IAEA-CN301-231

universität wien

VERA
Vienna Environmental Research Accelerator
Faculty of Physics - Isotope Physics

Ultra-trace analysis of anthropogenic long-lived radionuclides in the environment with AMS

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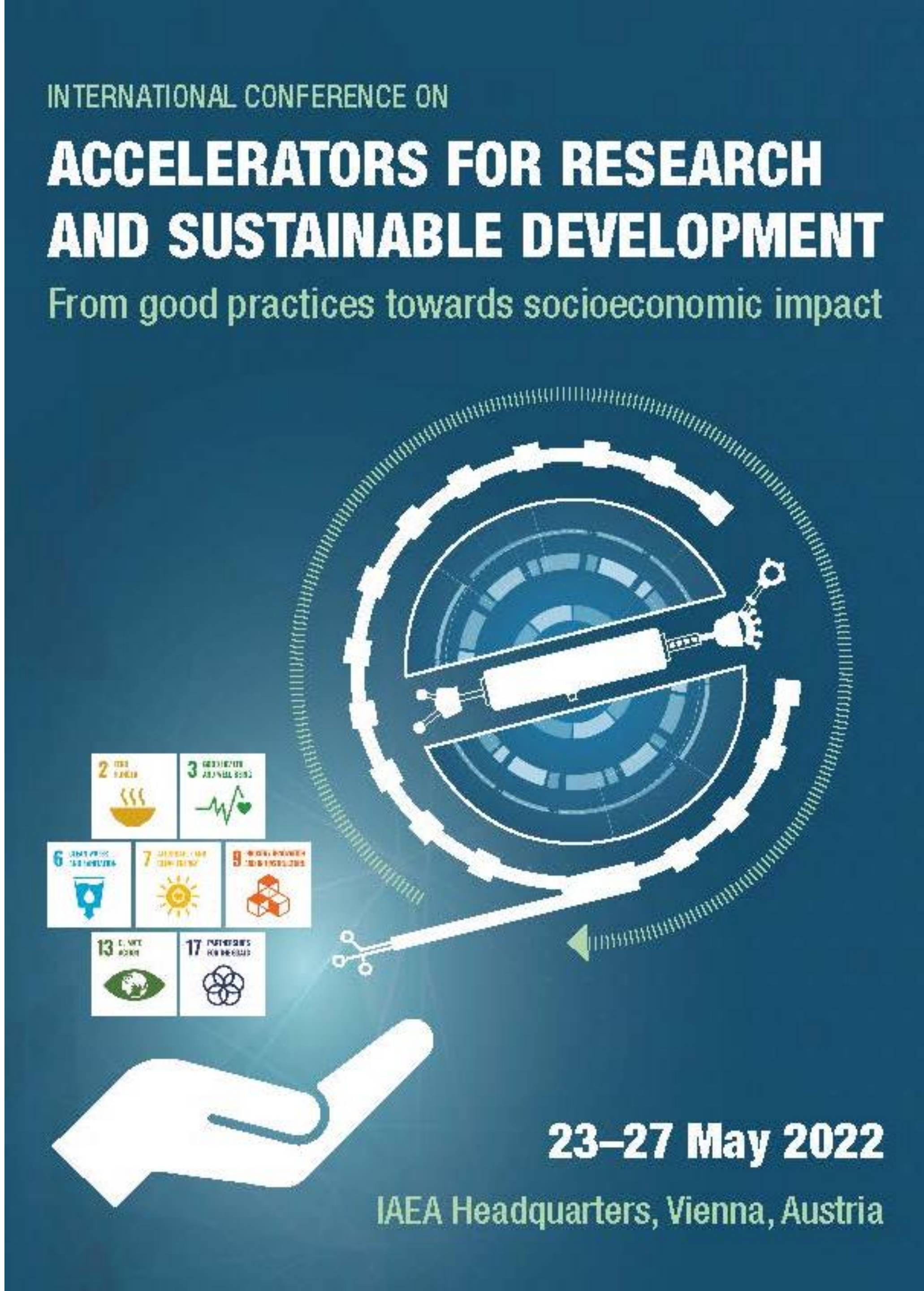


MIGRATION BEHAVIOUR

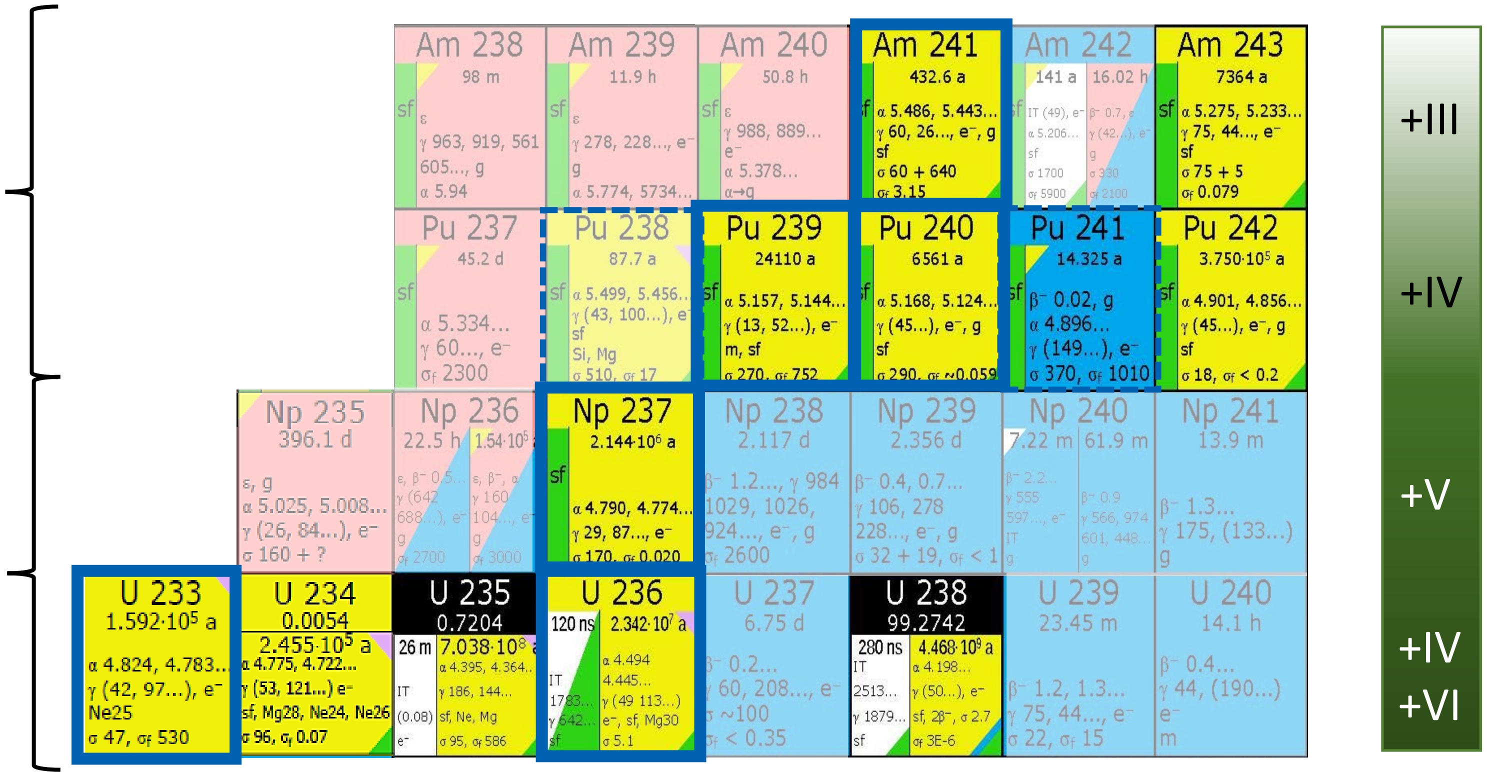
Particle-reactive

→ Stationary!?

Well soluble

→ Mobile!?

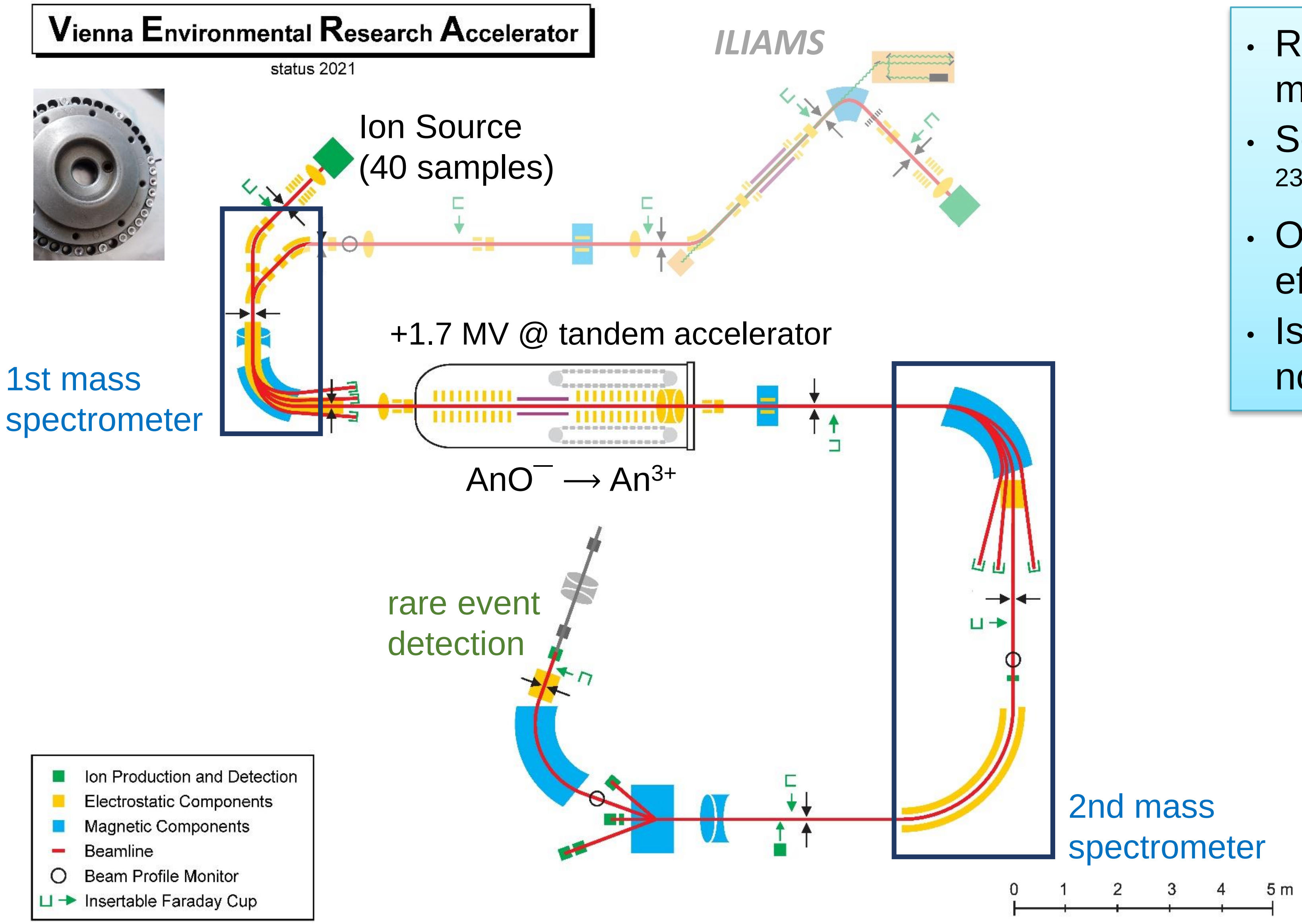
Oxidation state



Study migration behaviour in environmental systems at trace level

Radioecology, Nuclear Waste Management, Environmental Sciences (use as tracers)

AMS @ VERA

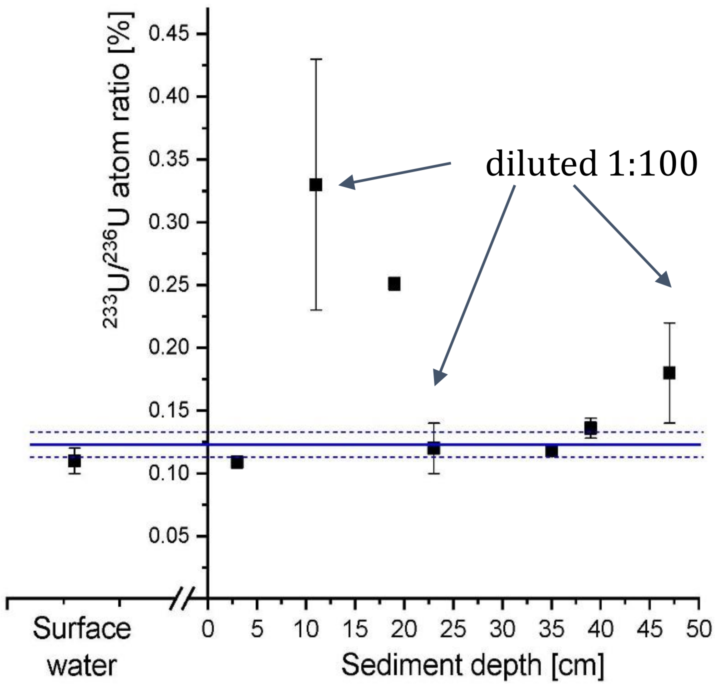


- Routine actinide measurement at VERA
- Sensitivity limit: $^{236}U/^{238}U < 10^{-12}$
- Overall detection efficiency: $5 \cdot 10^{-4}$
- Isotopic spike for normalisation

Detection Limit:
ag (10^{-18}g)

Irish Sea: nuclear reprocessing

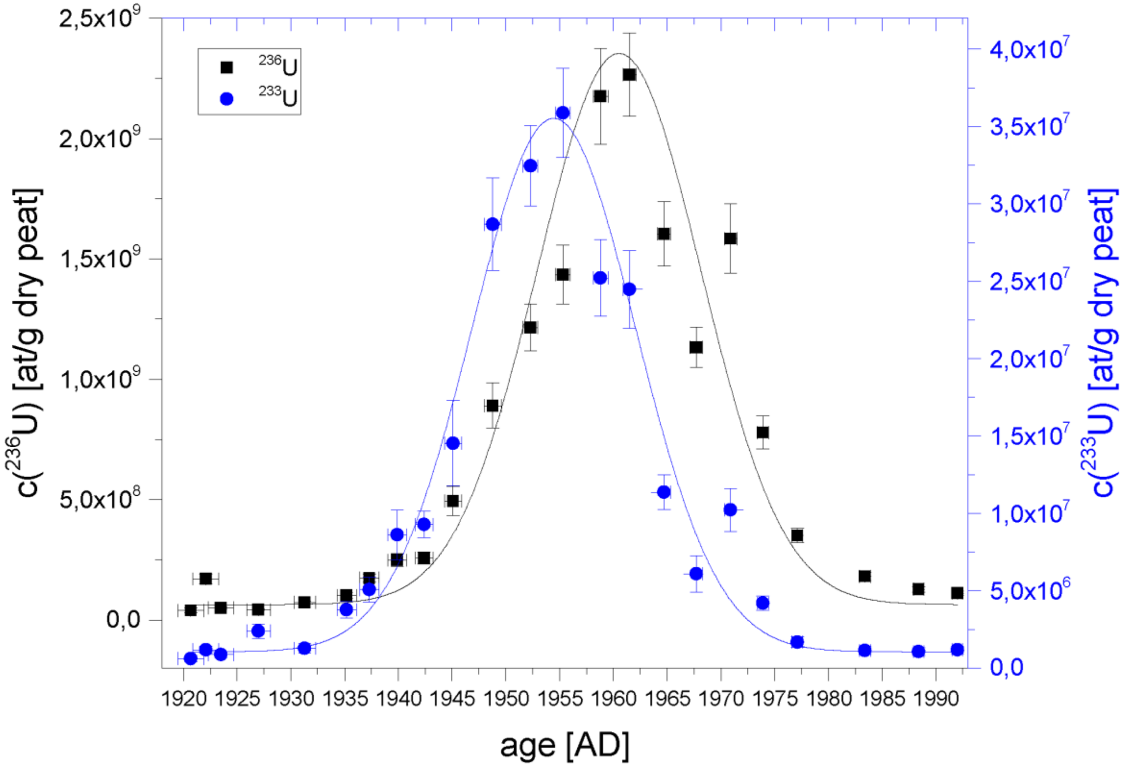
- Dated Sellafield Sediment core
- IAEA-381: Irish Sea Water (1993)



$^{233}U/ ^{236}U = 0.13 \pm 0.02$ %

Peat bog: global fallout

- Core from Black Forest (Germany)

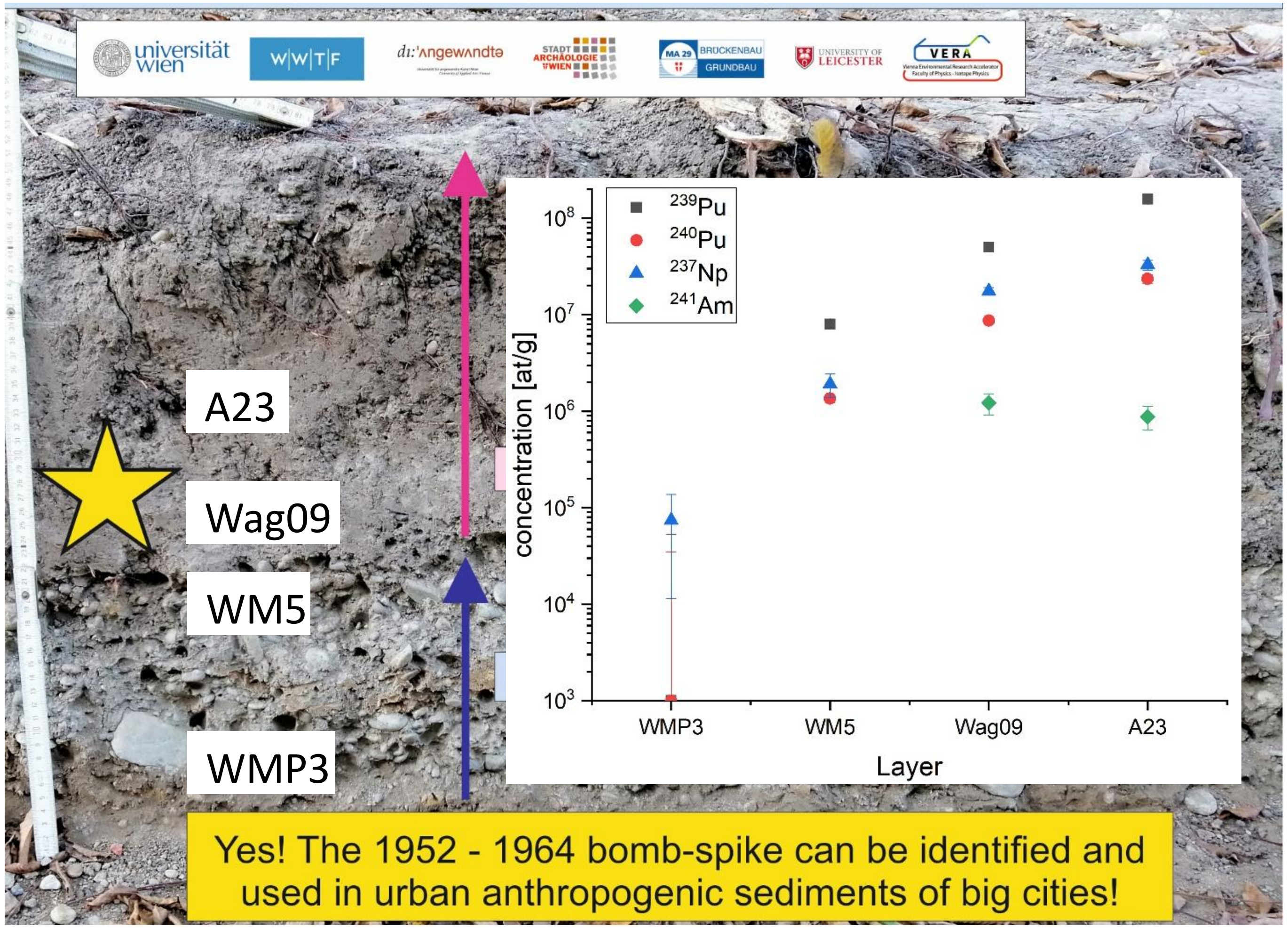


$^{233}U/ ^{236}U = 1.5 \pm 0.2$ %

M. Wagreich, M. Meszar, J.Wolf

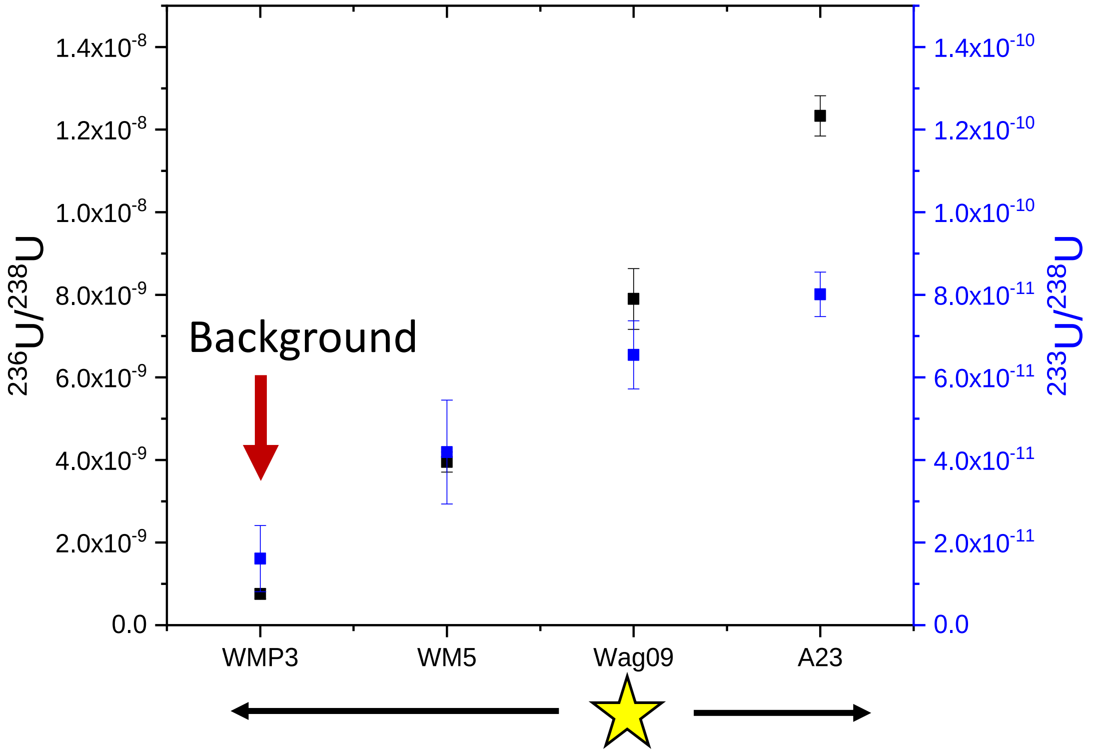
Karlsplatz, Vienna, Wien Museum

Sample mass: ≈ 10g (sieved and ground)



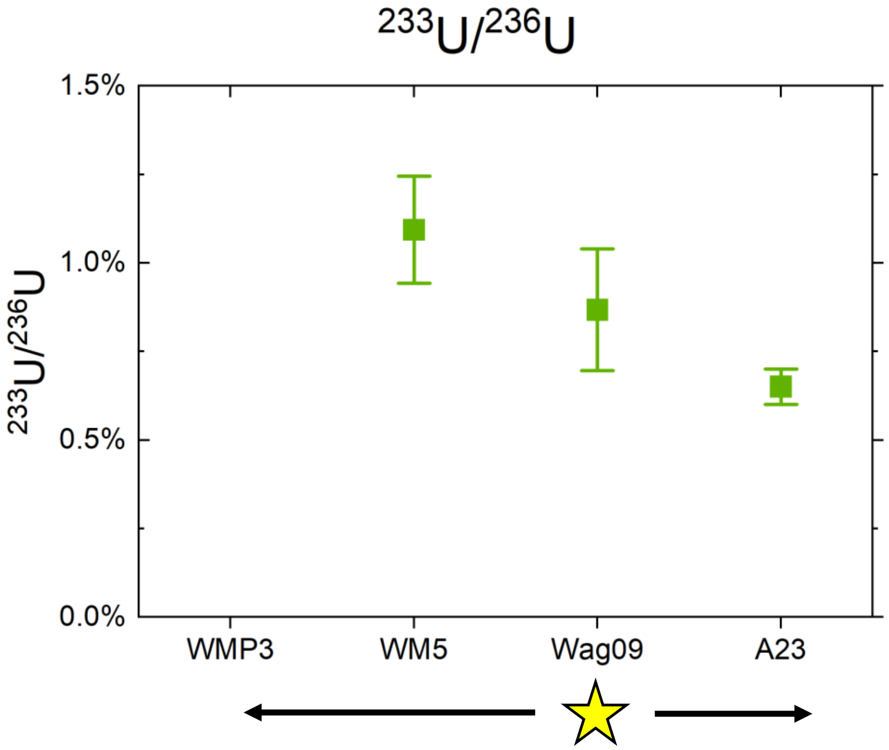
M. Wagreich, M. Meszar, J.Wolf

U ratios for source identification



1st half of 20th century
WW2 rubble

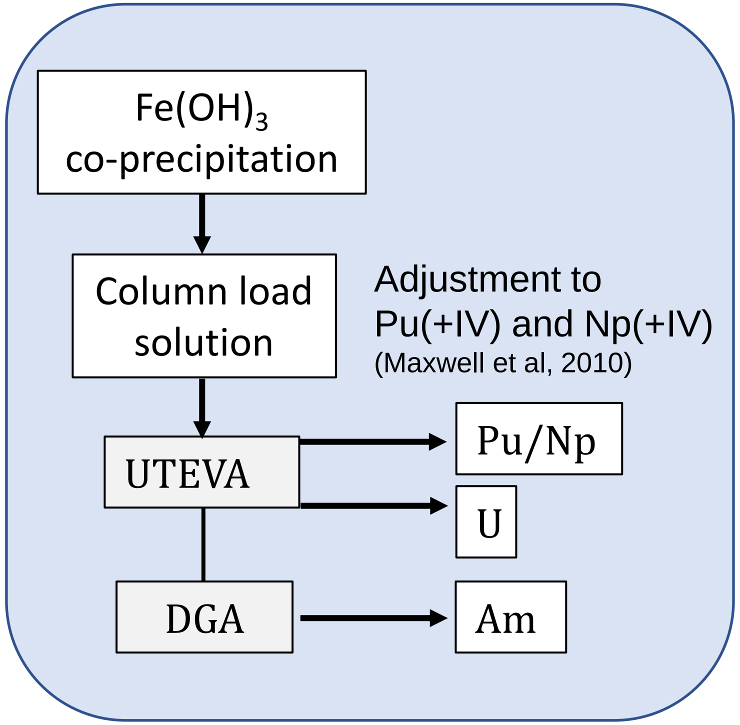
1959 to now



- In contrast to all other anthropogenic radionuclide signals $^{233}U/^{236}U$ **decreases!**

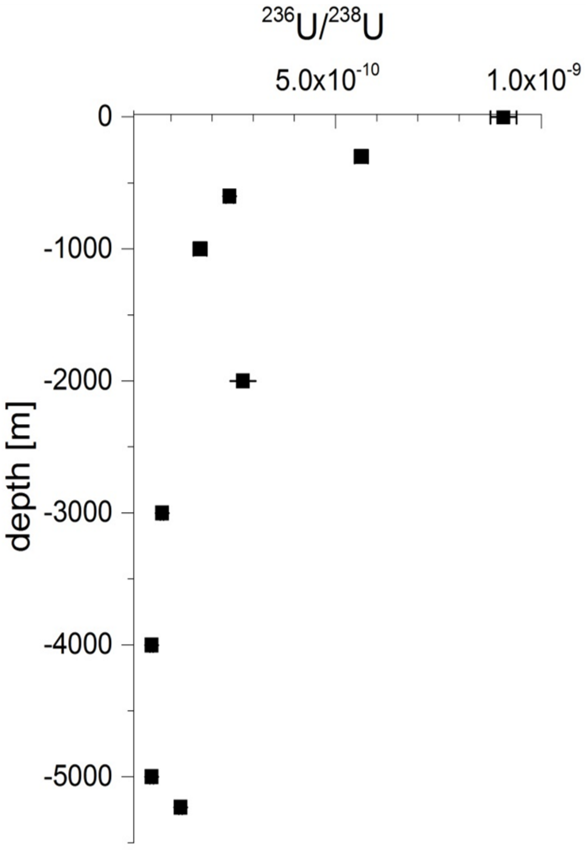
ANALYSIS OF ^{237}Np

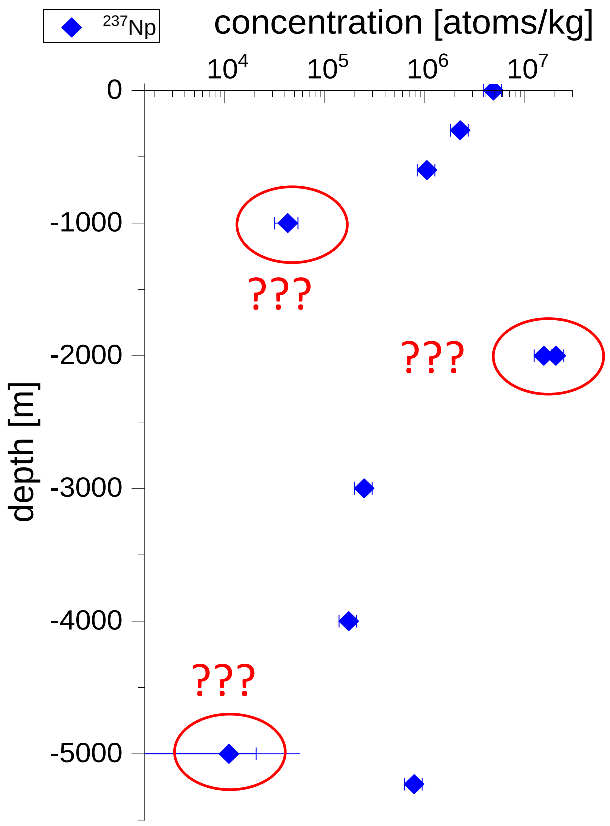
- Normalized to internal ^{242}Pu spike
- Ext. Standard with known $^{237}Np/^{242}Pu$



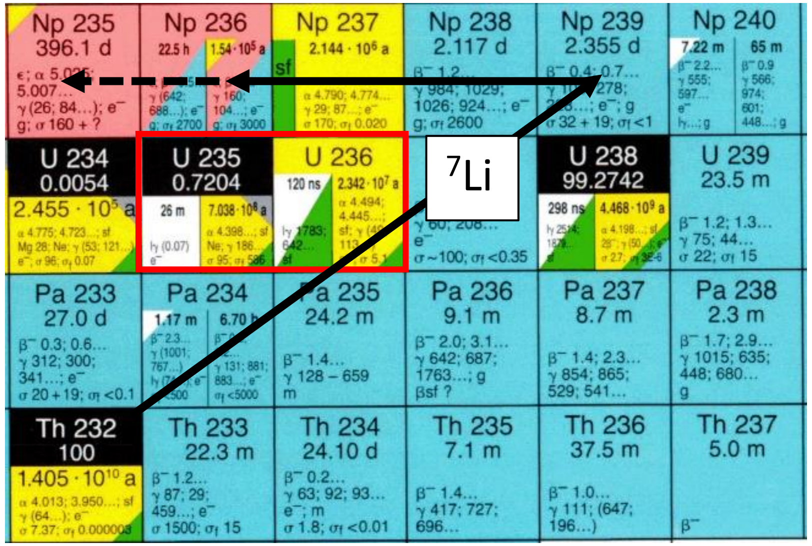
Lovett et al (1990): **c(Fe^{3+}) > 10 mg/L**
for quantitative co-precipitation of An(+V)

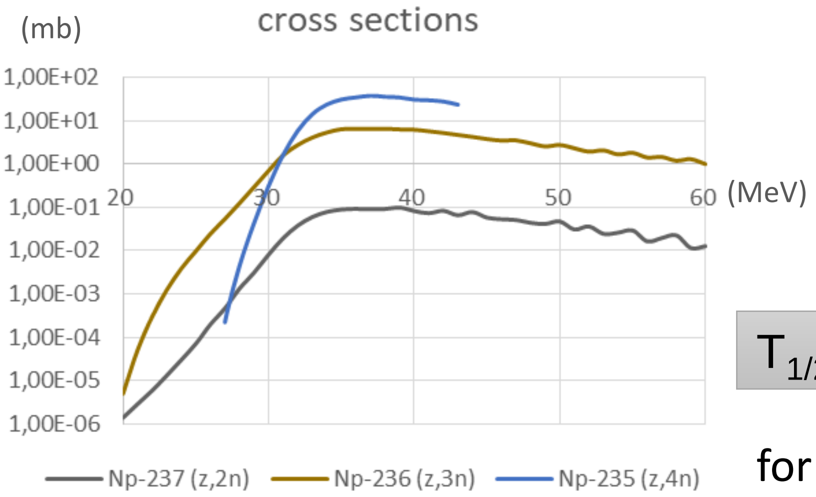
Northwest Pacific Ocean





SPIKE PRODUCTION FOR ^{237}Np



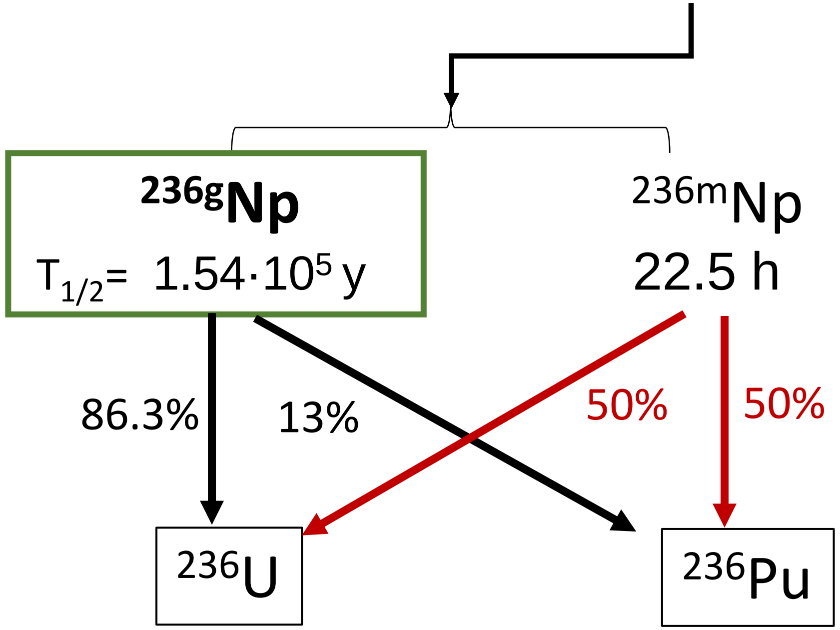


Calculated by EMPIREII code

$T_{1/2}$ (^{235}Np) = 396d!

for z = ^{7}Li

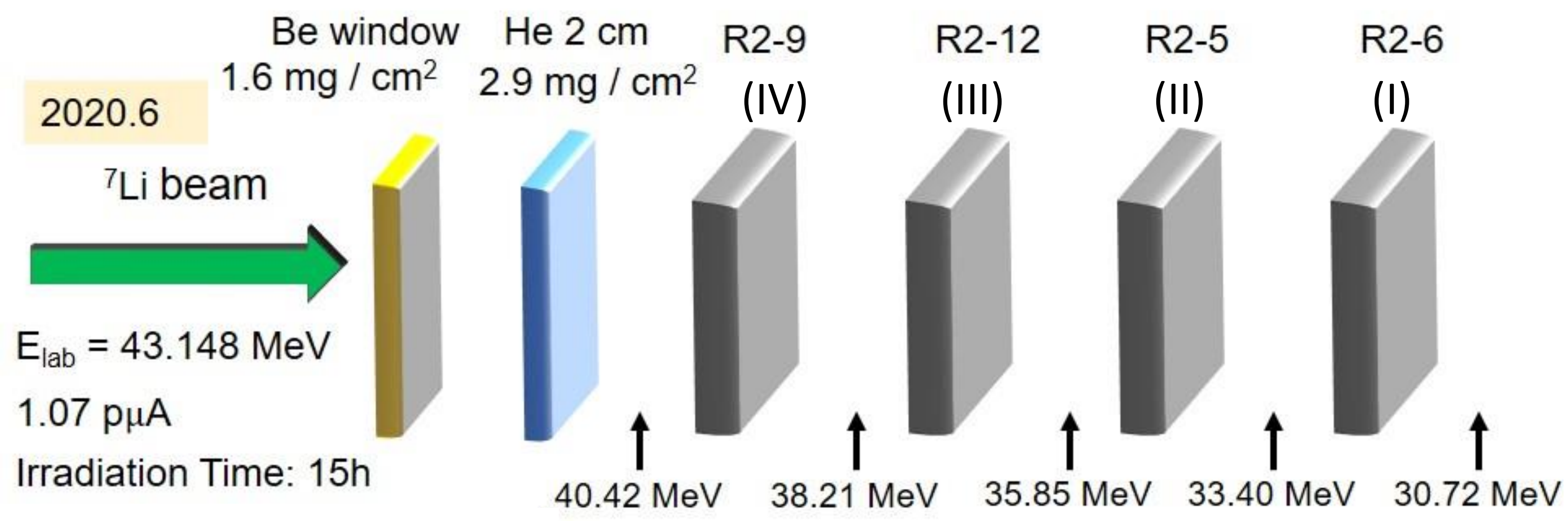
^{232}Th (^{7}Li , 3n) ^{236}Np

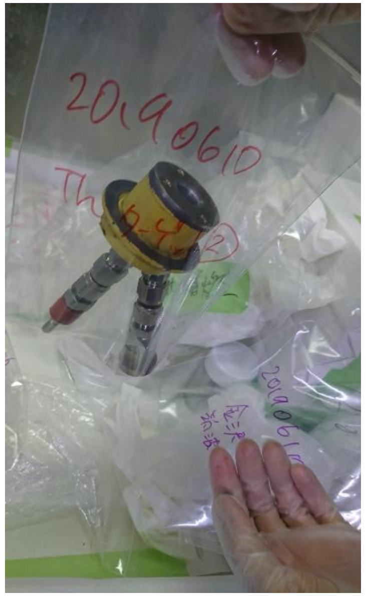


No experimental data available!

A. Sakaguchi,
University of Tsukuba

IRRADIATION @ RIKEN

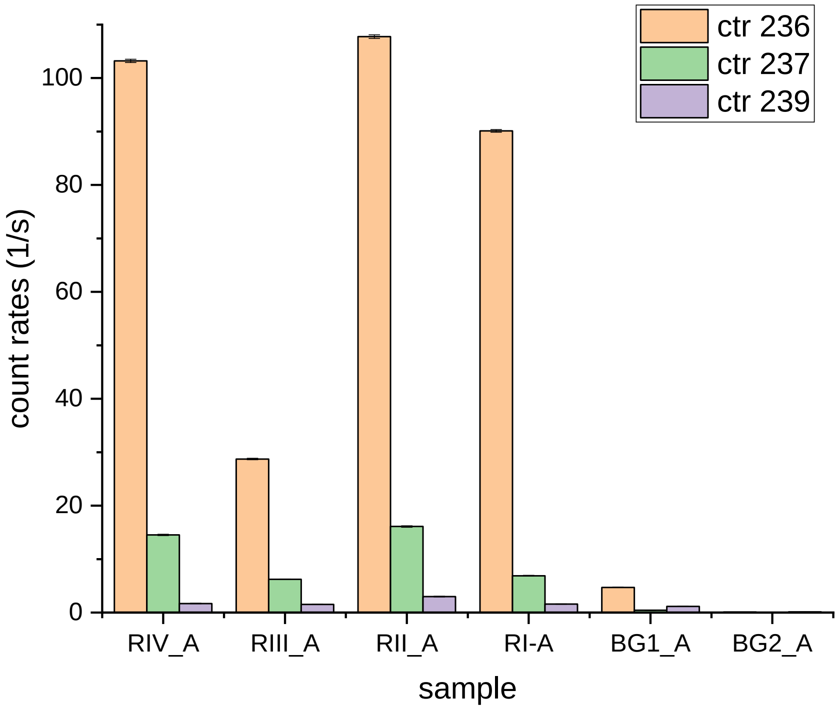




No.	Thickness (mg / cm^2)
R2 - 9	9.75
R2 - 12	10.15
R2 - 5	10.12
R2 - 6	10.58

A. Yokoyama,
Kanazawa University

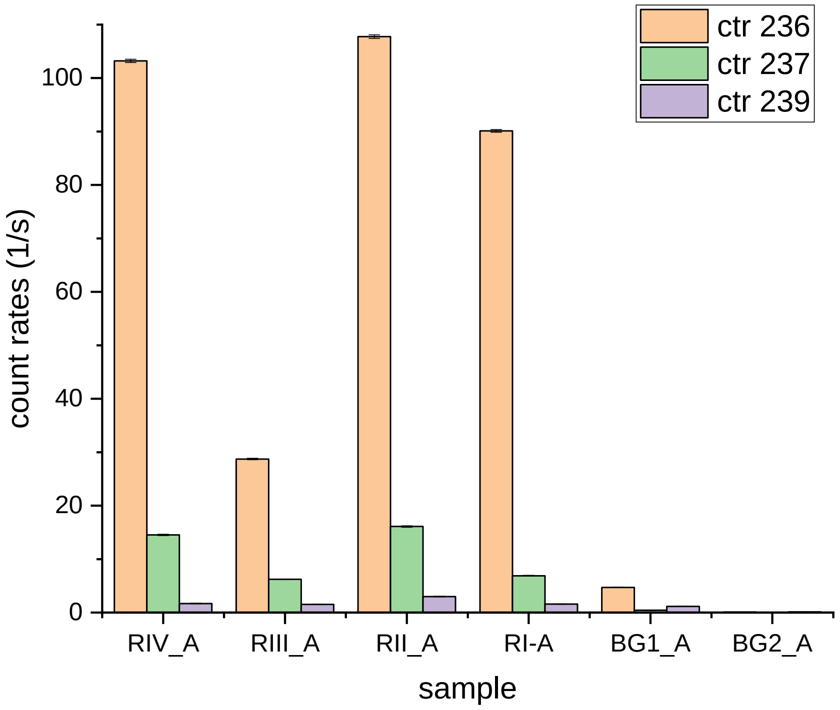
RESULTS FROM IRRADIATION

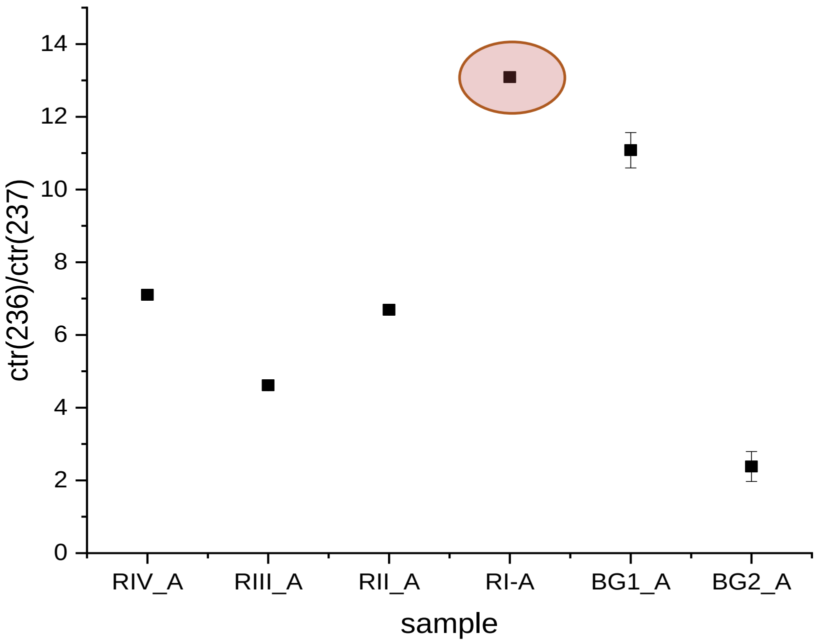


- **Mass 236 above BG level produced**
- Only statistical uncertainties included

RESULTS FROM IRRADIATION

Der Wissenschaftsfonds.





- Highest ratio 236/237 ≈ 13

↕

Empire Code: 236/237 ≈ 90

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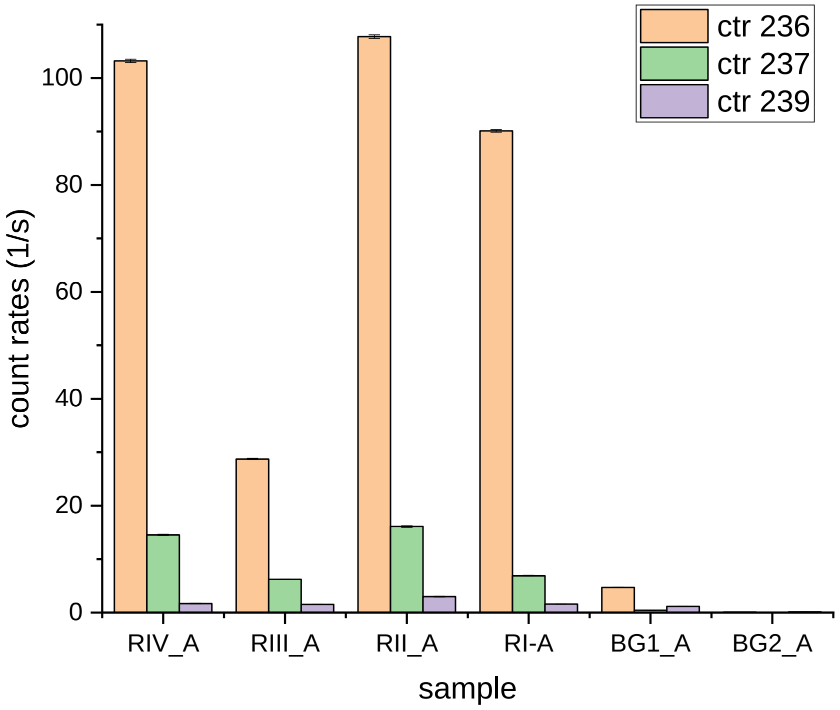
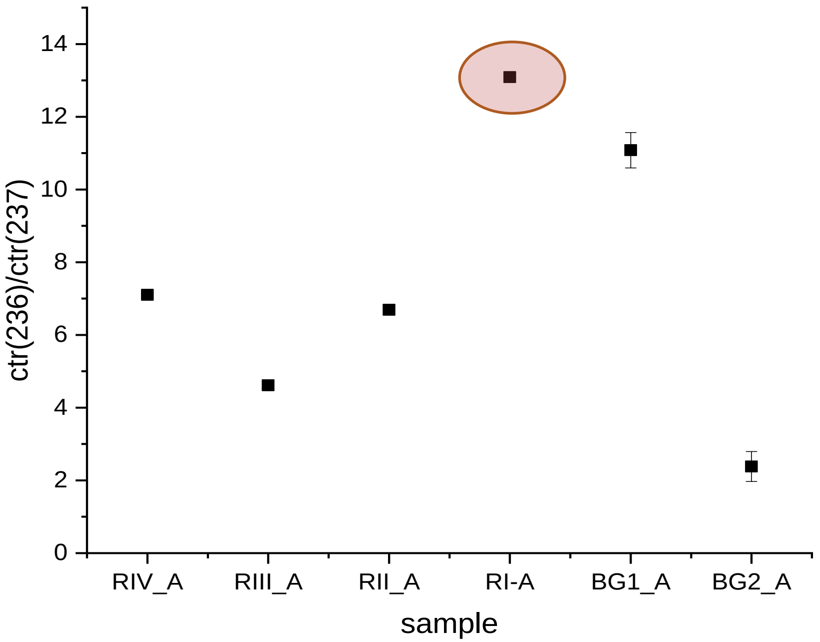
RESULTS FROM IRRADIATION





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↕

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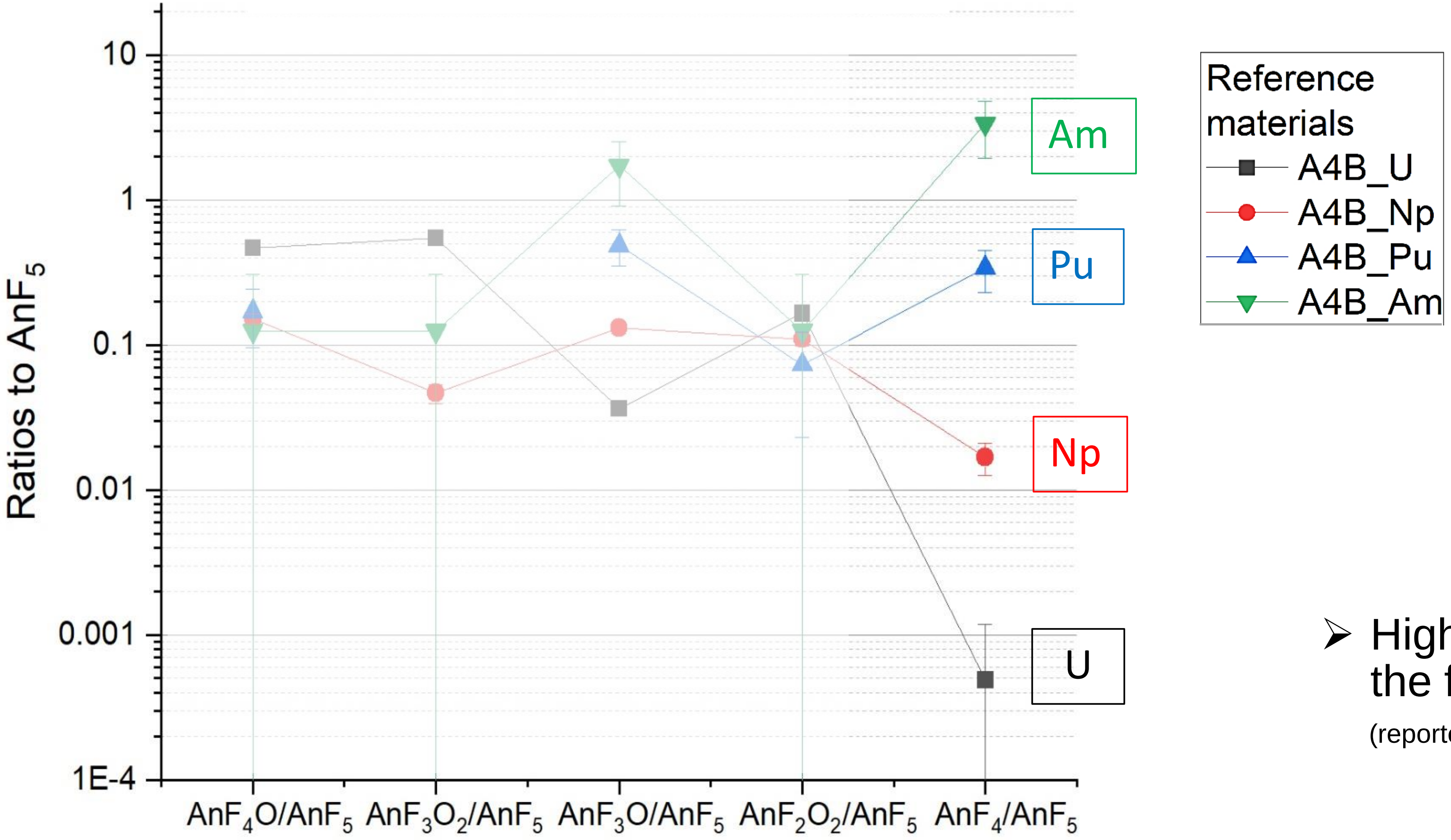
Problem: considerable ^{233}U production

Measured neutron flux (Au monitor)

thermal n flux: $6.5 \cdot 10^5/(scm^2)$
fast n flux: $2 \cdot 10^7/(scm^2)$

M(236) ≙ ^{236}Np?

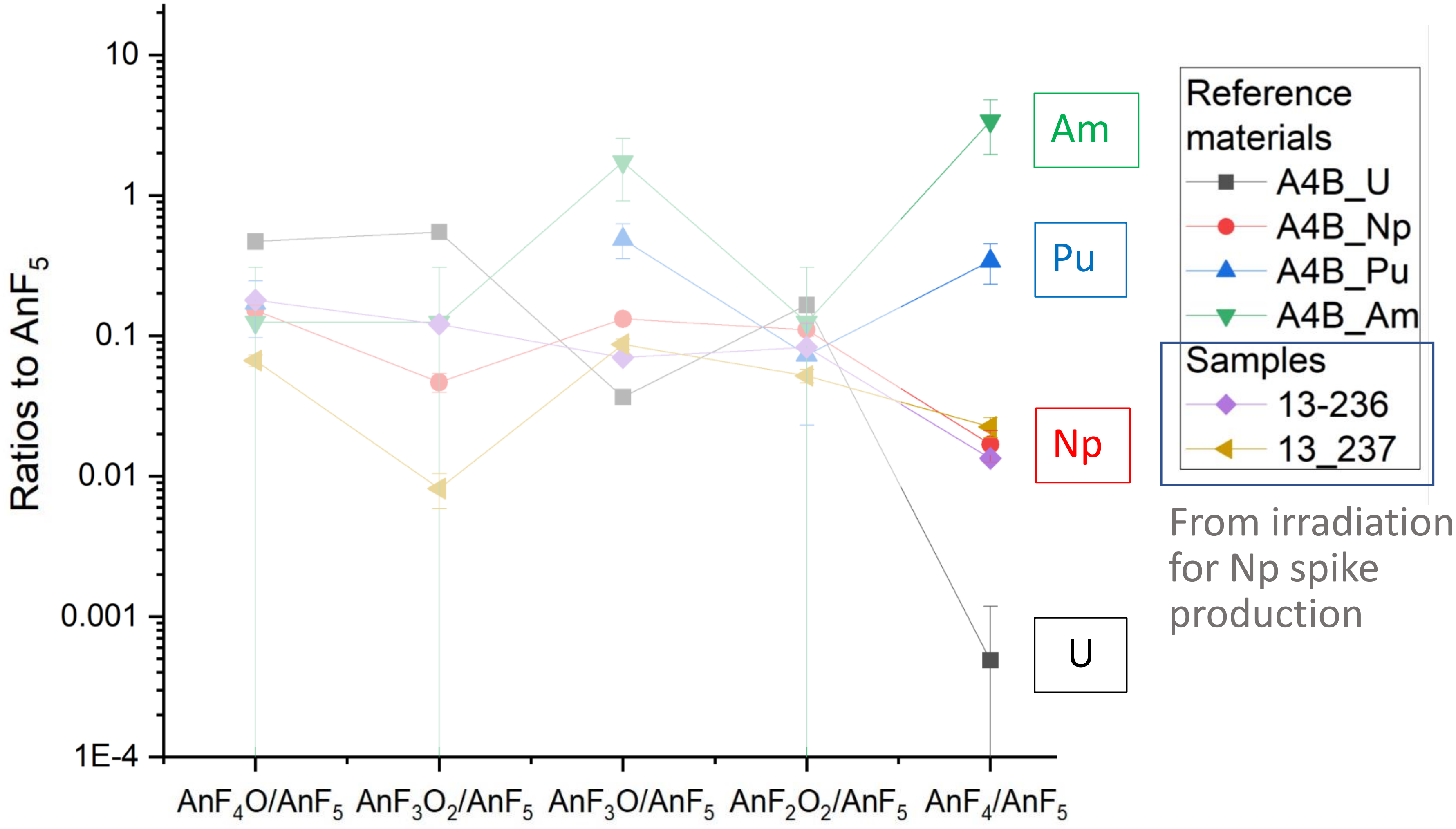
Relative formation of $AnF_mO_n^-$ with respect to $\mathbf{AnF_5^-}$



- Higher Actinides prefer the formation of AnF_4^-

(reported in Cornett et al, *NIMB,* 2015)

Relative formation of $AnF_mO_n^-$ with respect to $\mathbf{AnF_5^-}$

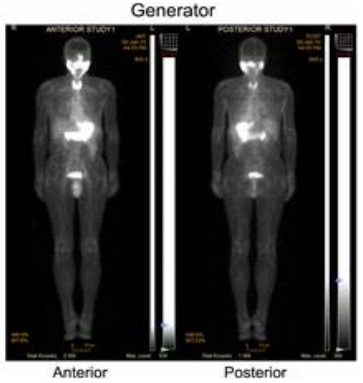


From irradiation for Np spike production

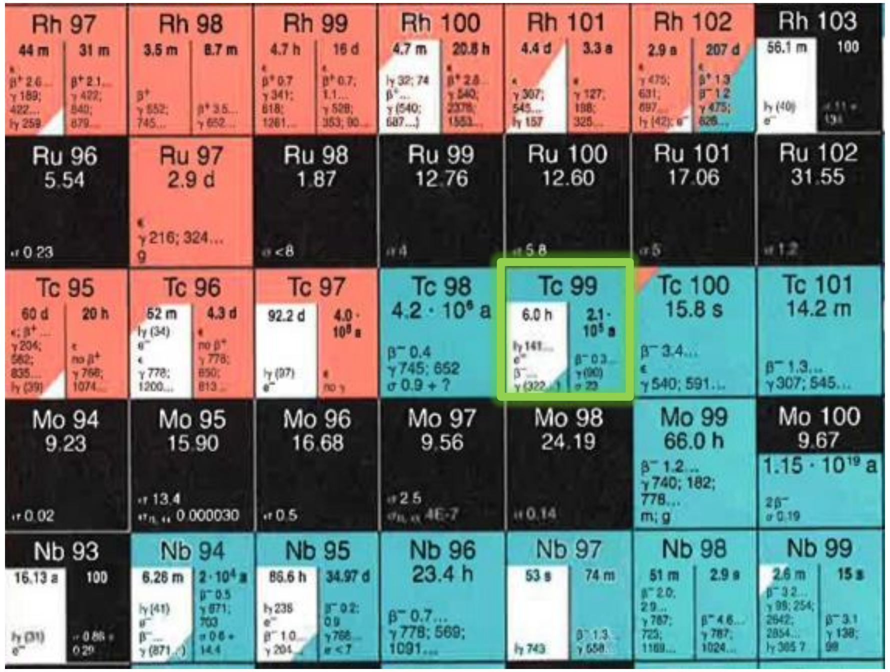
FWF Der Wissenschaftsfonds.

Nuclear reprocessing plants

(Nuclear Medicine)



This project

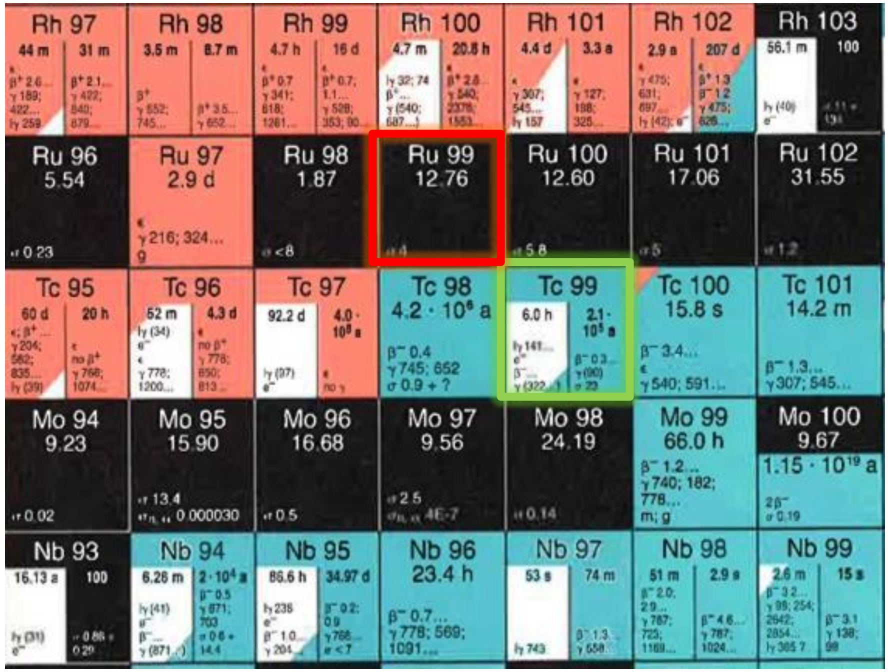


Atmospheric Nuclear Weapons Tests

Estimated total deposition: 140 TBq (220 kg)

DETECTION OF ^{99}Tc: ILIAMS

- Several proof-of-principle publications by TIMS, RIMS and AMS
- But hardly any studies on environmental concentrations far away from the contamination sources (100L, ICP-MS)



interfering isobar ^{99}Ru

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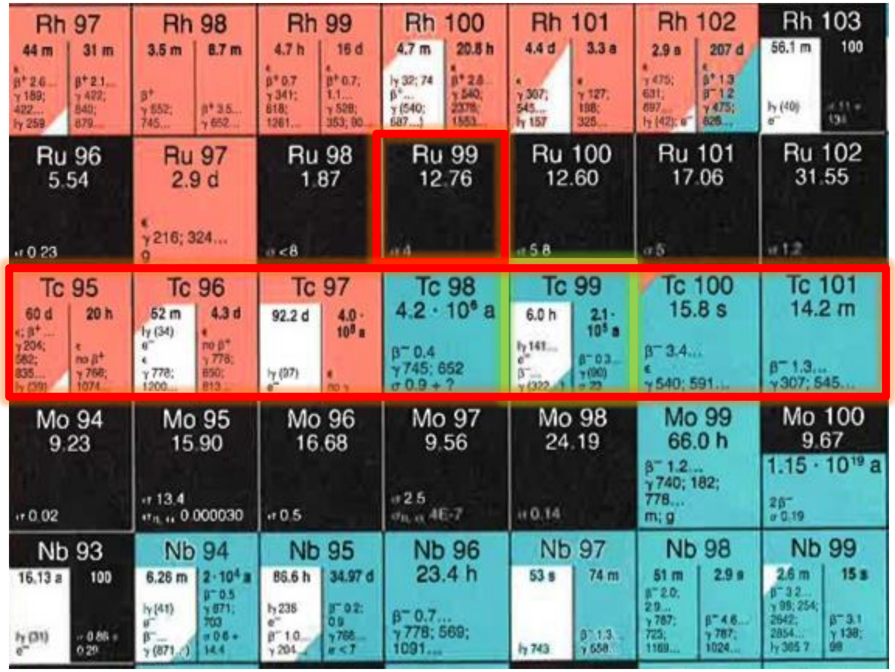
no stable isotopes for normalization



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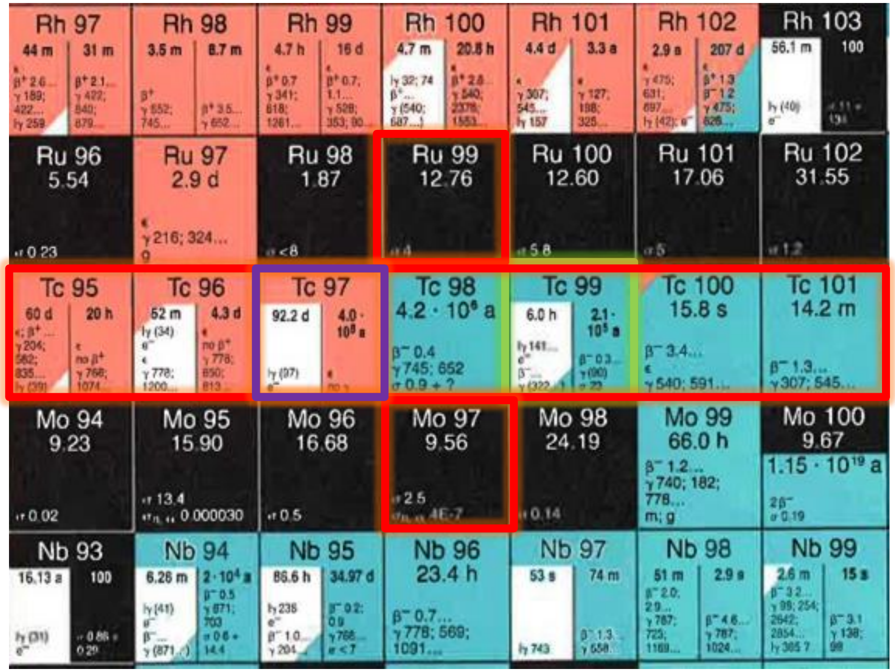
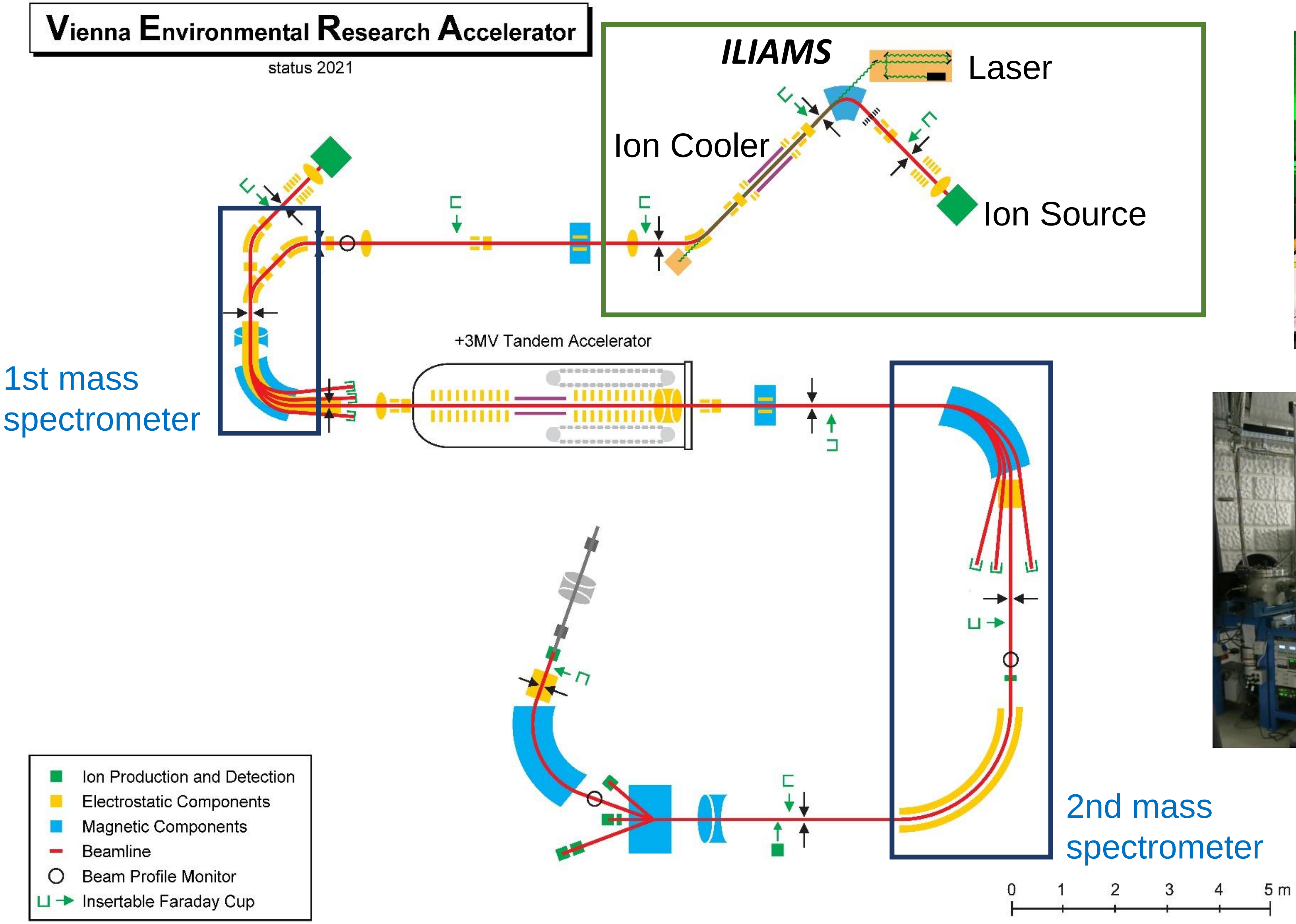


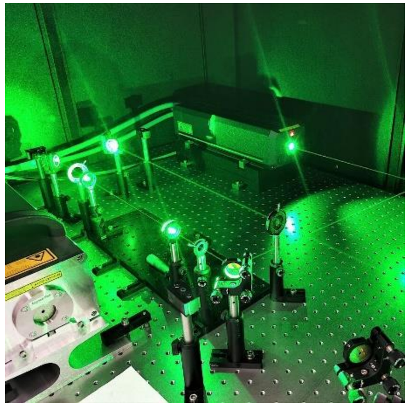
interfering isobar ^{99}Ru

use isotopic spike: **^{97}Tc** ⟶ Produced e.g. via **^{93}Nb(^{7}Li,3n)^{97}Tc**

@ 9.5 MV terminal voltage (MLL, Munich)

ILIAMS @ VERA


Vienna Environmental Research Accelerator
status 2021
ILIAMS
Laser
Ion Cooler
Ion Source
+3MV Tandem Accelerator
1st mass spectrometer
2nd mass spectrometer
Ion Production and Detection
Electrostatic Components
Magnetic Components
Beamline
Beam Profile Monitor
Insertable Faraday Cup
0 1 2 3 4 5 m

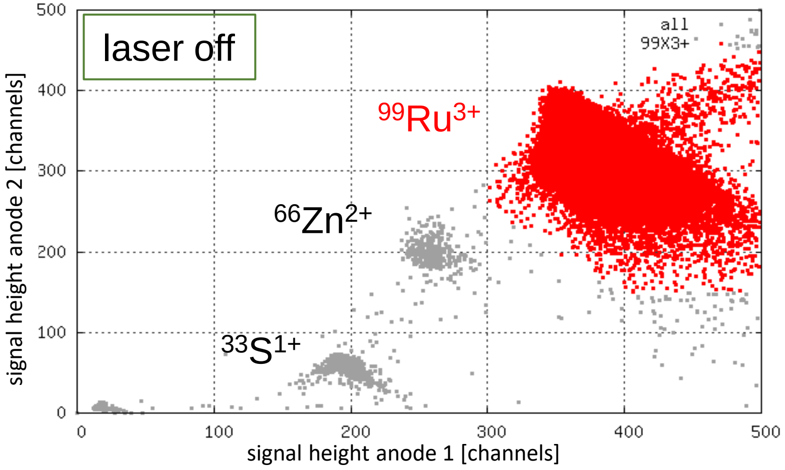



06/03/2017

M. Martschini, J. Pitters

FWF Der Wissenschaftsfonds.

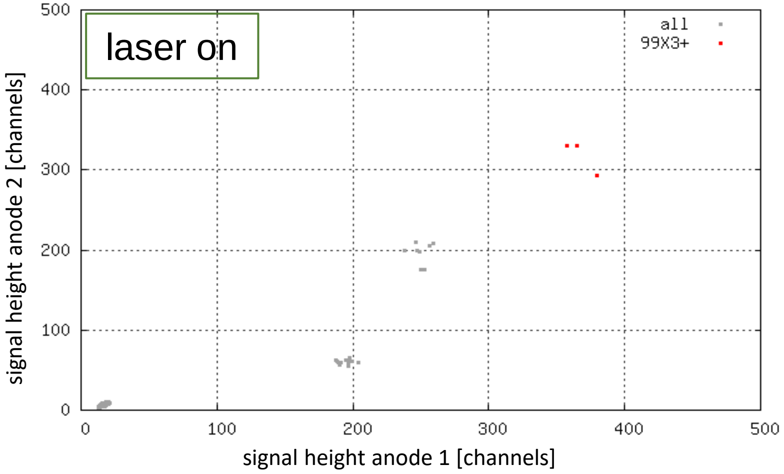
Sample with **7·10^14 atoms ^{99}Ru** added



Suppression factor

≈ **10^4-10^5**

Green laser on (5W transmitted power)



Ru SUPPRESSION

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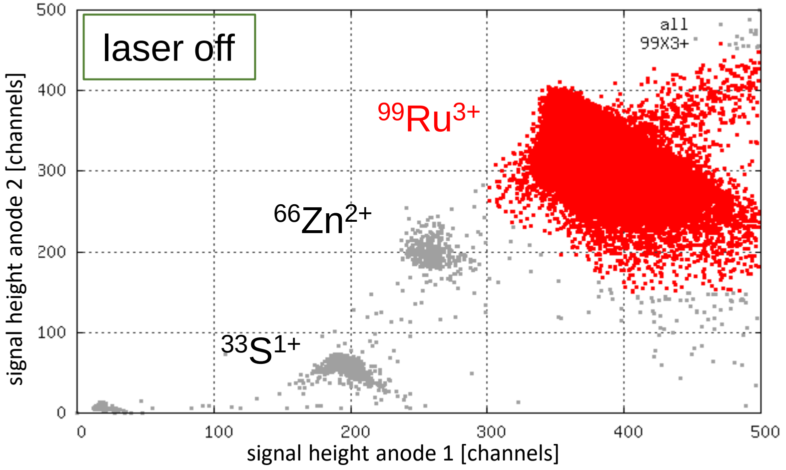
Der Wissenschaftsfonds.

Sample with **7·10^{14} atoms ^{99}Ru** added



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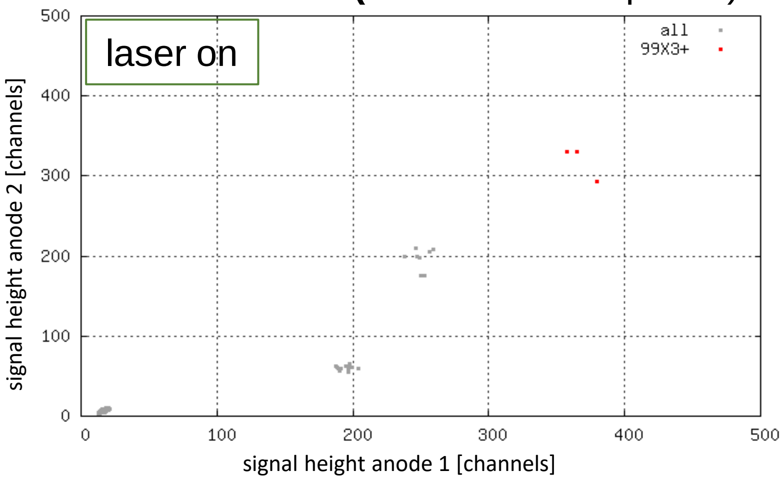
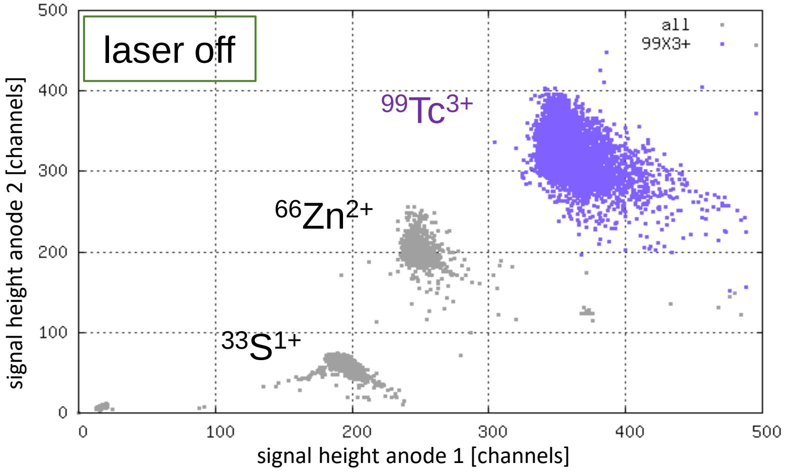
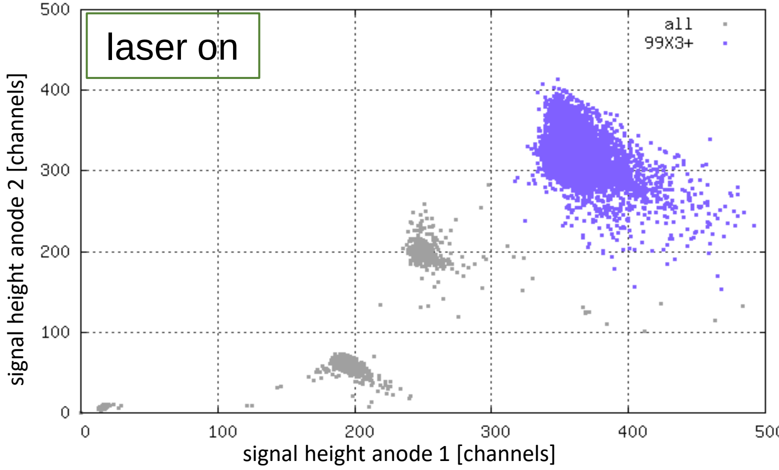


Sample with **4·10^{10} atoms ^{99}Tc** added



Green laser on (5W transmitted power)



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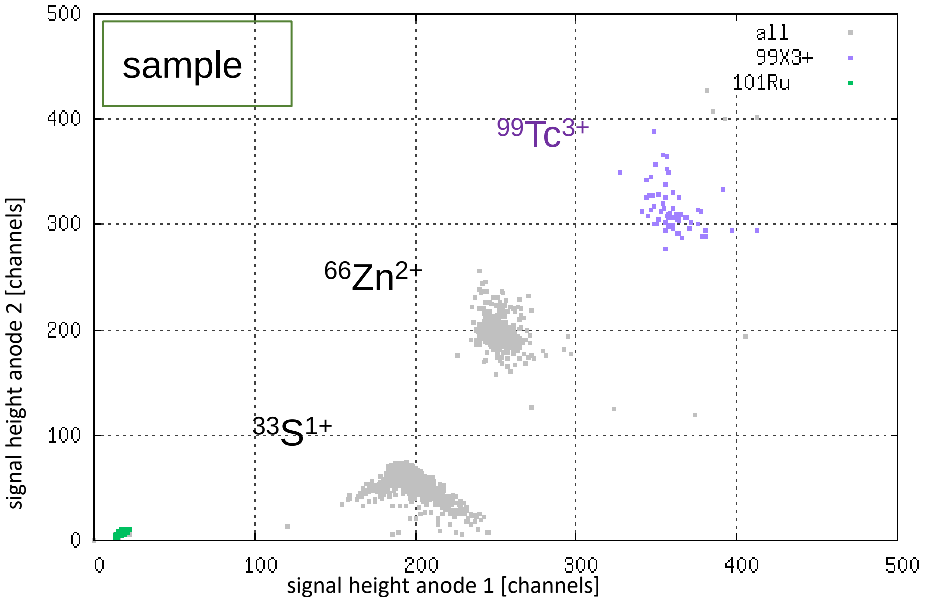
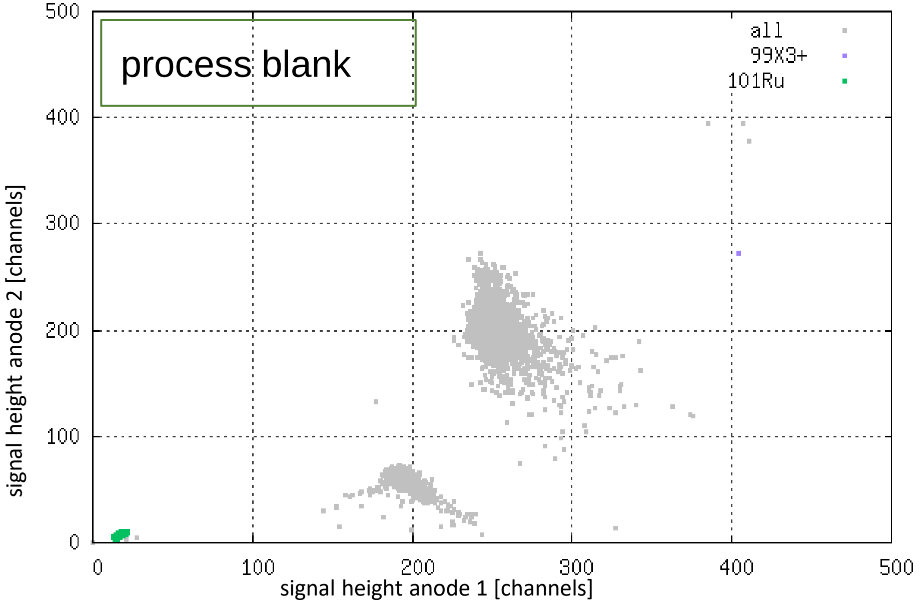
FWF Der Wissenschaftsfonds.

Ombrotrophic peat bog (Austria):
surface water (V = 10L)

Green laser on (5W transmitted power)





International Conference on Accelerators for Research and Sustainable Development #Accelerators2022 23–27 May 2022 IAEA, Vienna, Austria

SUMMARY: RESULTS ILIAMS

Helium as buffer gas

	Formation ion source	532 nm E = 2.33 eV	355 nm E = 3.40 eV	479 nm E = 2.79 eV
TcF_5^-		U ≈ 1	U ≈ 150 (?)	1
RuF_5^-	little	**U ≈ 10^5**	U > 100*	**U ≈ $4\cdot10^4$**
MoF_5^-	strong	U ≈ 20	U ≈ 250	U < 2.5
NbF_5^-	20 nA	U ≈ 1	U ≈ 1.2	1

* Upper limit:
no counts detected

- Normalization on ^{97}Tc will be challenging
  focus on Mo suppression
  (chemistry, other reactive gases, cooler settings)
- ✖ Normalization on Nb adds large uncertainties
  temporal behaviour of TcF_5^- differs from NbF_5^-
  Nb suppresses formation of TcF_5^-

Thank you

Acknowledgements

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FWF Der Wissenschaftsfonds.

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RADIATE