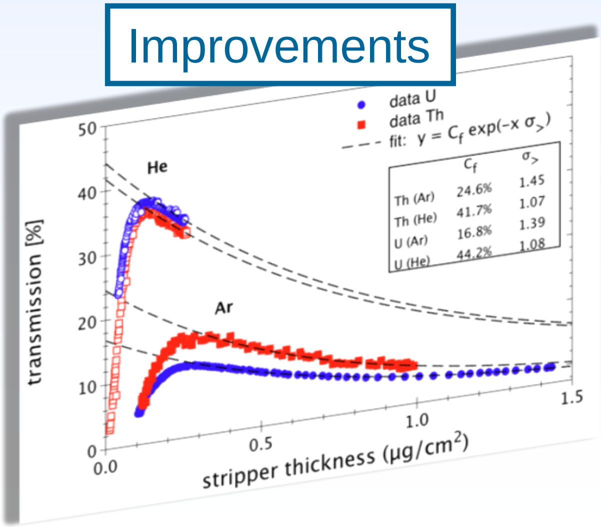


What do we measure?

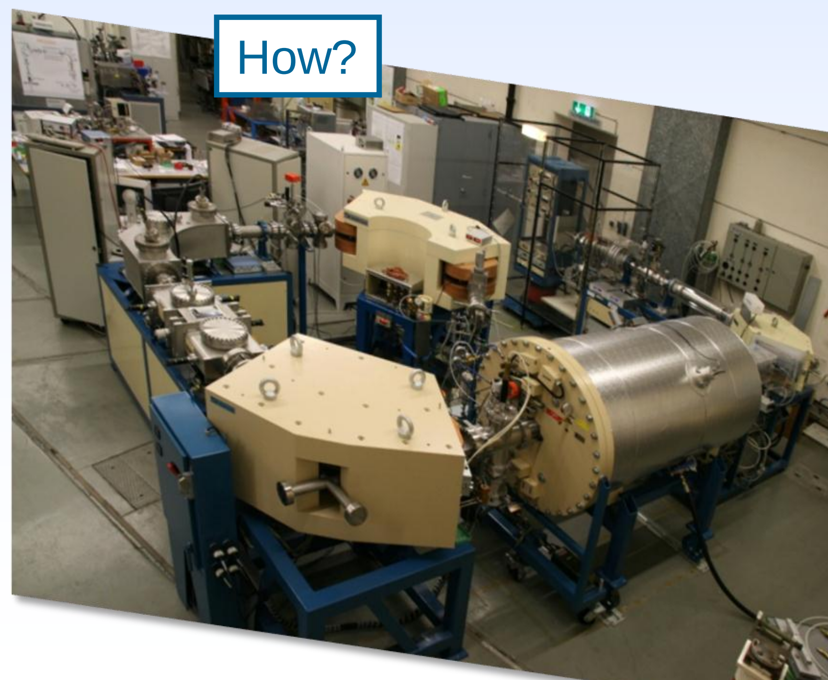
Why?



Improvements

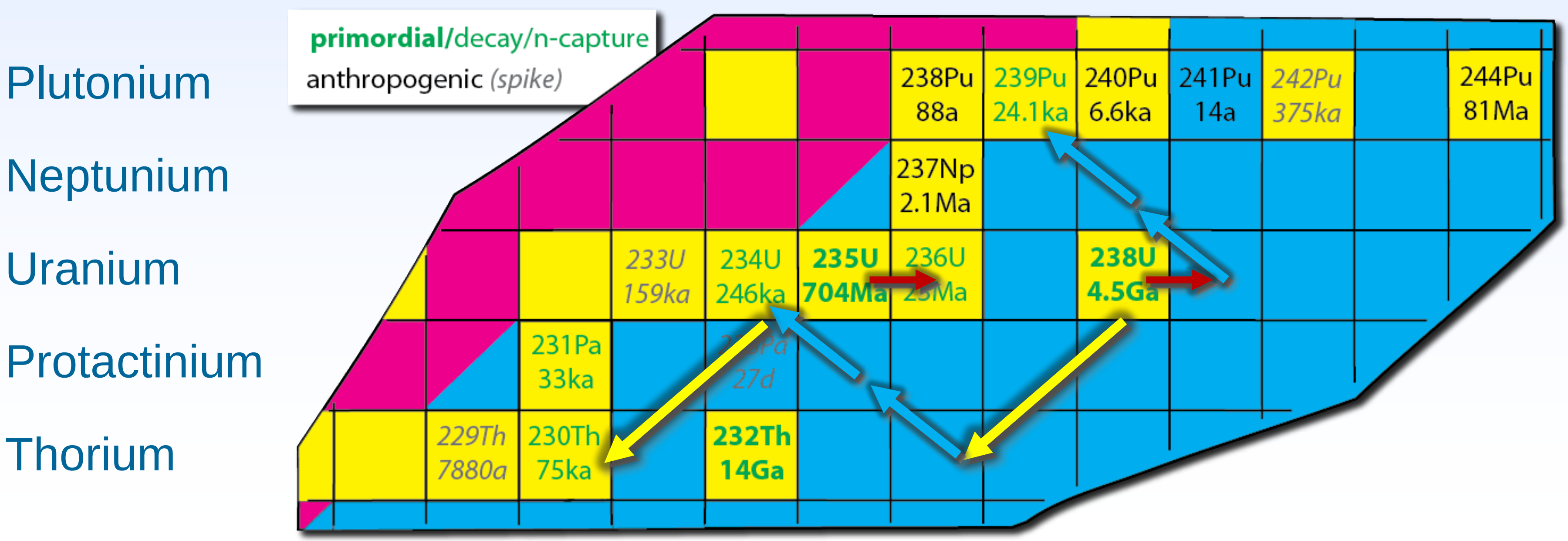


How?



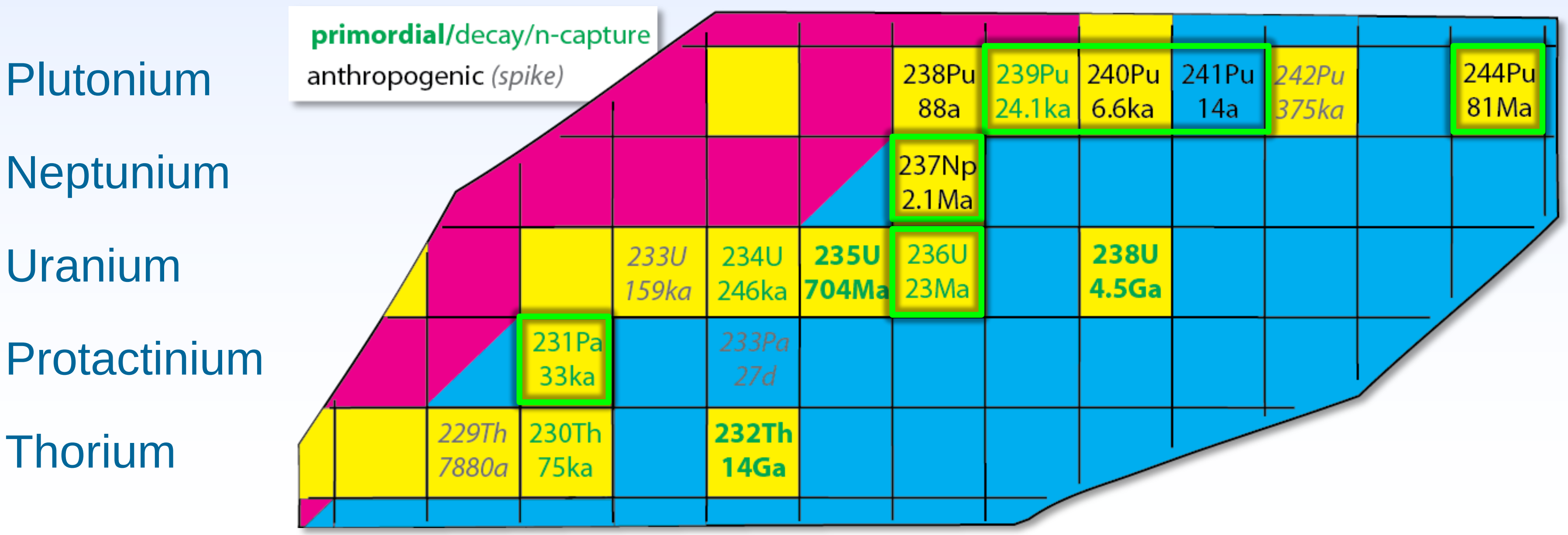
occurrence of the actinides

- natural occurrence:
  - long-lived ^{232}Th & ^{235,238}U in ores, water, sediment
  - elements from Uranium or Thorium decay chains, e.g. ^{230}Th, ^{231}Pa, ^{234}U
  - radioisotopes produced by natural neutron irradiation ^{236}U & ^{239}Pu
- anthropogenic input
  - nuclear reactors & reprocessing plants
  - atomic bomb explosions



applications

- exploration of the natural levels (^{231}Pa, ^{236}U,^{239}Pu)
- ^{236}U, ^{237}Np, Pu isotopes: transport processes of anthropogenic contamination in the environment
- ^{237}Np, ^{239,240}Pu: dosimetry for late effects of Pu/Np intake

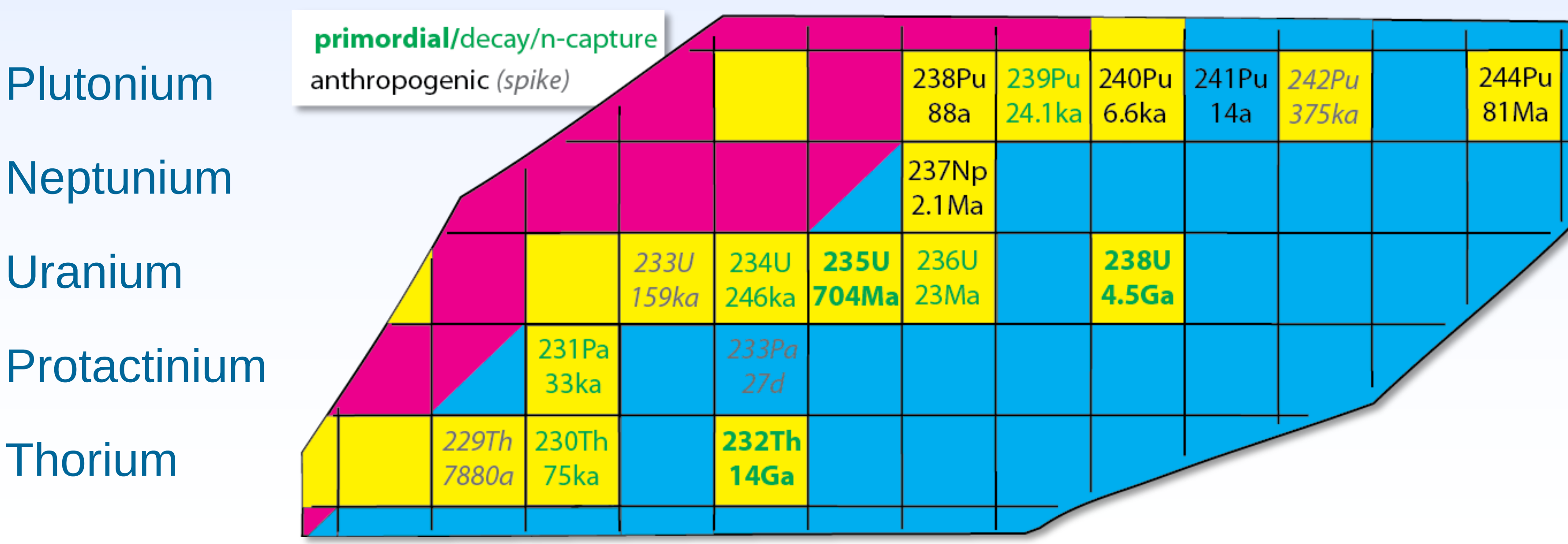


determining actinide concentrations

- AMS determines number of actinide atoms per volume or per mass of a liquid or solid sample
- overall efficiency of the determination has to be known

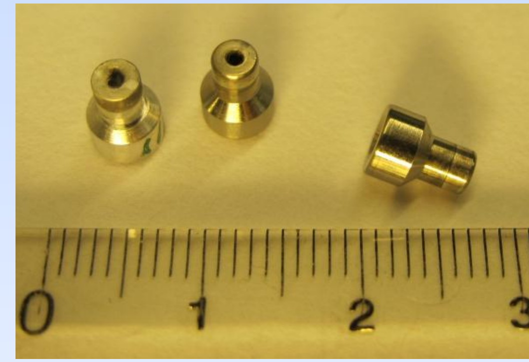
➔ sample is spiked with a known amount (e.g. 10^{10} at.) of an isotopic tracer (^{233}U, ^{242}Pu)

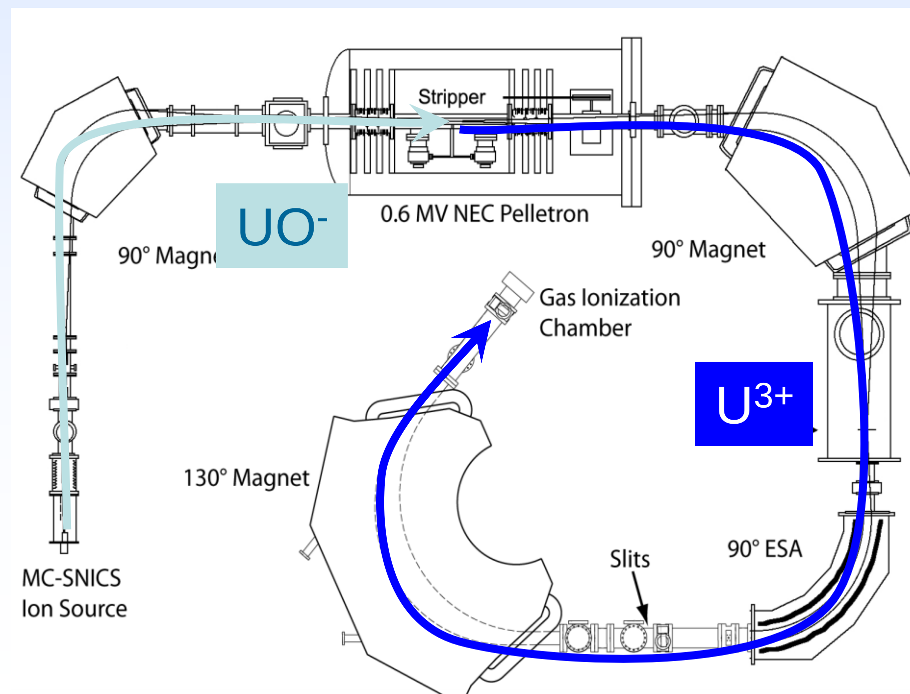
- efficiency is given by the tracer yield



AMS setup of actinide measurements

- target: Fe_2O_3 with actinide traces in Al cathode
- beam production: negative oxide molecules
  - ➔ ThO^-, UO^-, NpO^-, PuO^-
- accelerator operated at 300kV
- in the stripper at the middle of the accelerator: molecule-gas collisions → 3+ ions
- detection in gas ionization chamber



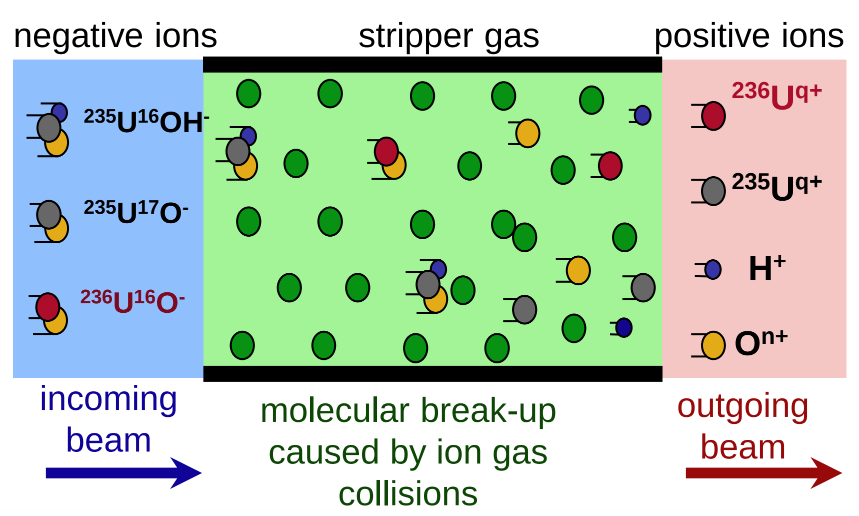


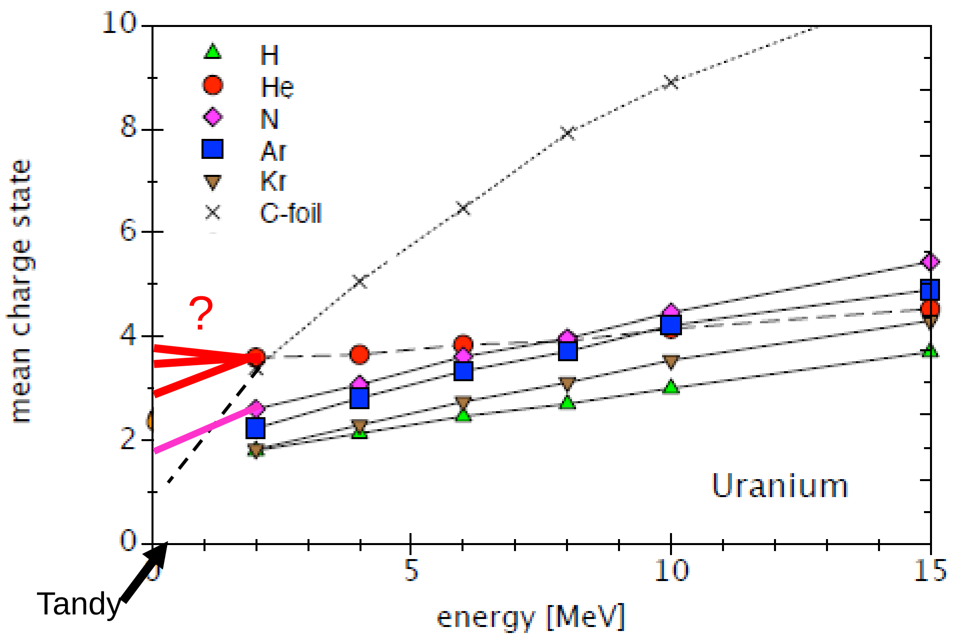
beam-gas collisions in the stripper

- charge exchange: $UO^- \rightarrow UO^{1+}, UO^{2+}, U^{1+}, U^{2+}, \mathbf{U^{3+}}, U^{4+}\ldots$
- high charge states suppress molecular background
- equilibrium charge state distribution with mean charge state q

  → depends on cross sections for e^--transfers between gas and beam

  → depends on stripper gas and beam energy
- high mean charge state at low beam energies ? → He as stripper gas!



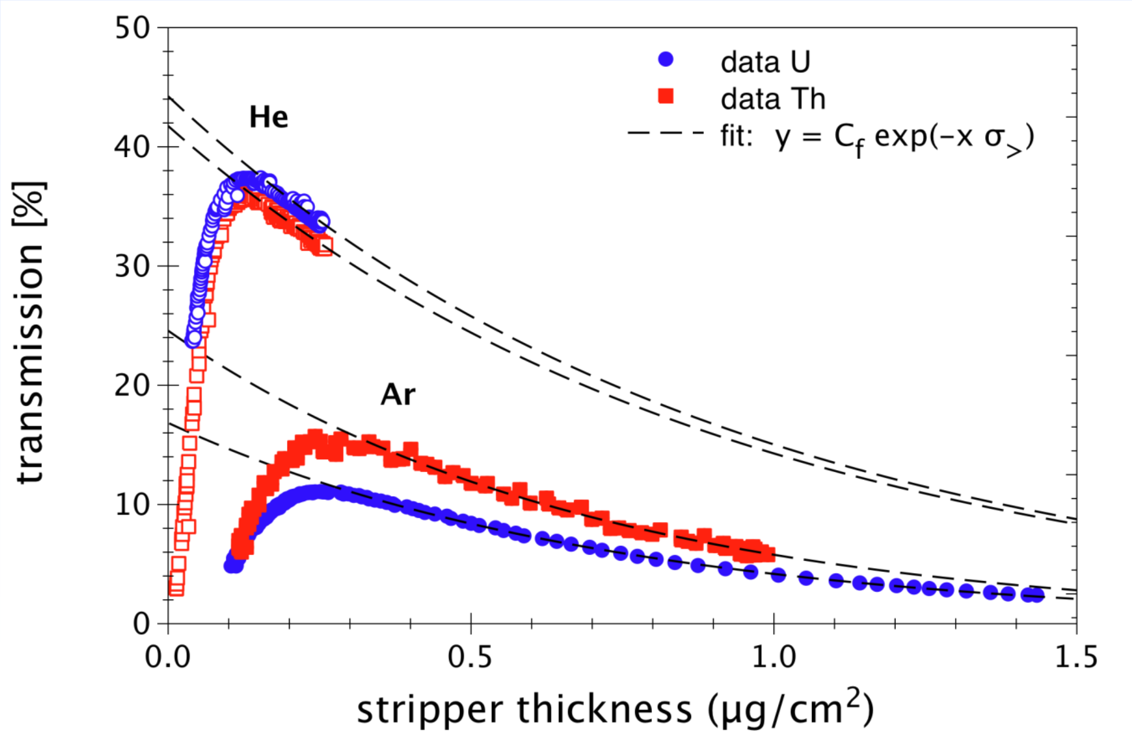


Wittkower & Betz, 1973; Vockenhuber et al., subm.

changing the stripper gas

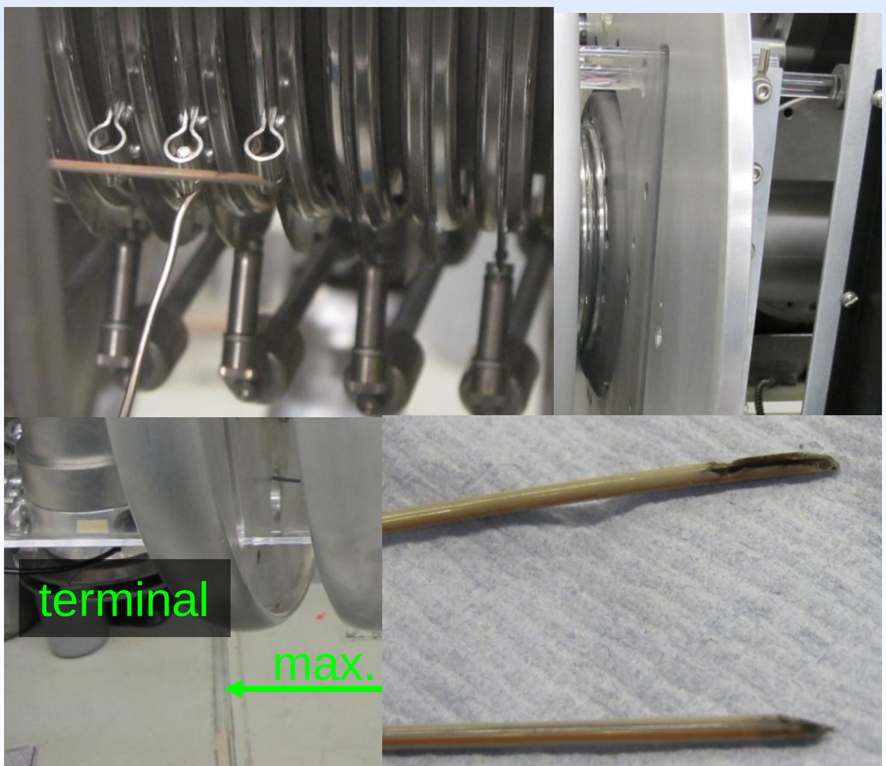
- with Ar as stripper gas: transmission $UO^- \rightarrow U^{3+}$ ~12%
- with He as stripper gas: transmission $UO^- \rightarrow U^{3+}$ ~37%

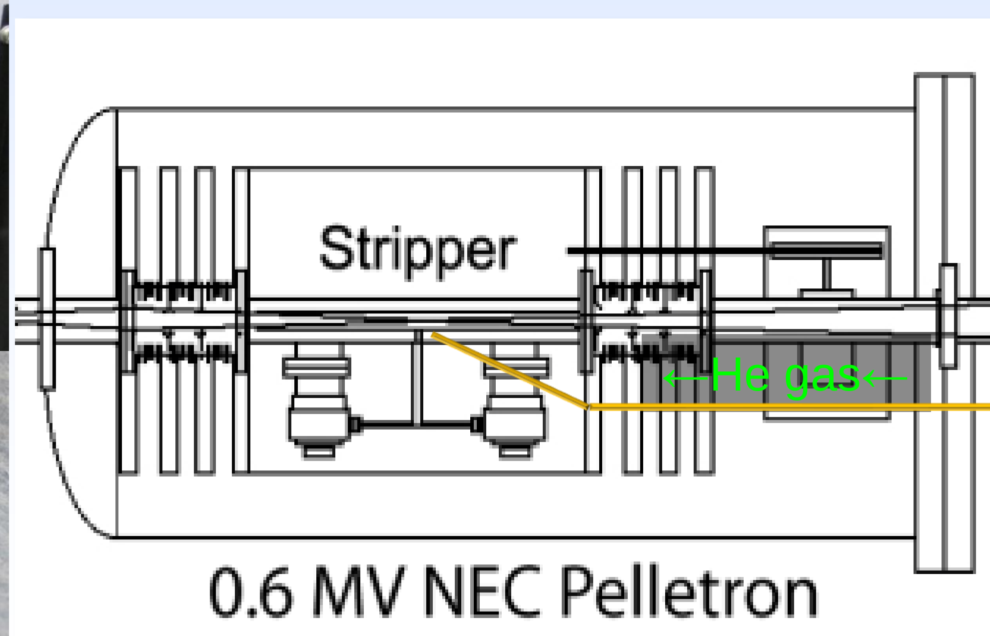
→ improvement by a factor 3 for actinide measurements!



He gas transport to stripper

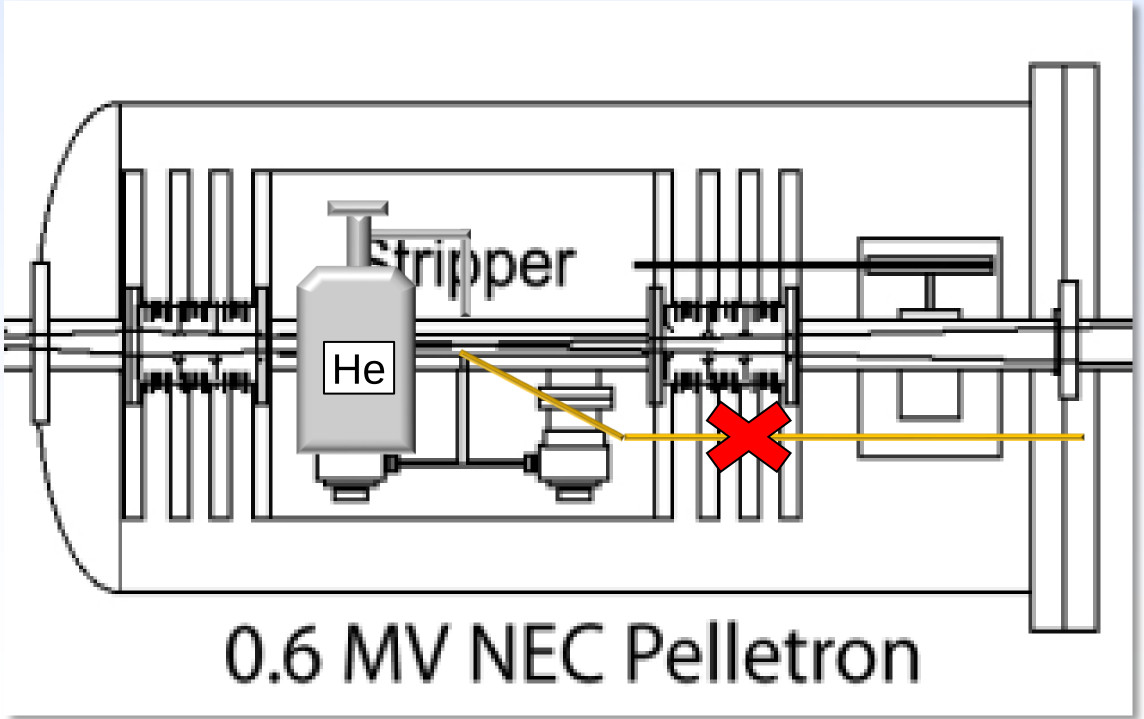
- He & Ar stripper gas in bottles outside the accelerator tank
- gas transport via an insulating capillary over a voltage gradient of max. 600kV/50cm
- electric discharges in He destroy the gas inlet capillary





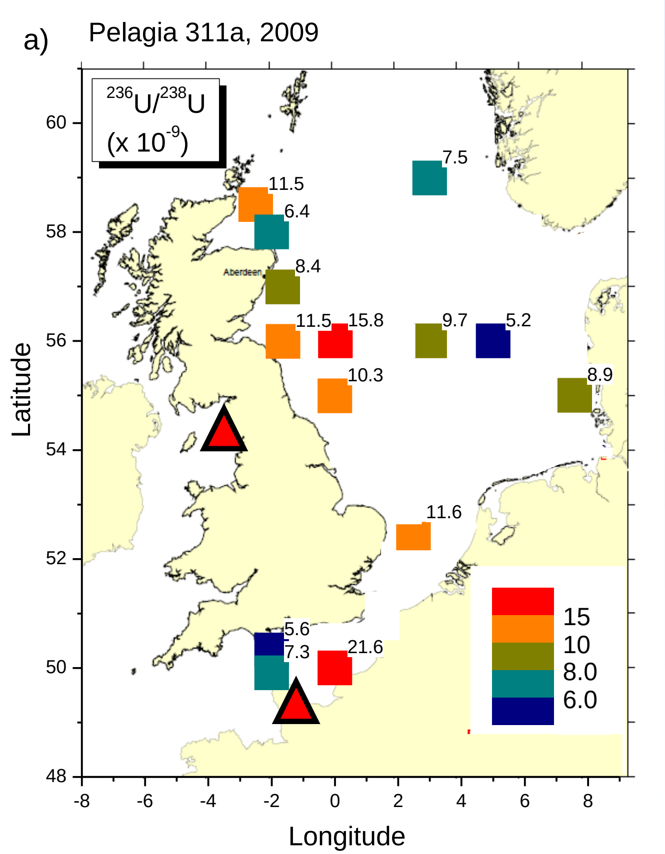
He gas transport to stripper

- removed the gas inlet capillary
- He bottle mounted on the terminal of the accelerator
- disadvantages: ~ no flexibility to use different stripper gases
  ~ openings of the tank to refill He will be necessary

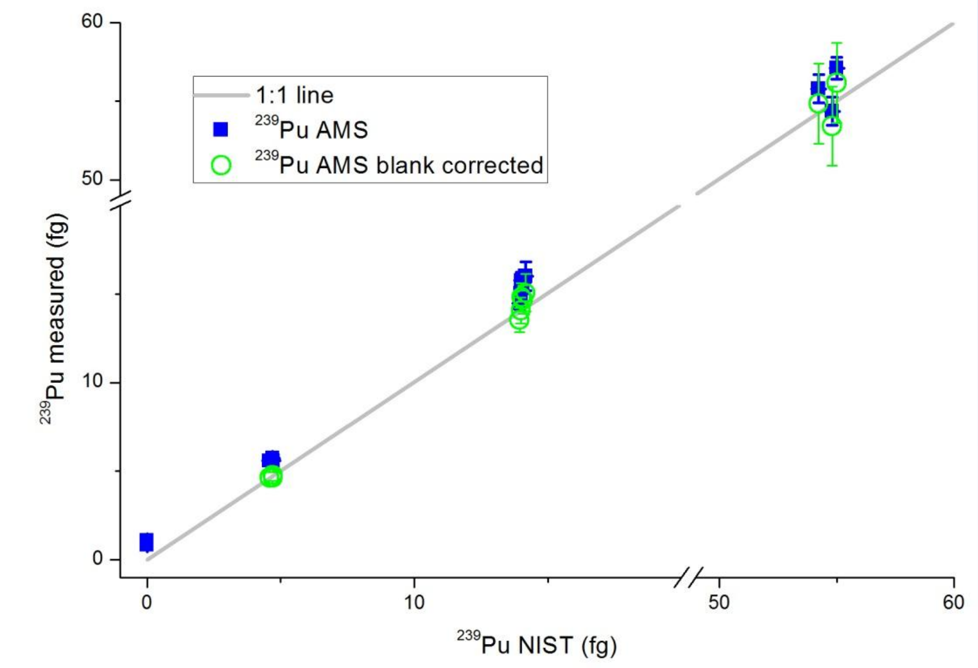


Two examples of our applications

^{236}U in the North Sea



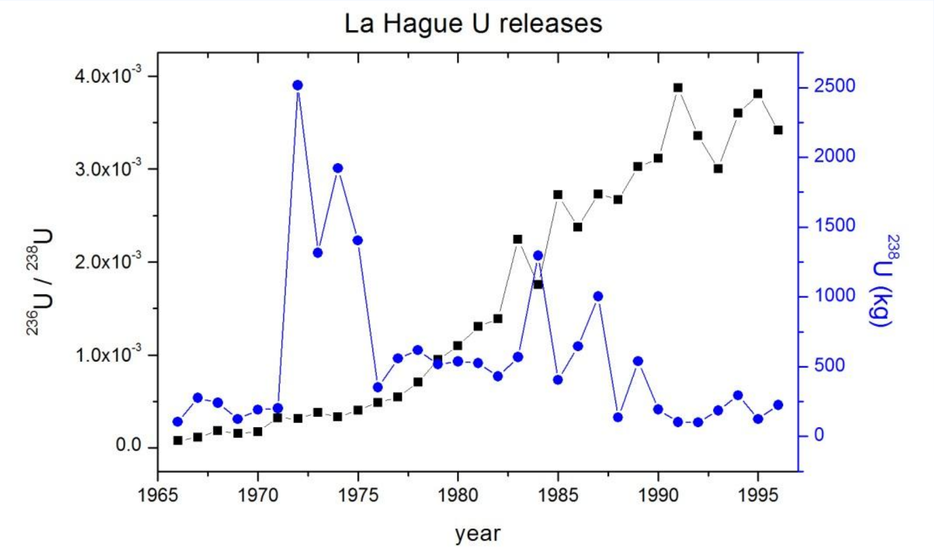
dosimetry with $^{239,240}Pu$

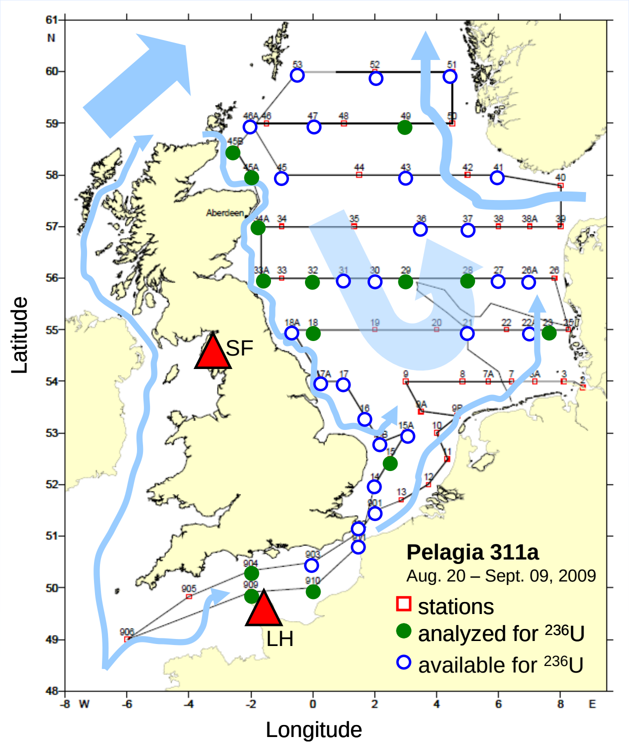


^{236}U release by reprocessing plants

- European reprocessing plants (La Hague, Sellafield) released ca. 40-50t U with increasing ^{236}U content
- $\sigma(^{235}U(n,f))$=586 barn, $\sigma(^{235}U(n,\gamma)^{236}U)$=95 barn

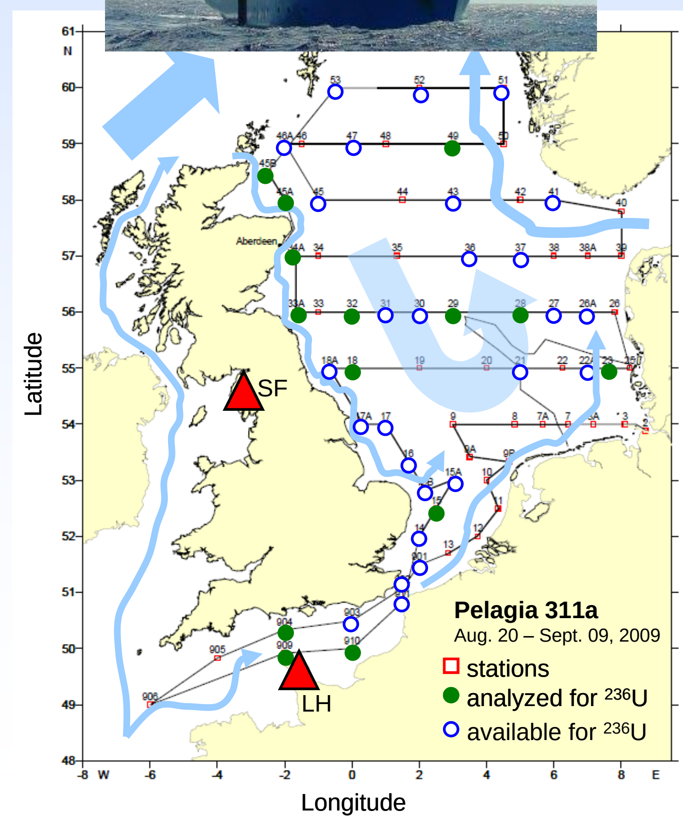
➔ in reprocessed fuel: $^{236}U/^{238}U \sim 10^{-3}$





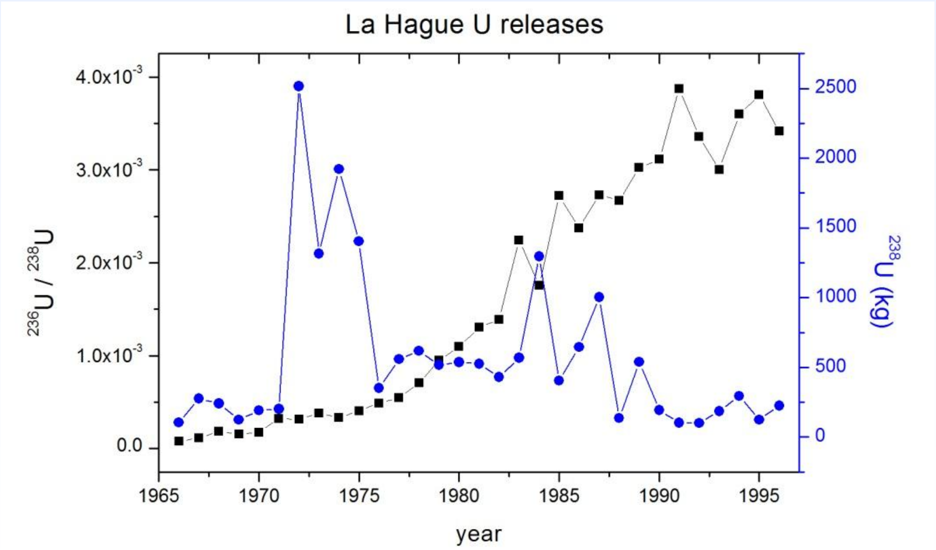
^{236}U – sampling and chemical preparation

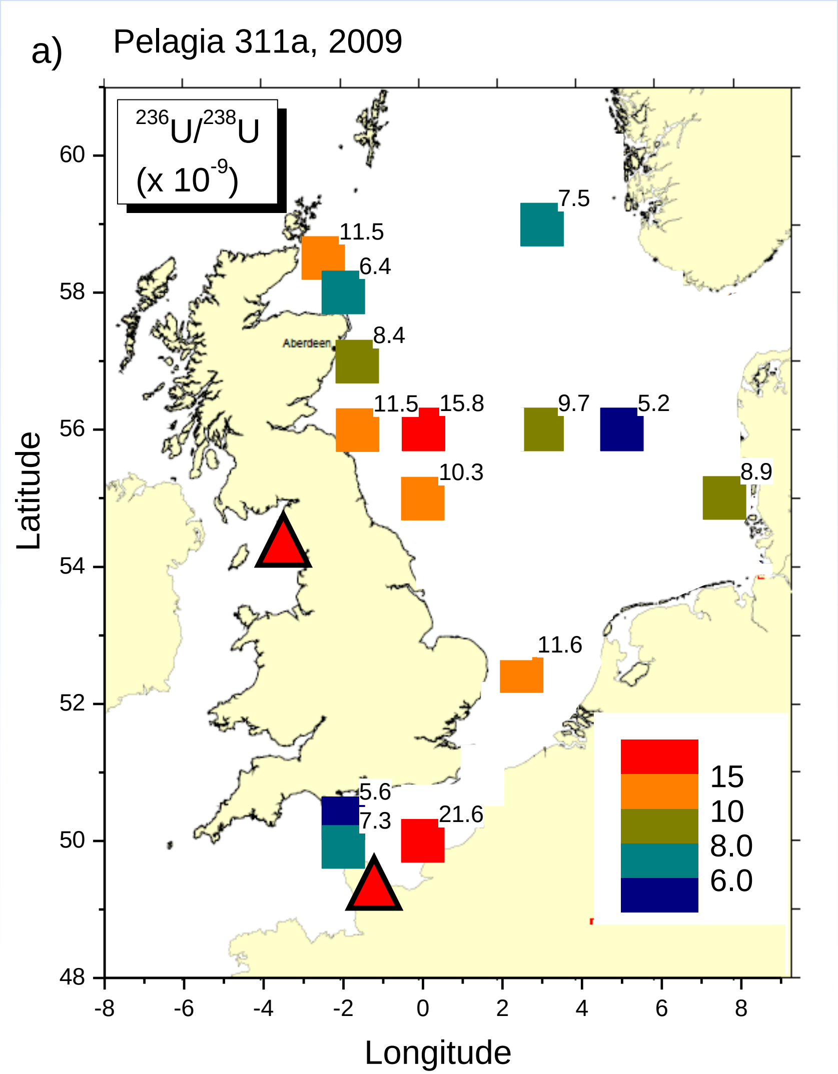
- trace the transport of the ^{236}U labelled water
- collaboration with BSH Hamburg
- annual sampling for monitoring of the North Sea
- analysis of ca. 30 surface water samples (15L)
- extraction of U in precipitations and purification on anion exchange columns (UTEVA resin)



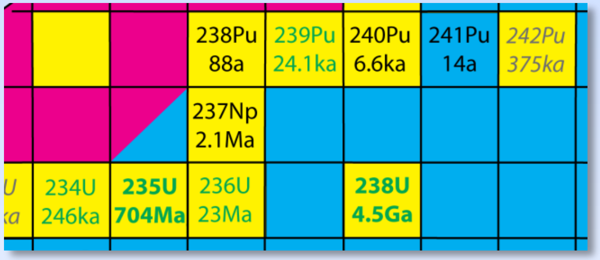
^{236}U in the North Sea

- in north sea surface water:
  $^{236}U/^{238}U \sim 10^{-8}-10^{-9}$
- in reprocessed fuel: $^{236}U/^{238}U \sim 10^{-3}$
- in the ocean water: 3µg/L U
  Atlantic Surface water: $^{236}U/^{238}U \leq 10^{-9}$





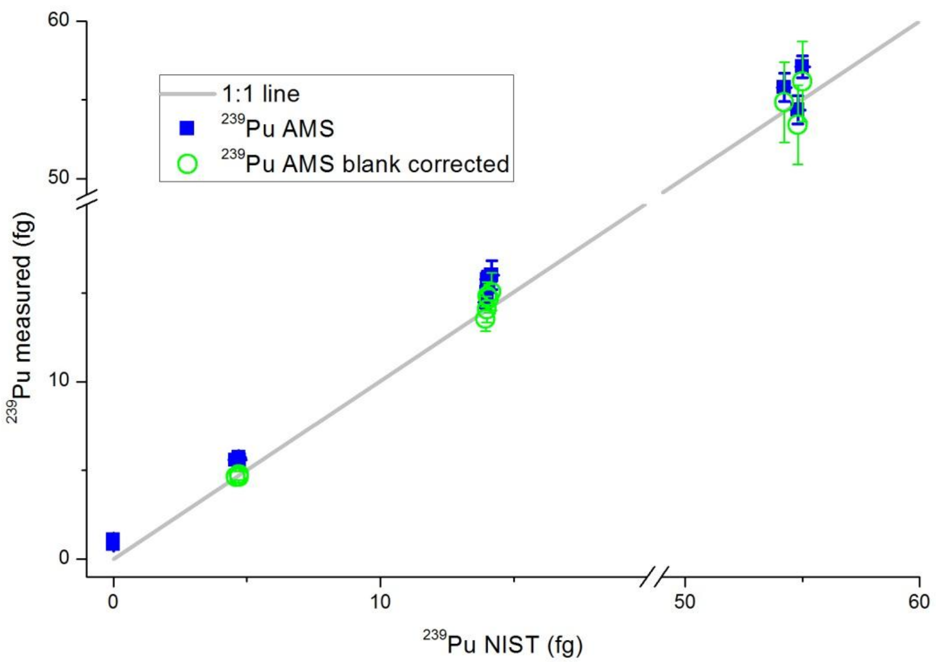
^{239,240}Pu for dosimetry

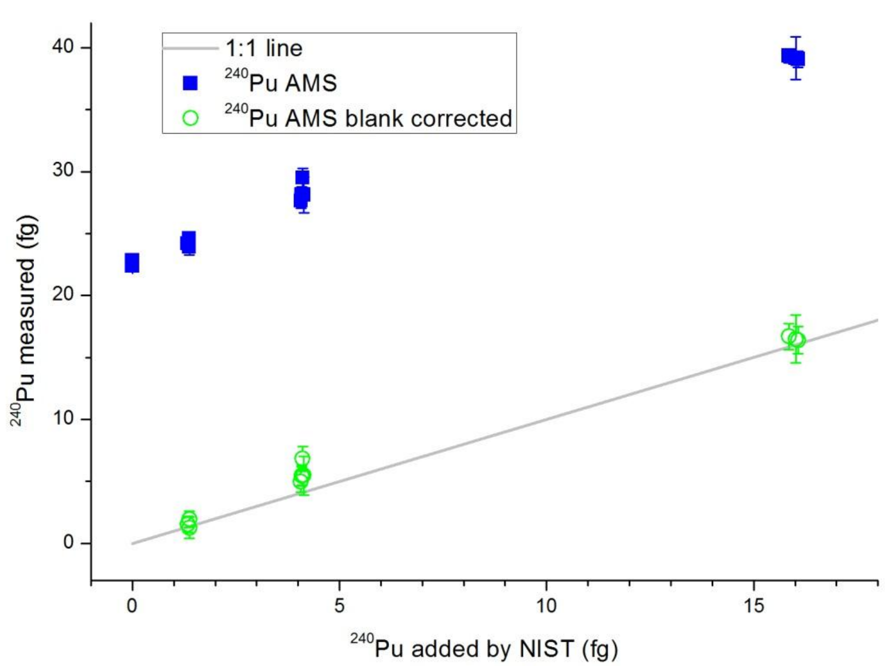


- monitoring of radioactivity intake in exposed workers of research and nuclear industry in blood & urine samples
- inhalation may lead to a long-term exposure and to tumours in lung, skeleton and liver
- usually performed by counting techniques
- general aim in monitoring: verify that no exposure is/was present
- ^{239}Pu activities of µBq per liter urine sample: 10 µBq ≈10^7 atoms ≈ 2 fg
- AMS is an attractive method for long-lived radioisotopes
  → overall efficiency up to 10^{-4}
- measurement relative to a ^{242}Pu tracer gives the concentration

^{239,240}Pu for dosimetry

- collaboration with Femtomass/Chalk River Labs, Canada
- available from NIST: standard dilution series with unknown concentrations of ^{239}Pu and ^{240}Pu in artificial urine
- returned raw measured values to NIST
- blank corrected values agree perfectly with 1:1 line (deviation of 1.5%±3.5%)
- concentrations in the fg level can be determined





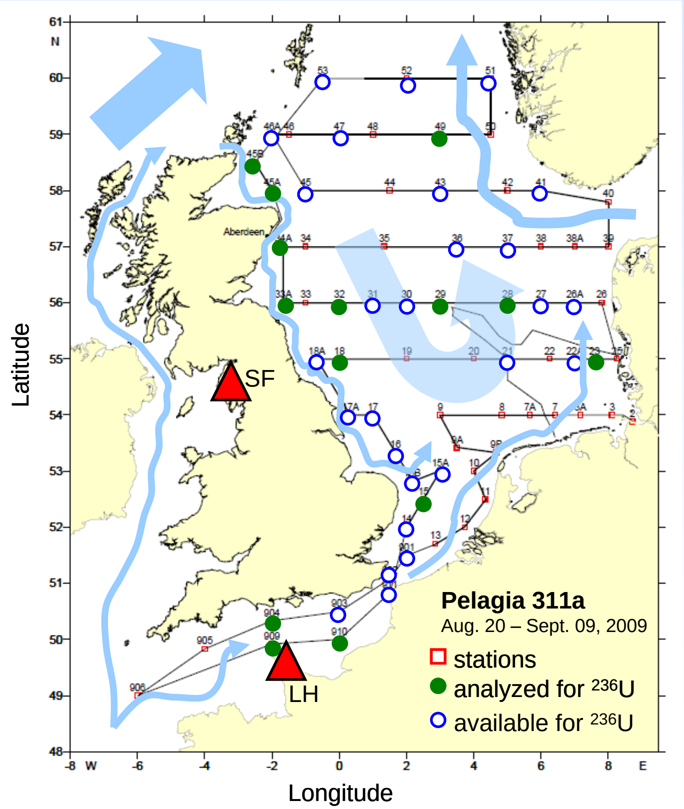
Summary & Outlook

setup & improvements of the spectrometer

- He as stripper gas improves efficiency
- new construction of the stripper gas supply was necessary
- further reconstruction to regain flexibility of using different stripper gases

routine measurements

- *$^{236}U/^{238}U$ in water*
  - more samples from the North Sea 2009, 2010, 2011
  - Atlantic transect
- *actinides for dosimetry*
  - measurement of Pu samples is certified and ready for external users
  - development of Np detection in progress



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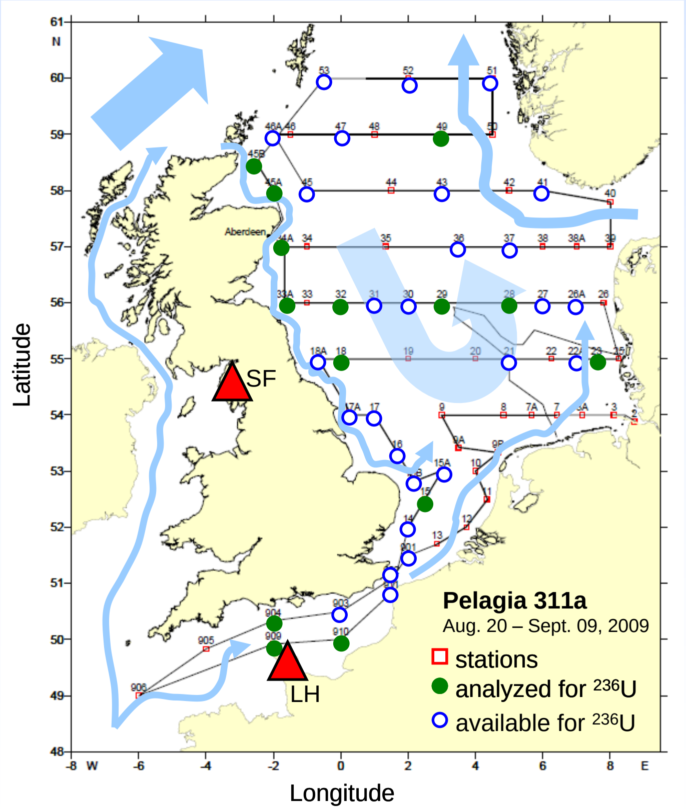
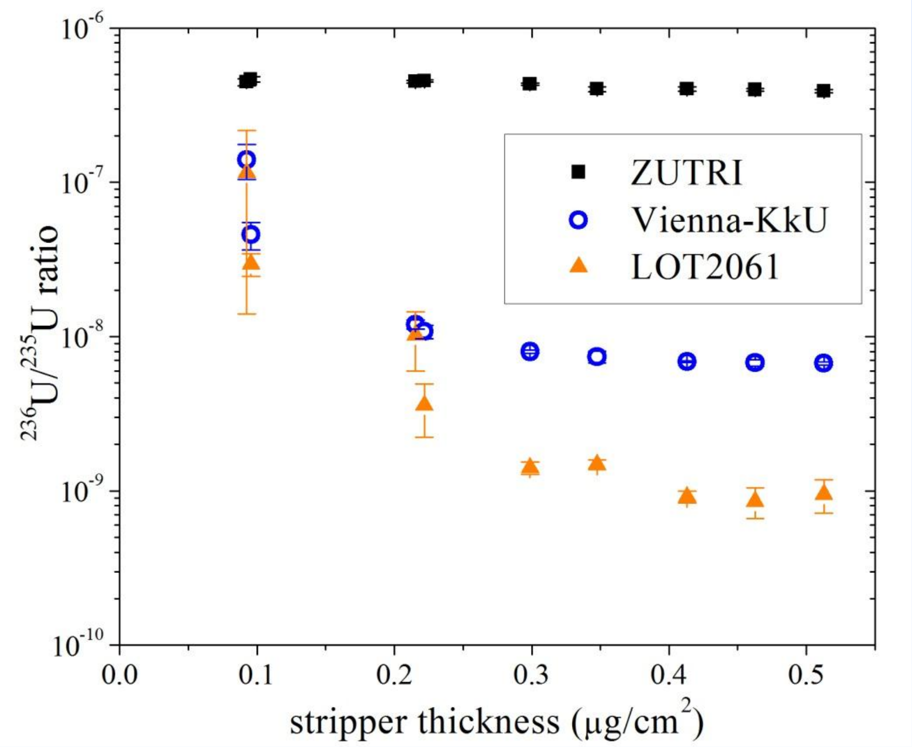


3+ molecules surviving the stripping process

- stripper gas density dependent counting rates indicate existence of 3+ molecules:
  $^{232}ThH^{3+}$, $^{235}UH^{3+}$ and $^{238}UH^{3+}$
- dangerous for sensitive ^{236}U studies: molecular isobars cannot be separated in the detector

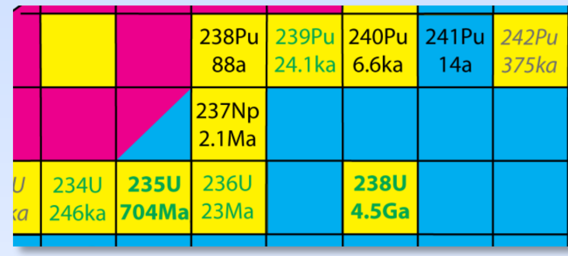
➔ suppression of $^{235}UH^{3+}$ molecules demands for higher stripper thicknesses



Sample name	$^{236}U/^{238}U$	
ZUTRI	$4.1*10^{-9}$	ETHZ in-house standard
Vienna-KkU	$6.98*10^{-11}$	material from P. Steier, VERA
LOT2061	$6*10^{-12}$	

^{237}Np for dosimetry

- collaboration with DTU Risø
- no appropriate Np isotope available as tracer
  ➔ measurement normalization against spike ^{242}Pu
- critical: same behaviour of both elements in chemistry and AMS measurement?



✓ reliable chemical separation of both elements possible

ϟ blanks show too much ^{237}Np

ϟ AMS efficiency for Np detection lower than for Pu (0.23:1)

ϟ formation of NpO^- depends on the target matrix, while the PuO^- formation does not