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Faculty of Physics - Isotope Physics

Accelerator Mass Spectrometry of ^{182}Hf and ^{135}Cs for nuclear astrophysics

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2nd ChETEC-Infra General Assembly

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Introduction

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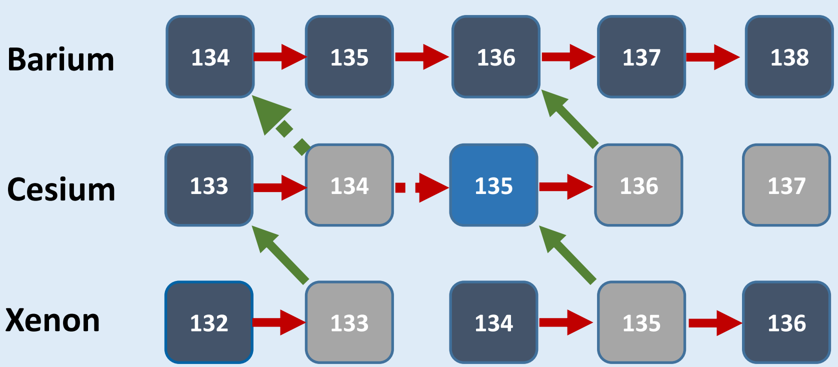
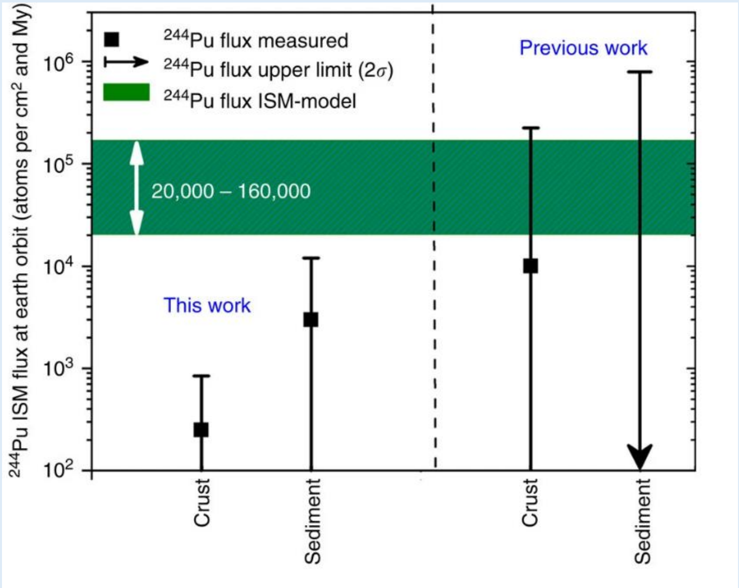
^{134}Cs is an important branching point in the **s-process** which defines the ^{134}Ba/^{136}Ba ratio (Li et al. 2021) by two parameters:

- The **beta-decay rate** at stellar temperatures
- The **^{134}Cs(n,γ)^{135}Cs** cross sections at keV energies

Detection limits of ^{135}Cs/^{133}Cs ≈ 10^{-14} needed

- Discrepancy of astrophysical sites for **r-process** following detection of live ^{60}Fe and ^{244}Pu in deep-sea reservoirs
- Detection of **live ^{182}Hf** ($T_{1/2}$ = 8.9 Ma) can help in solving this puzzle. No natural production on earth → live ^{182}Hf must stem from **recent nucleosynthesis**

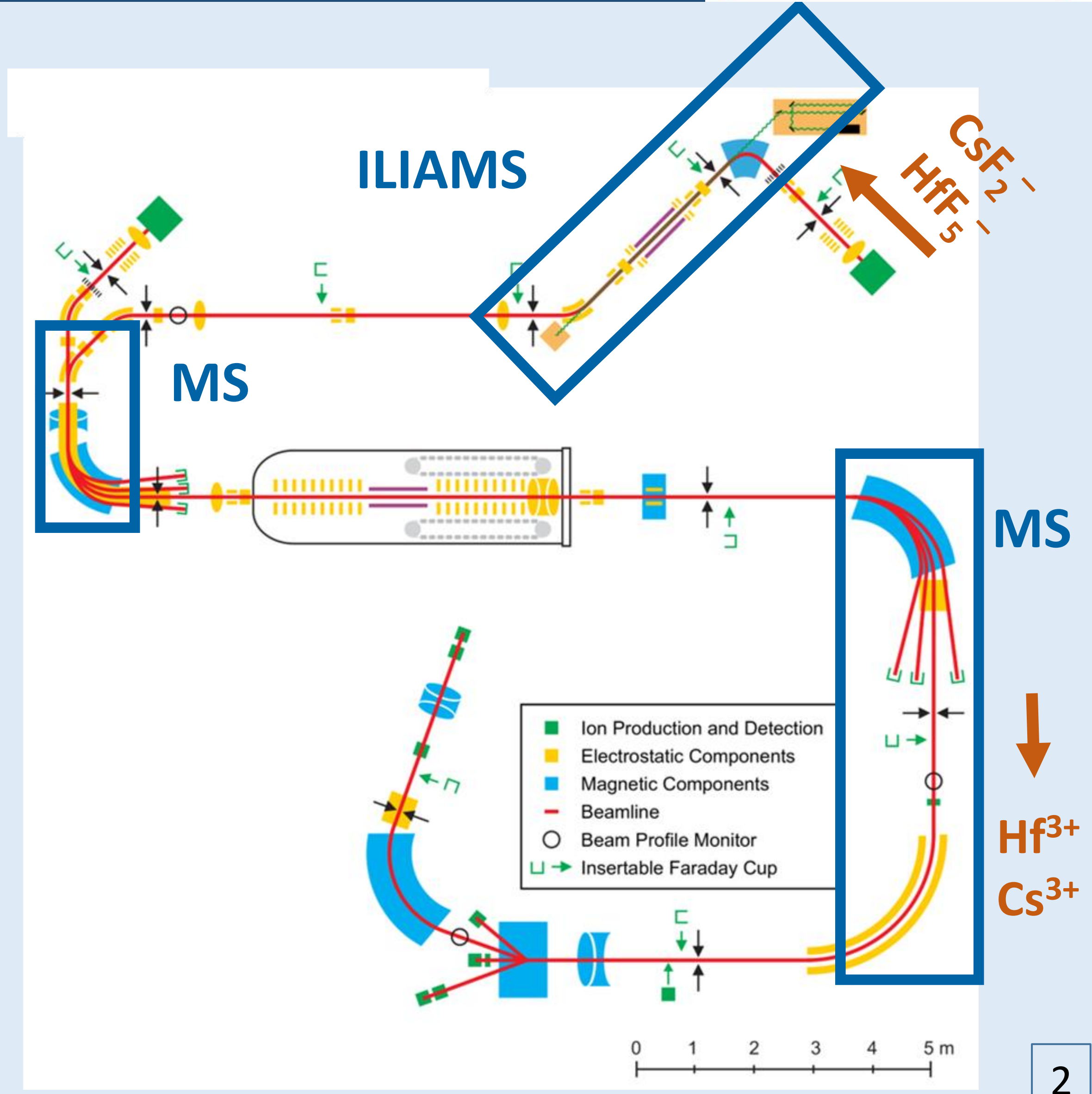




Detection via AMS at VERA

- Both nuclides (^{135}Cs & ^{182}Hf) have half-lifes in **Ma range and low abundances of 10^{-14}** need to be detected → **AMS** is the measurement method of choice

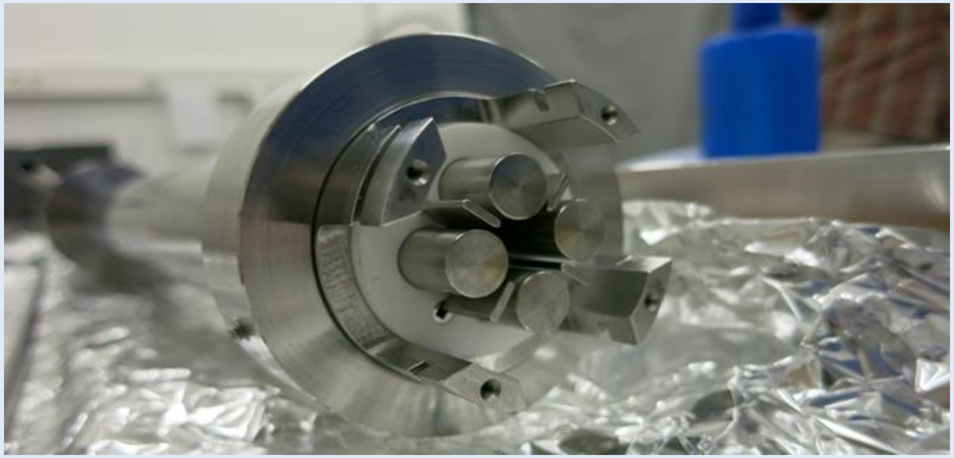
- **No separation of isobars** (^{135}Ba & ^{182}W) in classical AMS at this level possible → **ILIAMS setup** at VERA absolutely necessary for isobar suppression



ILIAMS setup

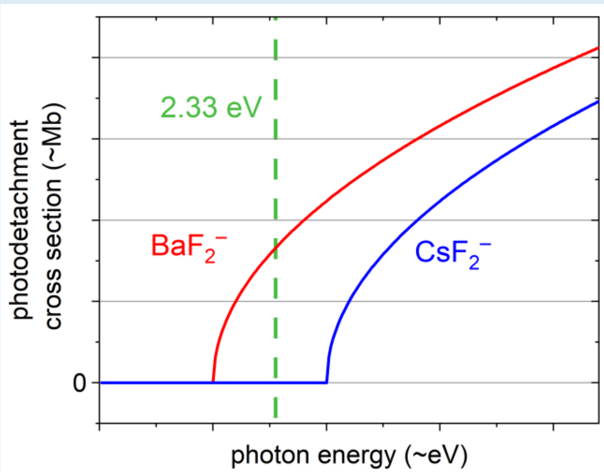
- ILIAMS is a **buffer-gas** filled RFQ where 30 keV anions are slowed to **near-thermal energies** and are collinearly overlapped with a laser beam → Low-energy isobar suppression

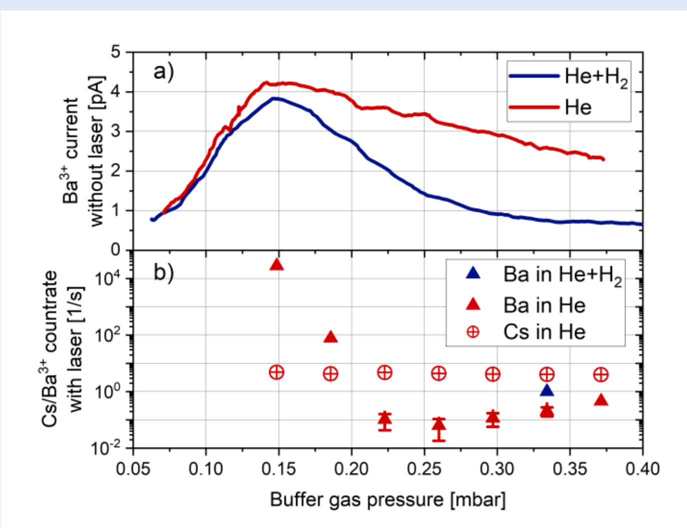
- It can act as an
  - ion cooler to **extend the interaction time between ion beam and laser beam** for laser photodetachment ($A^{-} + \gamma \rightarrow A + e^{-}$) or
  - as a **gas reaction cell** to suppress interfering isobars by molecular transitions



ILIAMS for laser photodetachment

- ^{135}Cs is extracted as CsF_2^- because it makes suppression of BaF_2^- with a green laser achievable





- Barium reduction by Ion-Laser interaction by 10^7 **and Cs stays unaffected!**
- BaF_2^- has a lower ionization efficiency by factor 10
- Measurements on Ba-spiked material give isobaric **suppressions > 10^8** at VERA and make detection limits of $^{135}Cs/^{133}Cs \approx 10^{-11}$ with ILIAMS possible
- Cross contamination in the ion source prevent us from reaching lower ratios

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ILIAMS as a gas reaction cell

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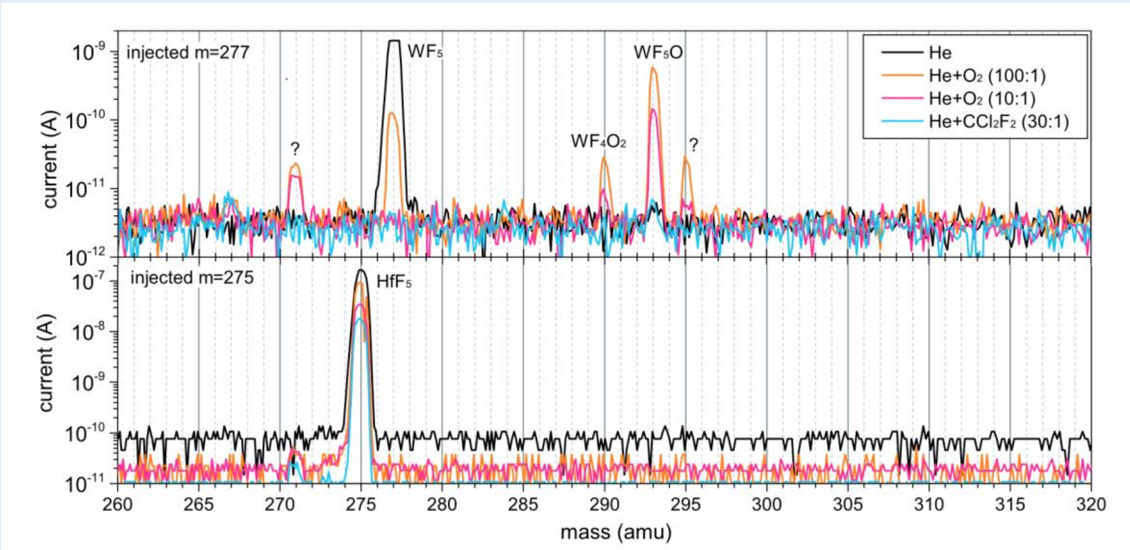
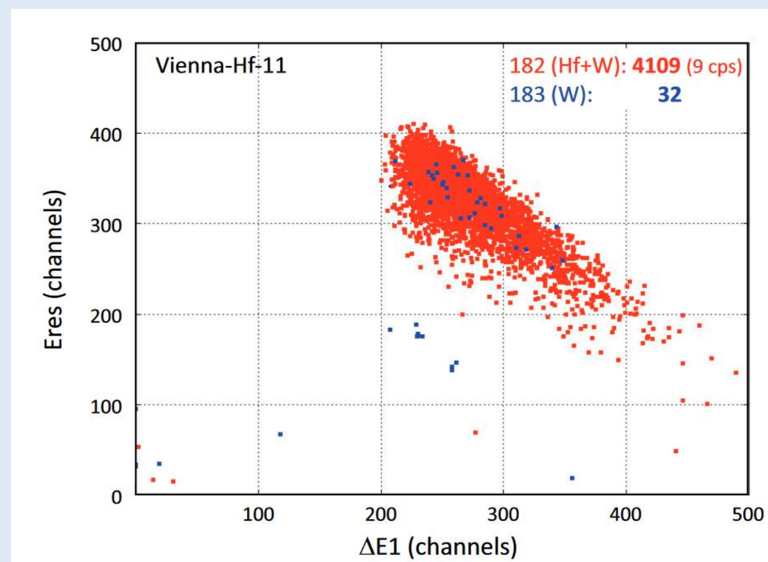


- ➢ ^{182}Hf is extracted as $^{182}HfF_5^-$ since $^{182}WF_5^-$ can be suppressed by a **molecular transition** $^{182}WF_5^- \rightarrow$ $^{182}WF_5O^-$ & $^{182}WF_4O_2^-$ with oxygen-containing buffer gas

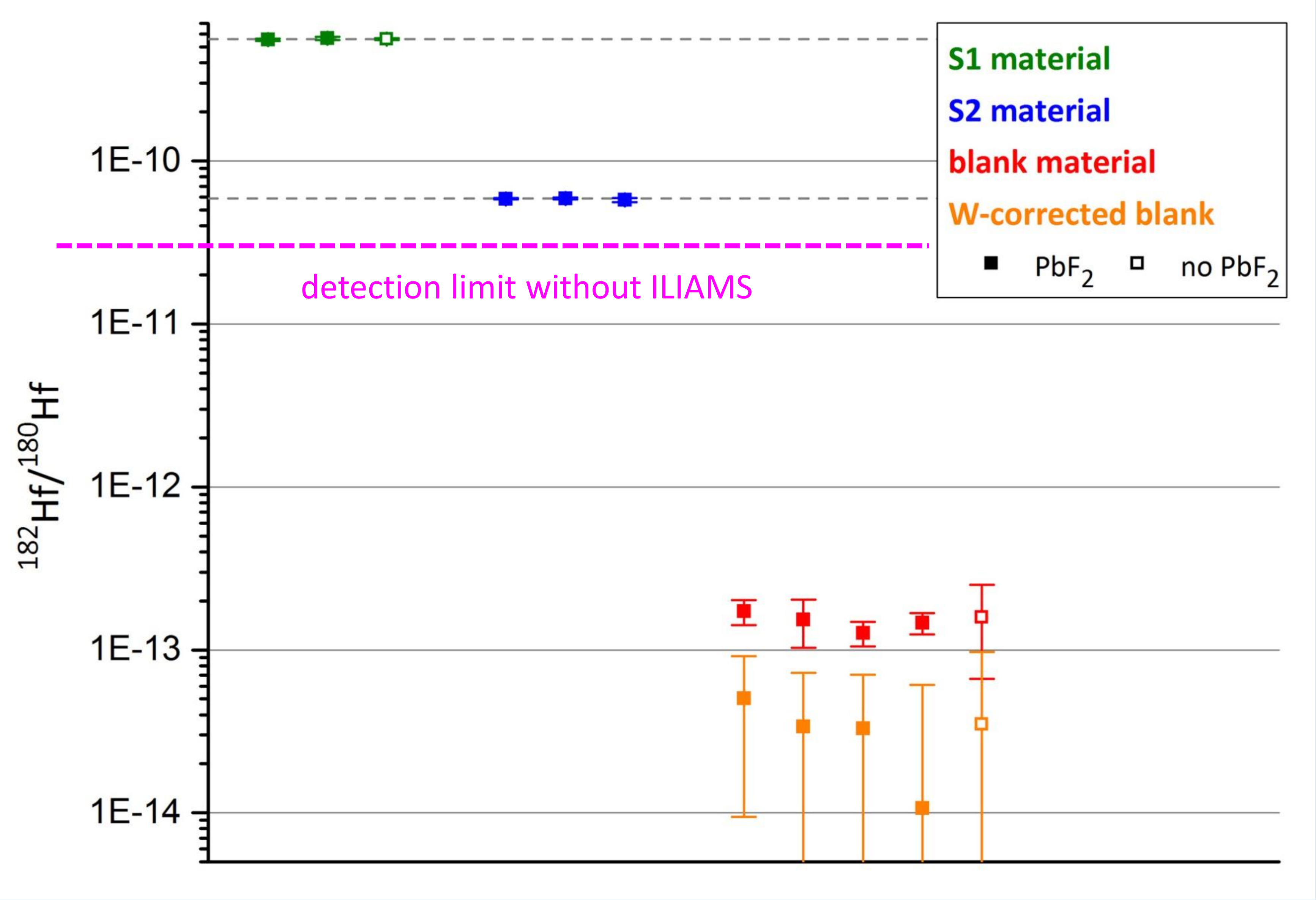
- ➢ Reproducibility on standard material < 5% with detection limit of **$^{182}Hf/^{180}Hf \approx 6\cdot10^{-14}$**
- ➢ as $^{182}W^{3+}$ and $^{182}Hf^{3+}$ are not separable in GIC → monitoring $^{183}W^{3+}$
- ➢ **Limited by ^{182}W suppression** in the gas reaction cell → Cross contamination in the ion source was not observed above this background level



Conclusion

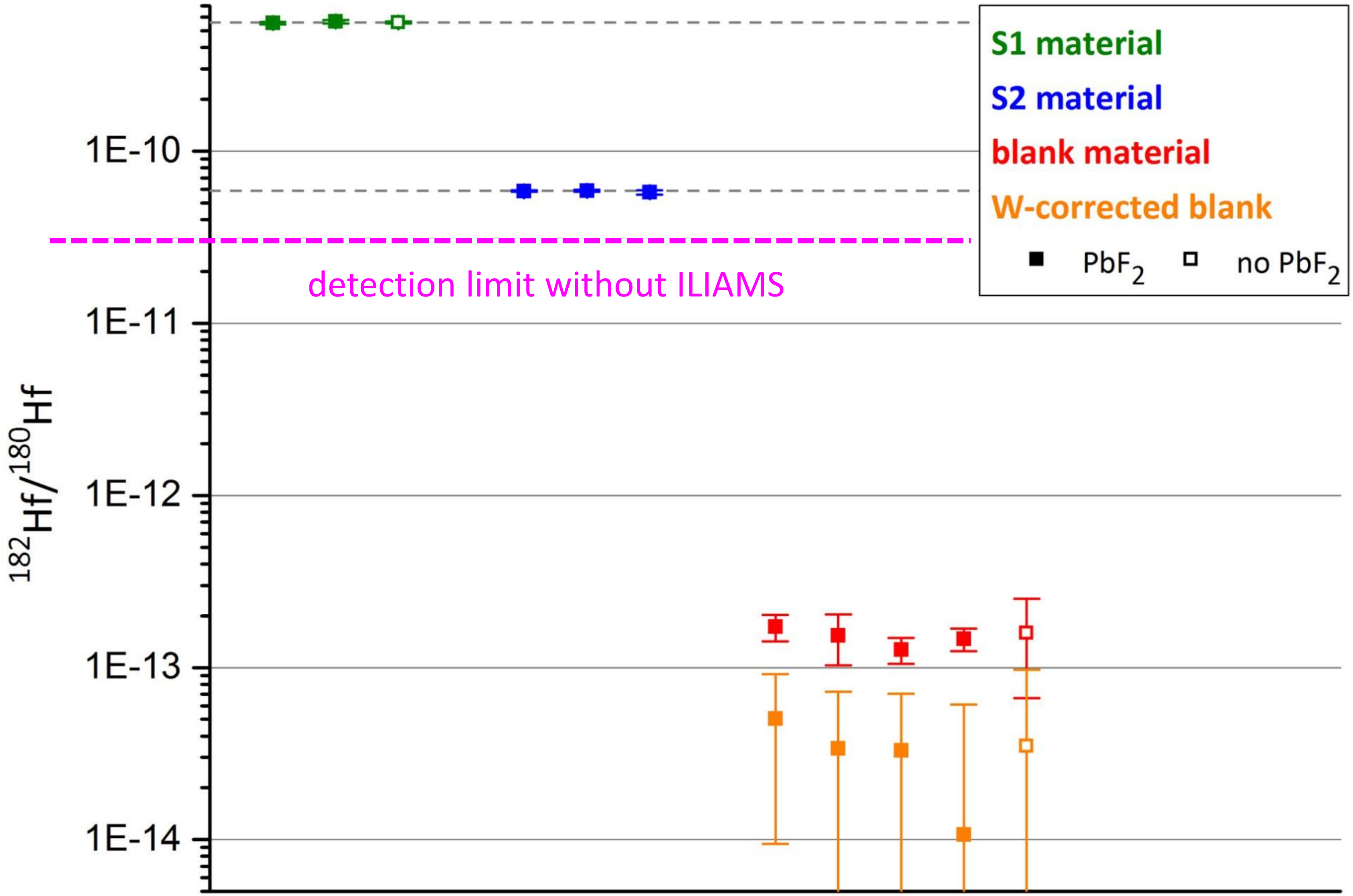
- The detection of both ^{135}Cs for neutron capture cross section measurements and live ^{182}Hf in deep sea archives is under development at VERA with the **ILIAMS setup**
- For both nuclides abundance sensitivities of 10^{-14} are needed
- For Cs, **cross contamination** and low ion source output prevent measuring lower isotopic ratios than 10^{-11}
- For Hf, the **W suppression** by molecular transitions in the ion cooler is limiting the blank values
- For both nuclides, AMS at VERA provides the **most sensitive measurement method**, still the detection limits need to be improved to detect ^{182}Hf also in **low-signal r-process scenarios** and ^{135}Cs in irradiated ^{133}Cs samples

Ab hier backupslides



S1 material
S2 material
blank material
W-corrected blank
PbF2
no PbF2
detection limit without ILIAMS
1E-10
1E-11
1E-12
1E-13
1E-14
182Hf/180Hf

^{182}Hf results



^{135}Cs and ^{137}Cs – Production and Motivation

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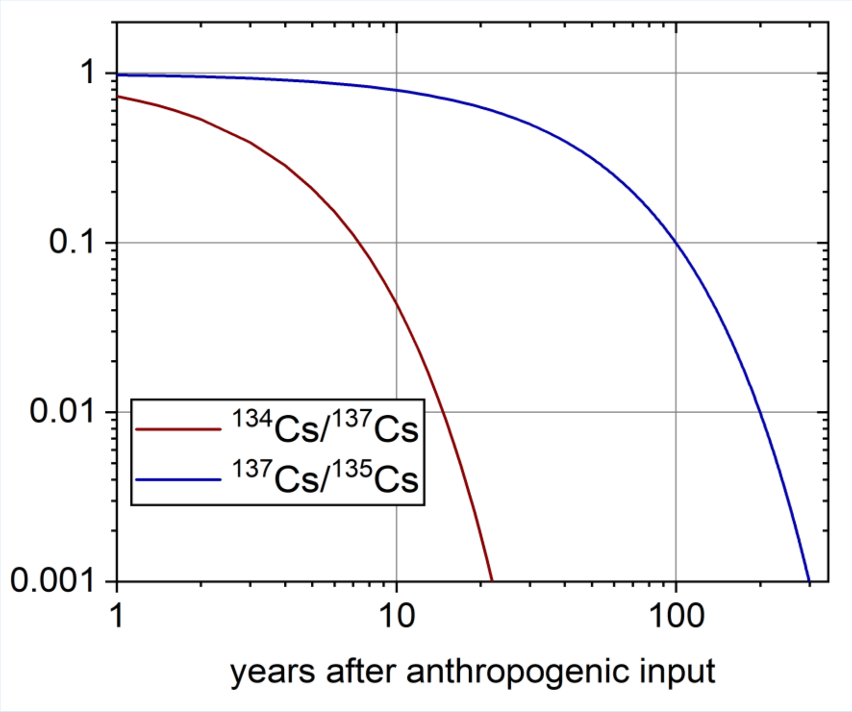
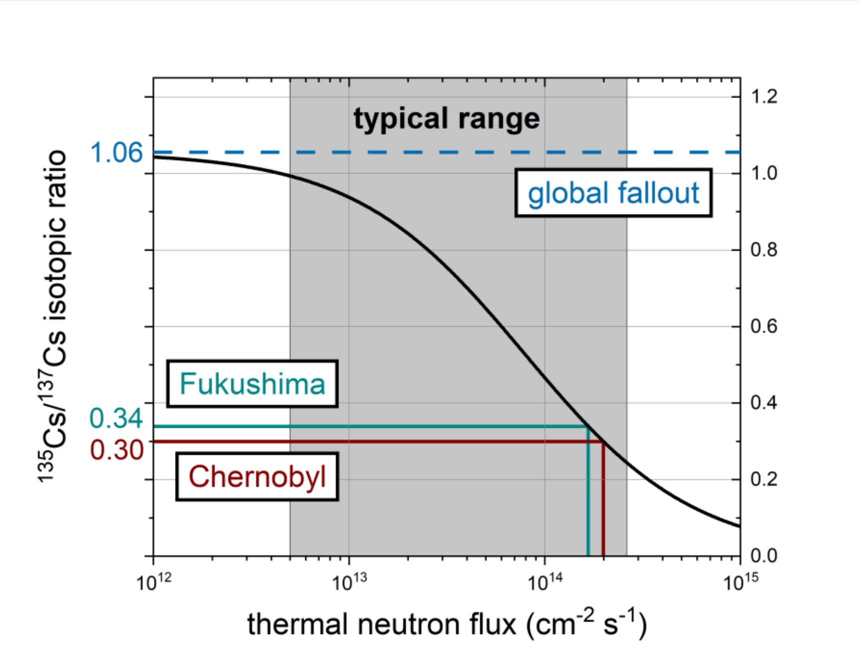
- γ detection of ^{134}Cs ($T_{1/2}$=2.06 yrs) is **half-life limited** → unsuitable for long-term tracing
- ^{135}Cs is a pure beta-emitter with long half-life → radiometric measurement hardly possible

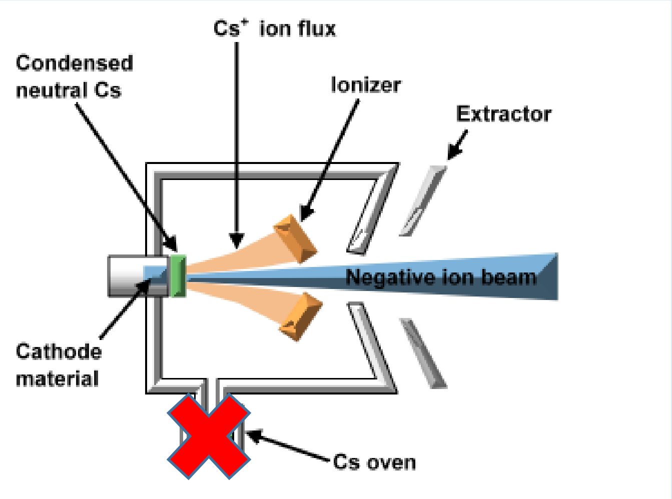
- First results on TIMS measurements give **consensus values on $^{135}Cs/^{137}Cs$** for Chernobyl and Fukushima [Bu et al., J. Anal. At. Spectrom. 2019]





AMS … or how it's normally done

- MC SNICS: Cs^+ sputtering to get suitable negative ions → Rb^+ or „self-sustainable" sputtering process

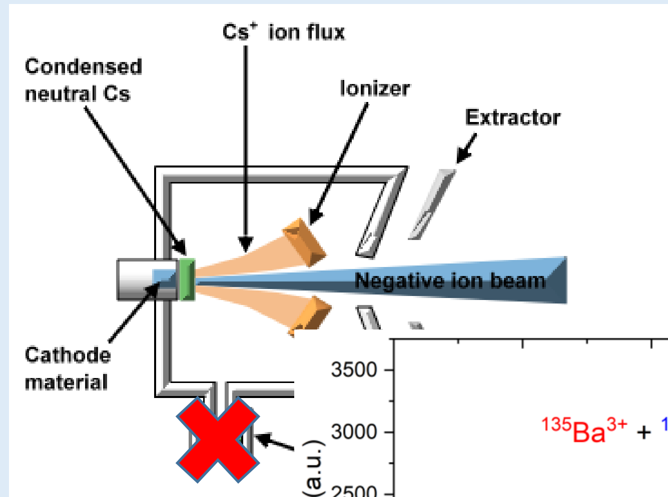


[https://tandem.nipne.ro/9MV_Pelletron.php]

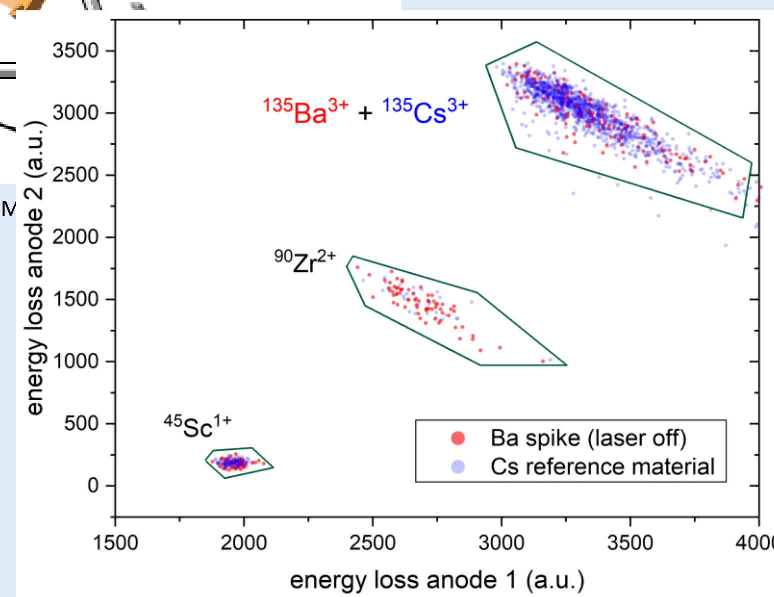
AMS … or how it's normally done

- MC SNICS: Cs^+ sputtering to get suitable negative ions → Rb^+ or „self-sustainable" sputtering process

- Elemental analysis methods in the gas-filled ionization chamber → complete **suppression of Ba** is necessary



[https://tandem.nipne.ro/9M



ILIAMS (Ion-Laser InterAction Mass Spectrometry)

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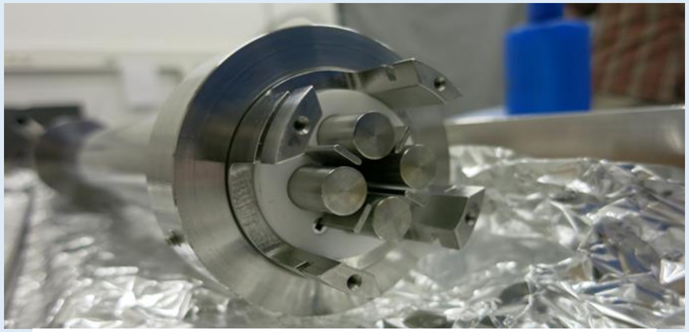
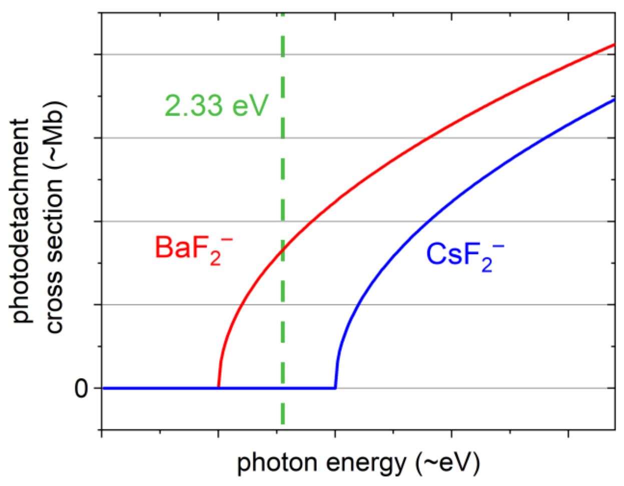
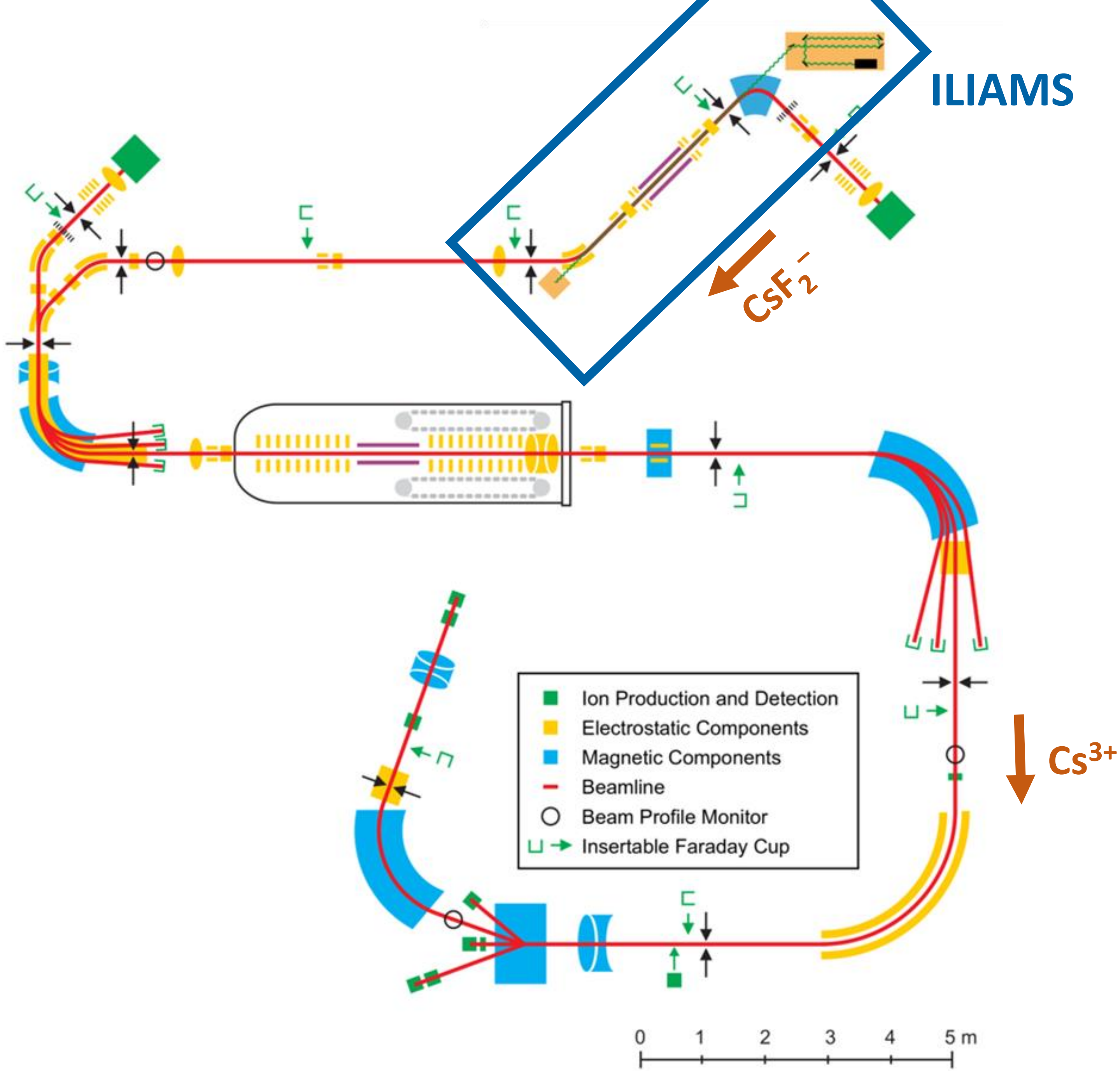
- buffer-gas filled RF quadrupole
- ion beam is overlapped with a **laser beam of suitable energy**
- non-resonant neutralization of anions with electron affinities < photon energy





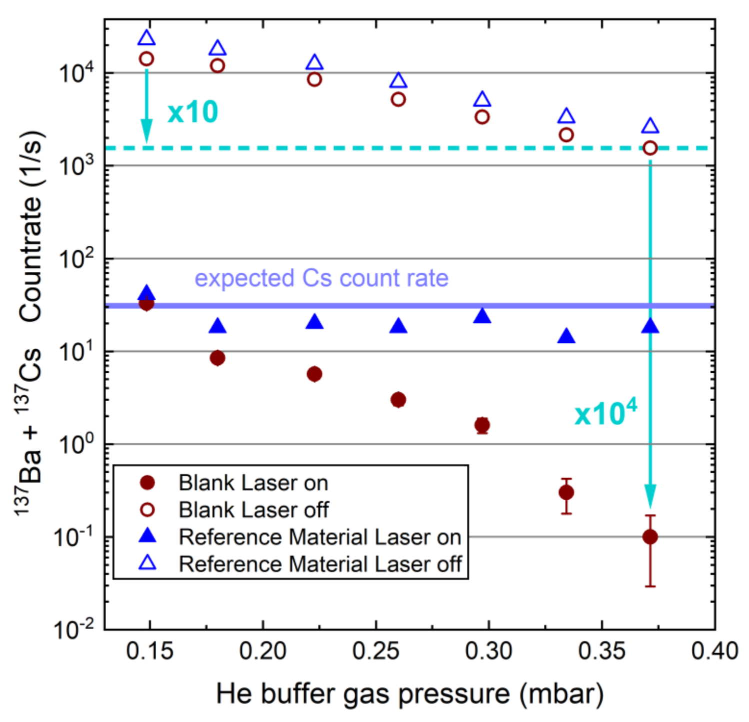
- electrostatic deceleration to ≈ 30 eV
- buffer gas cooling down to ≈ 1 eV
- RF quadrupole field keeps ions on track
- ion residence times: 1-10 ms



ILIAMS
CsF_2^-
Cs^{3+}
Ion Production and Detection
Electrostatic Components
Magnetic Components
Beamline
Beam Profile Monitor
Insertable Faraday Cup
0
1
2
3
4
5 m

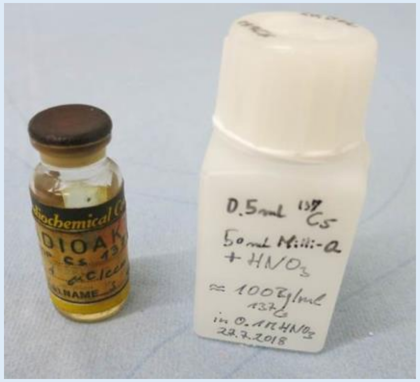
$^{135,137}CsF_2^-$ for isobar suppression with ILIAMS

Extracting **CsF_2^-** from the ion source makes suppression of Ba achievable with green (2.33 eV) laser.



- Ba reduced by collisions with the gas by 1×10^1
- Further reduction by Ion-Laser interaction by 1×10^4 **and Cs stays unaffected!**
- 30% ion cooler transmission, 25% accelerator transmission for Cs
- BaF_2^- has a lower ionization efficiency by factor 1×10^1
- Measurements on Ba-Spike give isobaric **suppressions > 10^6** at VERA

Reference materials



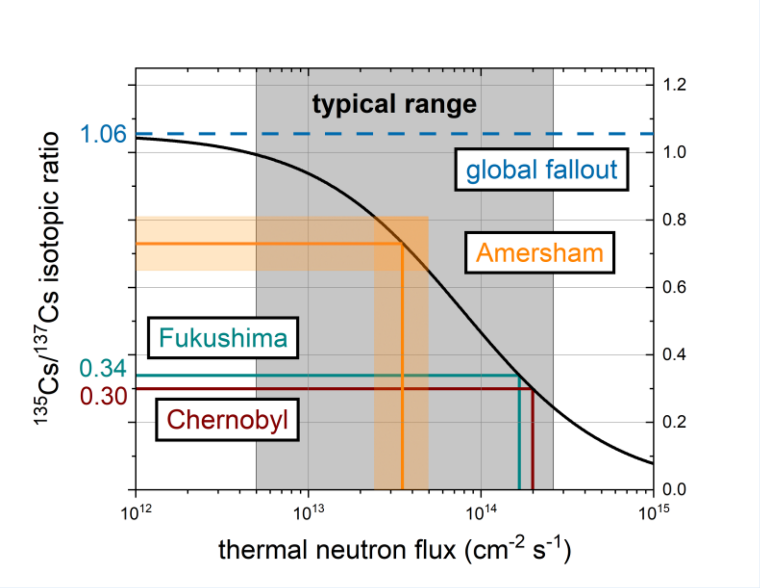
- No $^{135}Cs/^{133}Cs$ and no $^{135}Cs/^{137}Cs$ reference materials available

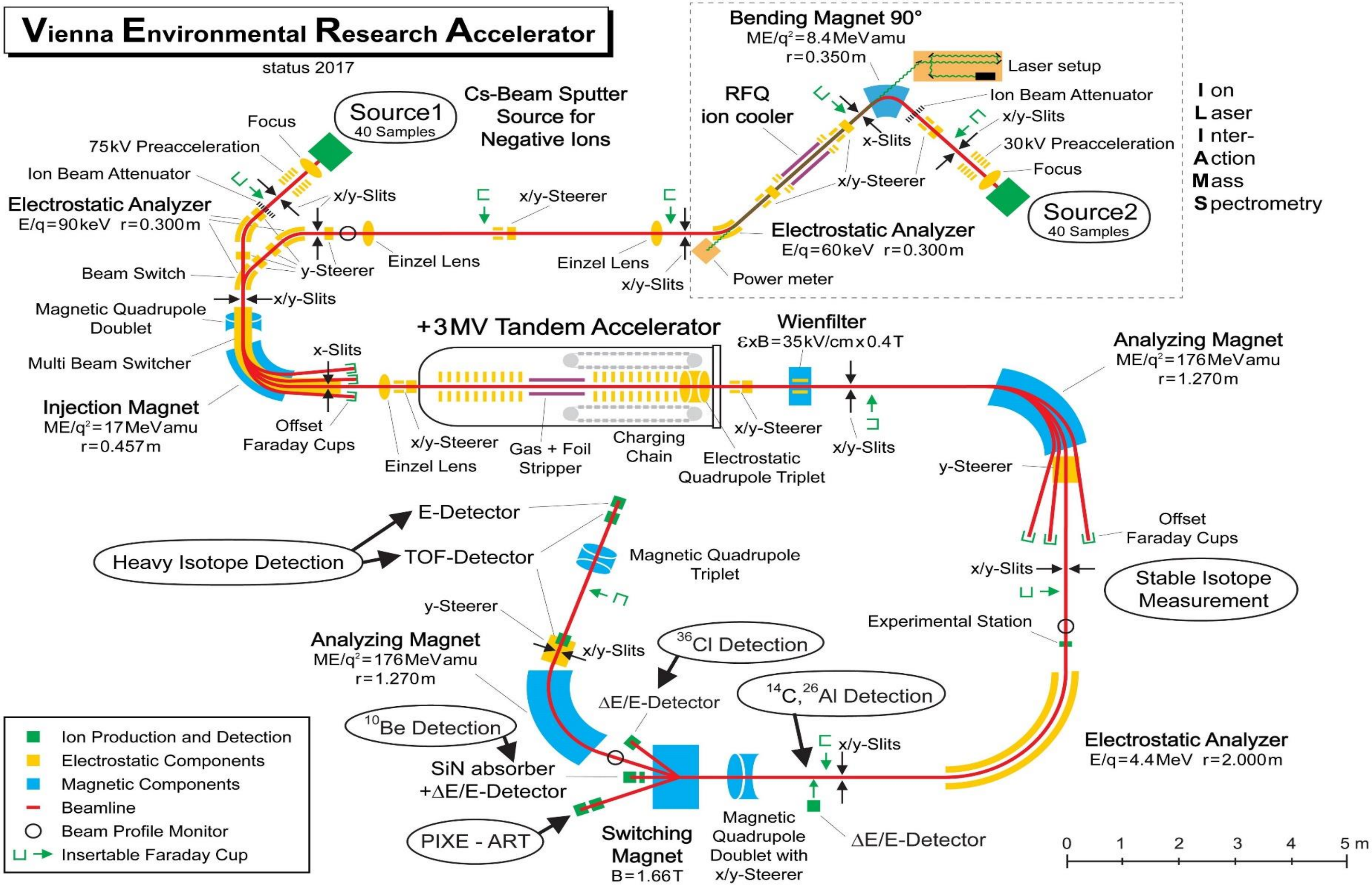
→ measurements relative to ICP-MS measured consensus values and in-house standard for $^{135}Cs/^{137}Cs$

- ^{137}Cs Amersham UK solution mixed with Cs_2SO_4-carrier and PbF_2 as reference material

- $^{135}Cs/^{137}Cs$ ratio not known a priori

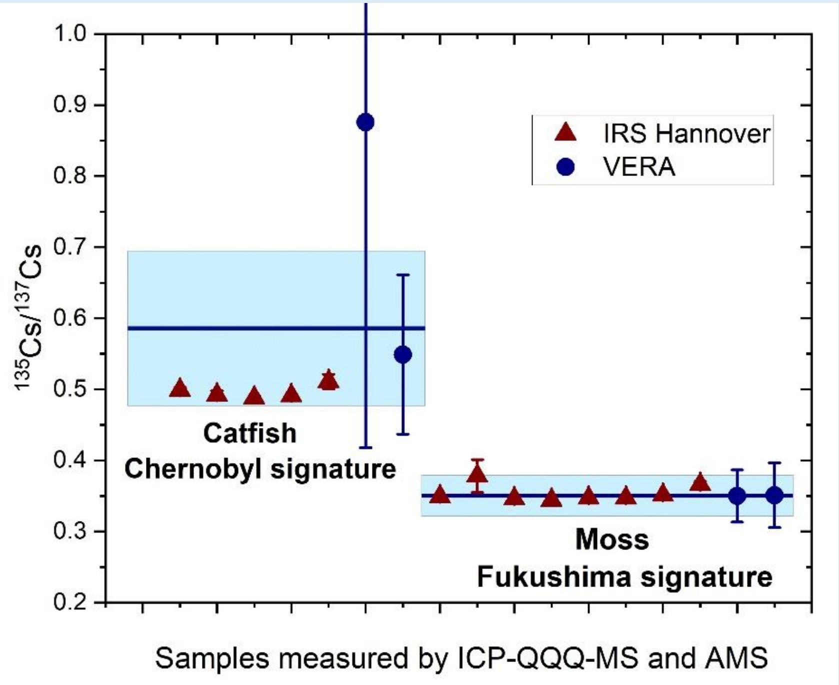
- First measurements at VERA give a ratio of **$^{135}Cs/^{137}Cs = 0.73 \pm 0.08$**





Vienna Environmental Research Accelerator
status 2017
Source1
40 Samples
Cs-Beam Sputter Source for Negative Ions
Focus
75kV Preacceleration
Ion Beam Attenuator
Electrostatic Analyzer
E/q=90keV r=0.300m
x/y-Slits
x/y-Steerer
y-Steerer
Einzel Lens
Beam Switch
x/y-Slits
Magnetic Quadrupole Doublet
Multi Beam Switcher
Injection Magnet
ME/q²=17MeVamu
r=0.457m
x-Slits
Offset Faraday Cups
Einzel Lens
x/y-Steerer
+3MV Tandem Accelerator
Gas + Foil Stripper
Charging Chain
Electrostatic Quadrupole Triplet
Wienfilter
εxB=35kV/cm x 0.4T
x/y-Steerer
x/y-Slits
Bending Magnet 90°
ME/q²=8.4MeVamu
r=0.350m
Laser setup
RFQ ion cooler
Ion Beam Attenuator
x/y-Slits
x-Slits
30kV Preacceleration
x/y-Steerer
Focus
Source2
40 Samples
Electrostatic Analyzer
E/q=60keV r=0.300m
Power meter
Ion Laser Inter-Action Mass Spectrometry
Analyzing Magnet
ME/q²=176MeVamu
r=1.270m
y-Steerer
Offset Faraday Cups
x/y-Slits
Stable Isotope Measurement
Experimental Station
Electrostatic Analyzer
E/q=4.4MeV r=2.000m
E-Detector
TOF-Detector
Heavy Isotope Detection
Magnetic Quadrupole Triplet
y-Steerer
x/y-Slits
Analyzing Magnet
ME/q²=176MeVamu
r=1.270m
36Cl Detection
ΔE/E-Detector
14C, 26Al Detection
x/y-Slits
10Be Detection
SiN absorber +ΔE/E-Detector
PIXE - ART
Switching Magnet
B=1.66T
Magnetic Quadrupole Doublet with x/y-Steerer
ΔE/E-Detector
Ion Production and Detection
Electrostatic Components
Magnetic Components
Beamline
Beam Profile Monitor
Insertable Faraday Cup
0 1 2 3 4 5 m

Environmental samples



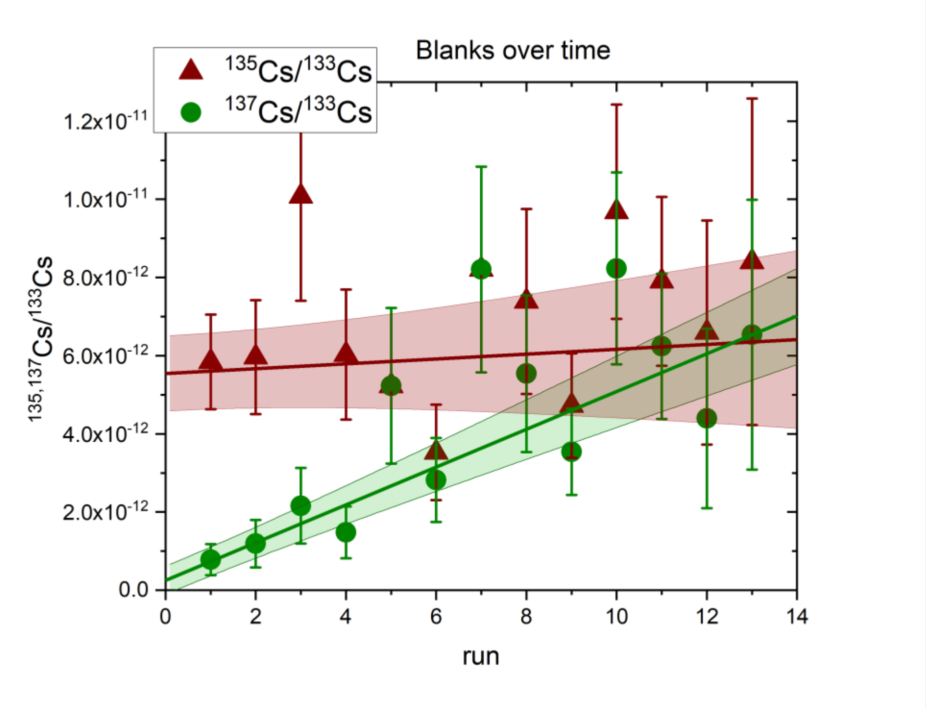
[Data taken from Zok et al., *Environ. Sci. Technol.* 2021]

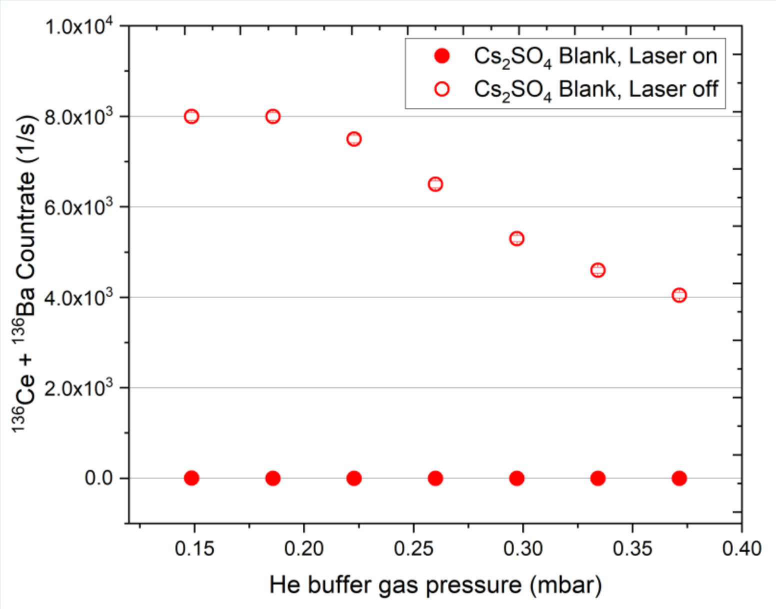
- First real samples measured at VERA show **Chernobyl and Fukushima signature**
- Samples prepared and measured at ICP-QQQ-MS at IRS Hannover

- Abundance sensitivity of ICP MS sufficient for heavily contaminated samples → estimations for **general oceanic samples** in ^{135}Cs/Cs ≈ 10^{-14} range → AMS needed

Next Phase

- Detection limit at VERA $^{135,137}Cs/Cs \approx 10^{-11}$
- Major problem for reaching desired detection limit of 10^{-14} is **cross contamination** in ion source (and not isobaric interferences from barium)





^{135}Cs – State of the Art

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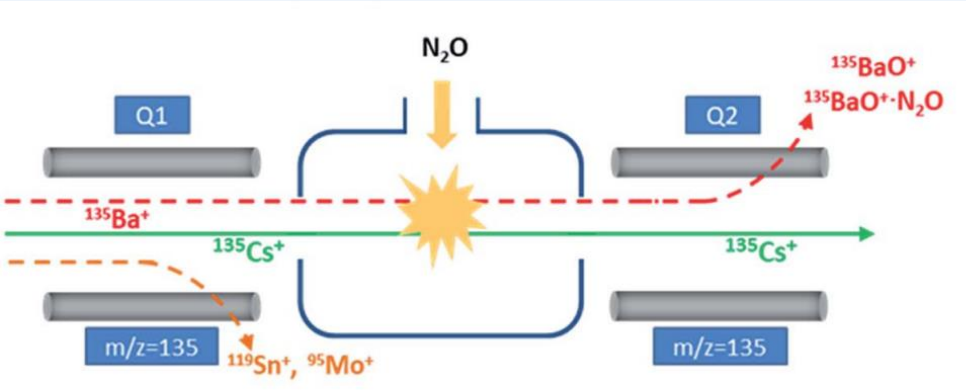
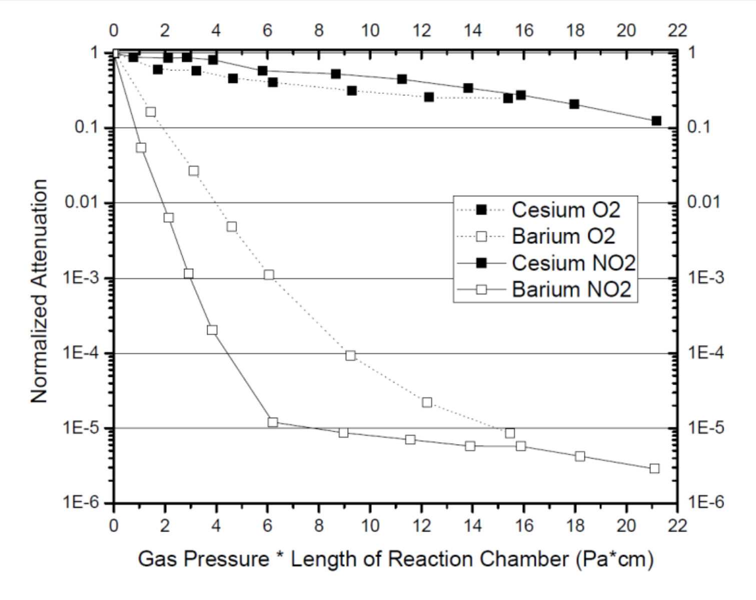
- ICPMS/MS measurements use concept of **reaction cell** with oxygen
- Abundance sensitivity $^{135}Cs/^{133}Cs \approx 10^{-9}$

- AMS measurements at ISO Trace Laboratory (University of Ottawa) → Cs ions captured in **RFQ-reaction cell**
- Abundance sensitivity $^{135}Cs/^{133}Cs \approx 10^{-10}$



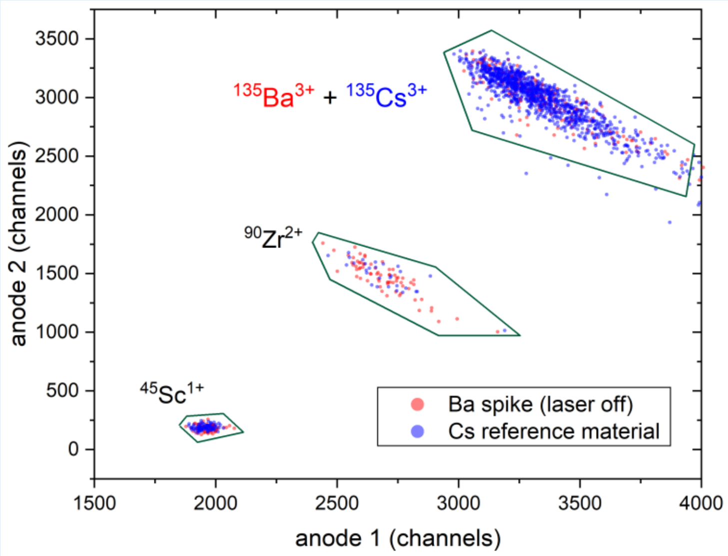
[Bu et al. 2018]



[MacDonald et al. 2014]

Why do we need ILIAMS?

- Barium (even at 8^{+}) **is not separable from Cesium** in the gas-filled ionization chamber
- Near **complete suppression** of barium is necessary
- M/q interferences are well separable



^{135}Cs and ^{137}Cs – Applications

half-life of ^{135}Cs not well known (0.7 – 3.0 Ma) → low level activity measurement and low level MS measurement necessary

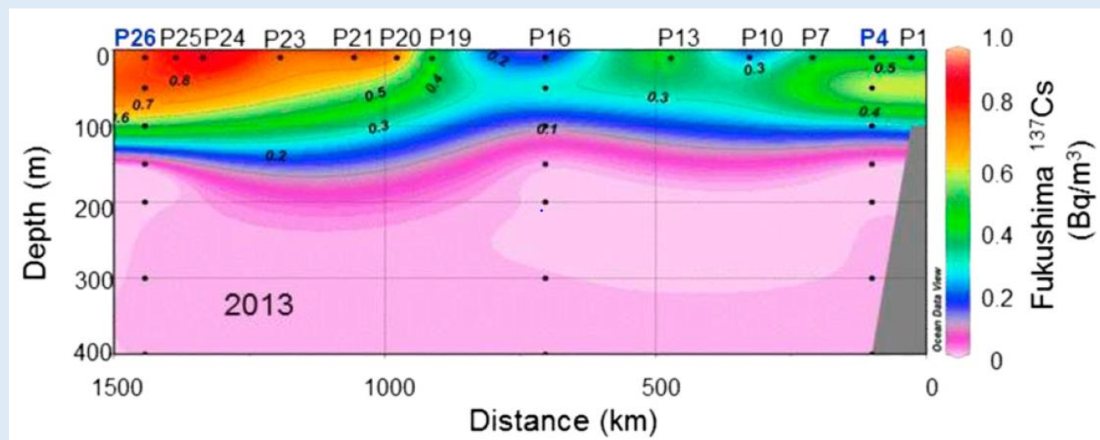
$$T_{1/2}(^{135}Cs) = \ln(2) \cdot (N_{135Cs}/N_{133Cs}) \cdot (N_{133Cs}/A_{135Cs})$$

AMS

Spiked material

Low-level activity measurement

- modeling of anthropogenic radionuclide dispersion
- studying erosion
- dating sediments



[Smith et al. 2015]