DEVELOPMENT OF ^{129}I AMS AT THE NUCLEAR SCIENCE LABORATORY FOR MEASUREMENTS OF THE GREAT LAKES REGION

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FNAL

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OUTLINE

1. ^{129}I and ^{129}I Content in the Environment
2. Accelerator Mass Spectrometry
3. Accelerator Mass Spectrometry for ^{129}I Measurement
4. Sample Collection and Chemistry
5. ^{129}I Measurement and Results
6. Future Work / Conclusions

^{129}I AND ^{129}I CONTENT IN THE ENVIRONMENT

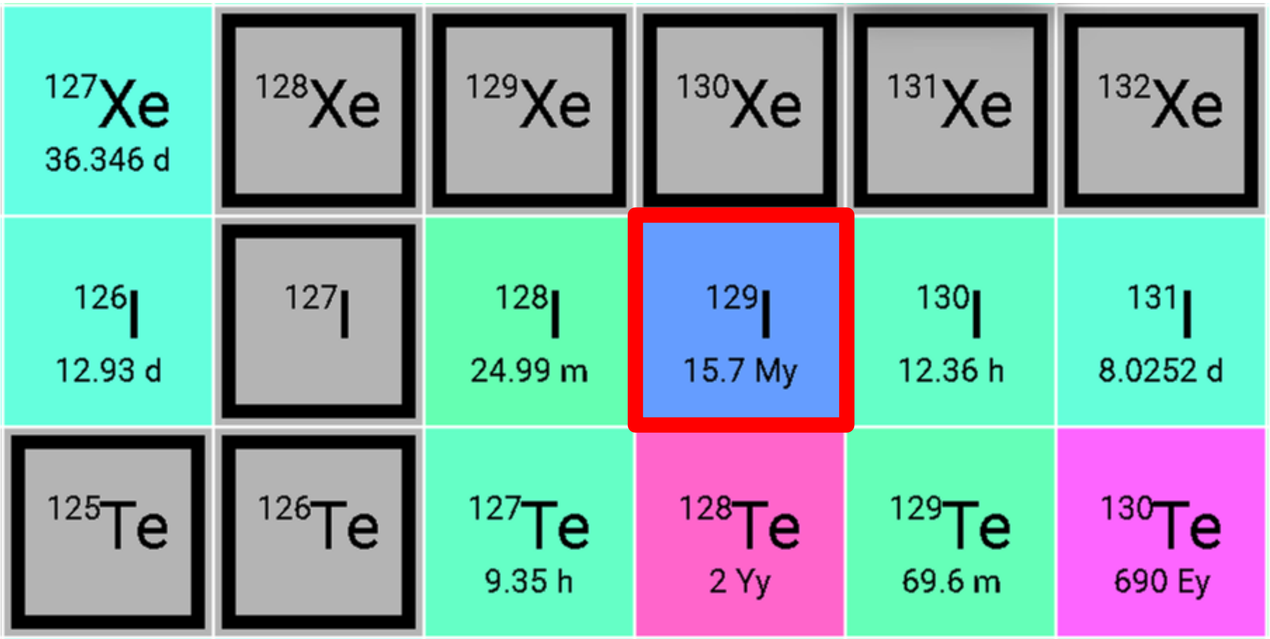
ENVIRONMENTAL RADIONUCLIDES

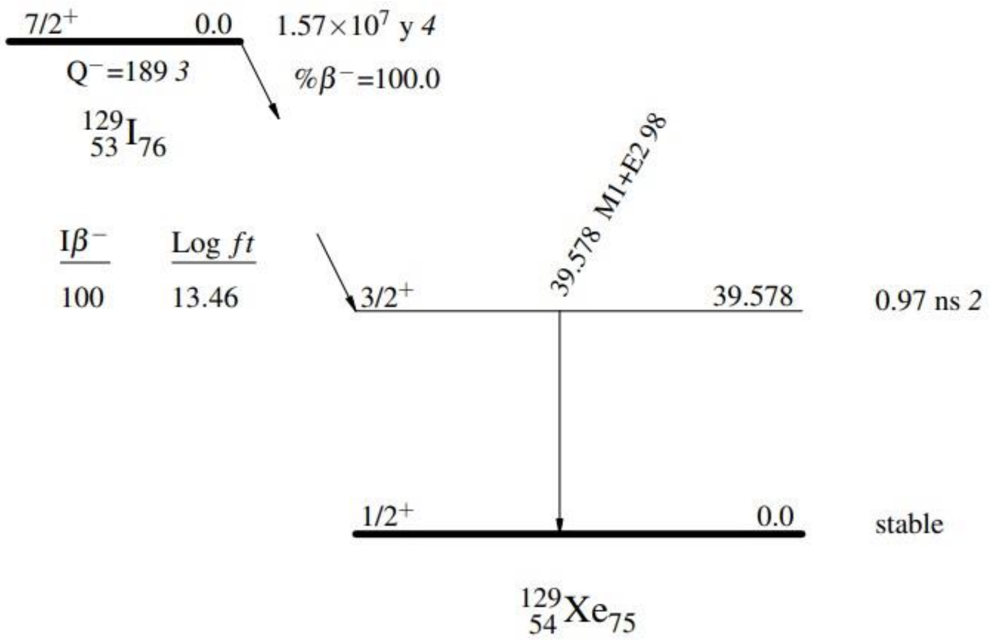
Radionuclide	$t_{1/2}$ (Myr)
^{236}U	23.4
^{129}I	15.7
^{10}Be	1.5
^{26}Al	0.7
^{36}Cl	0.3
^{41}Ca	0.1

- There are many radionuclides in the environment that have half-lives ≥ 0.1 Myr, both naturally-produced and anthropogenic (human-produced)
- Special attention to ^{129}I for environmental tracing
  - Volatile
  - Soluble
  - Biophilic (easily incorporated into organisms)

129I

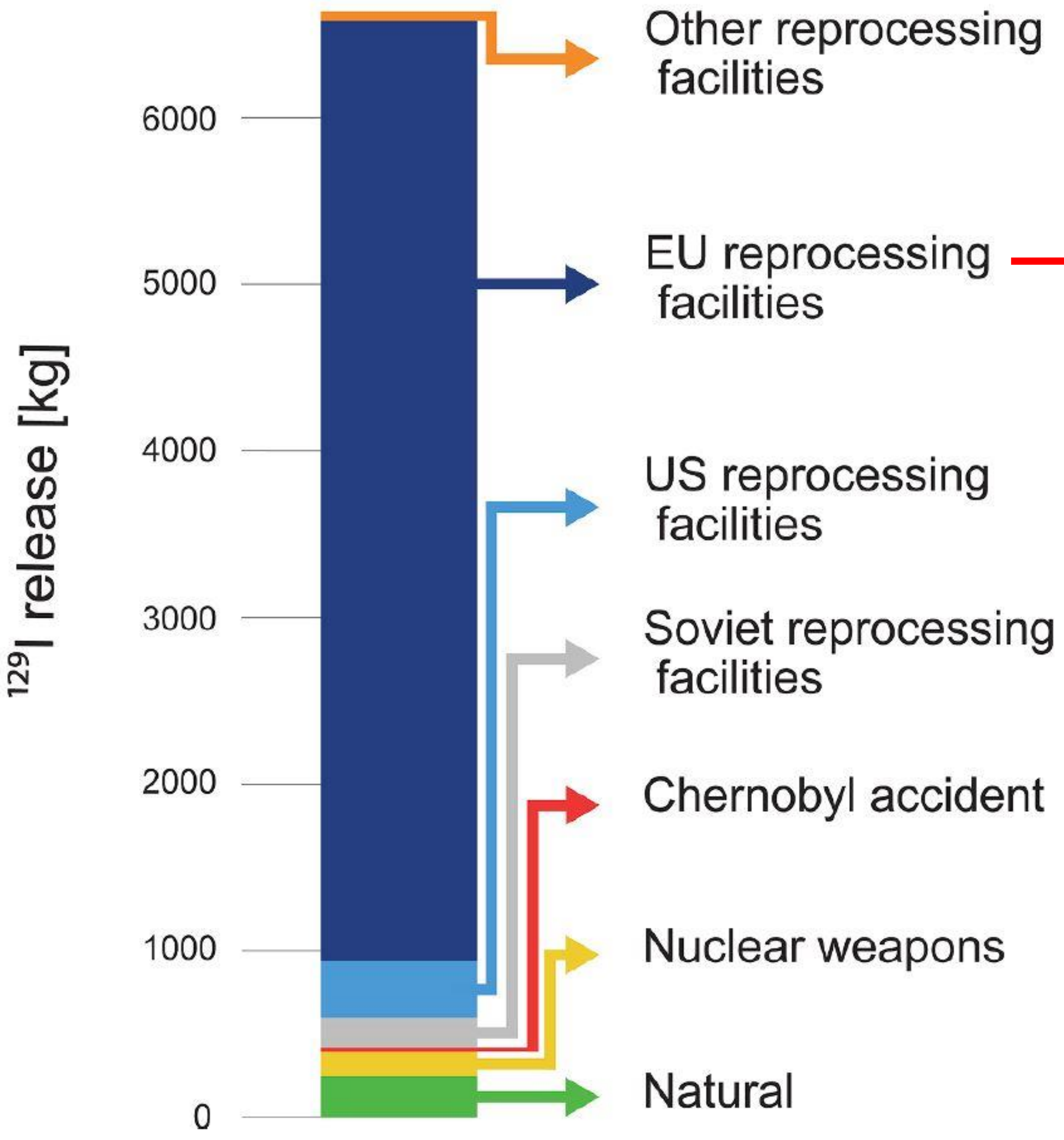
- $t_{1/2}$ = 15.7 Myr
  - Longest-lived radioisotope of iodine
  - Decays to stable ^{129}Xe
- Production
  - Cosmic-ray-induced reactions on Xe
  - Spontaneous fission of U
  - Thermal-neutron-induced fission
    - ^{235}U – 0.7%
    - ^{239}Pu – 1.4%





CONTRIBUTIONS TO THE GLOBAL ^{129}I CONTENT

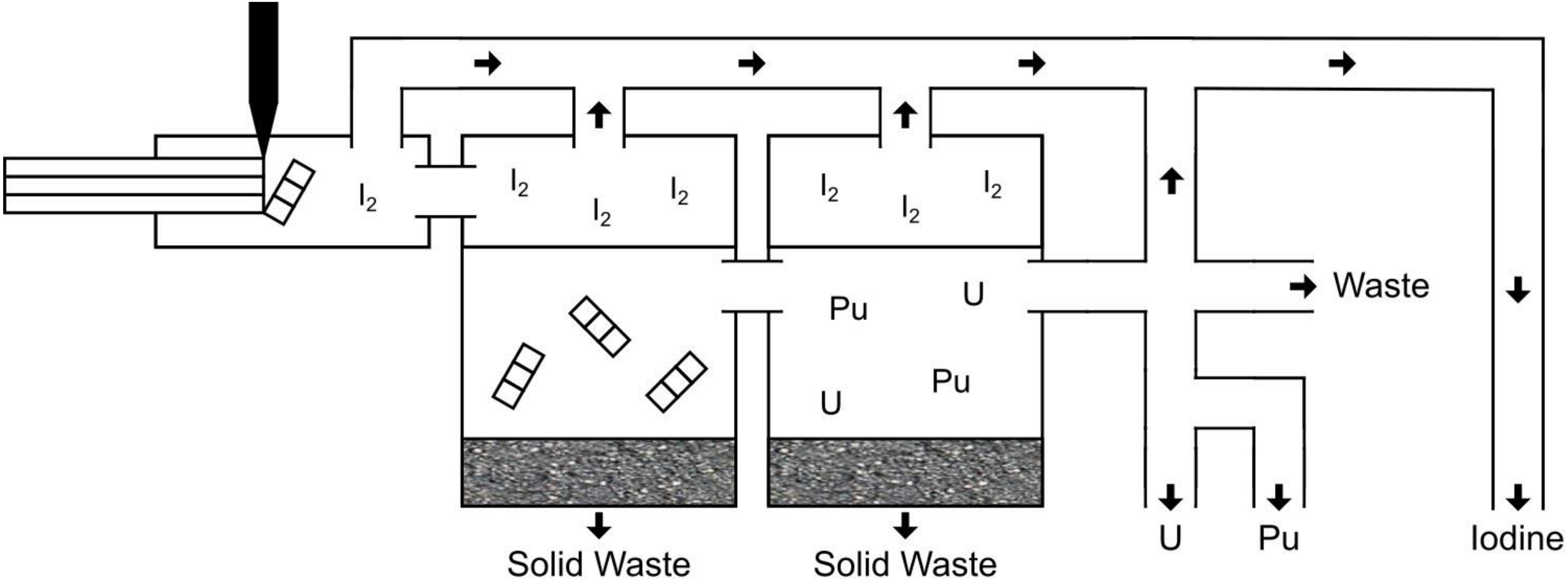
- Natural amount
  - ~100 kg
- Nuclear weapons tests
  - ~50 kg
- Chernobyl/Fukushima
  - ~a few kilograms
- Spent fuel reprocessing
  - ~5300 kg



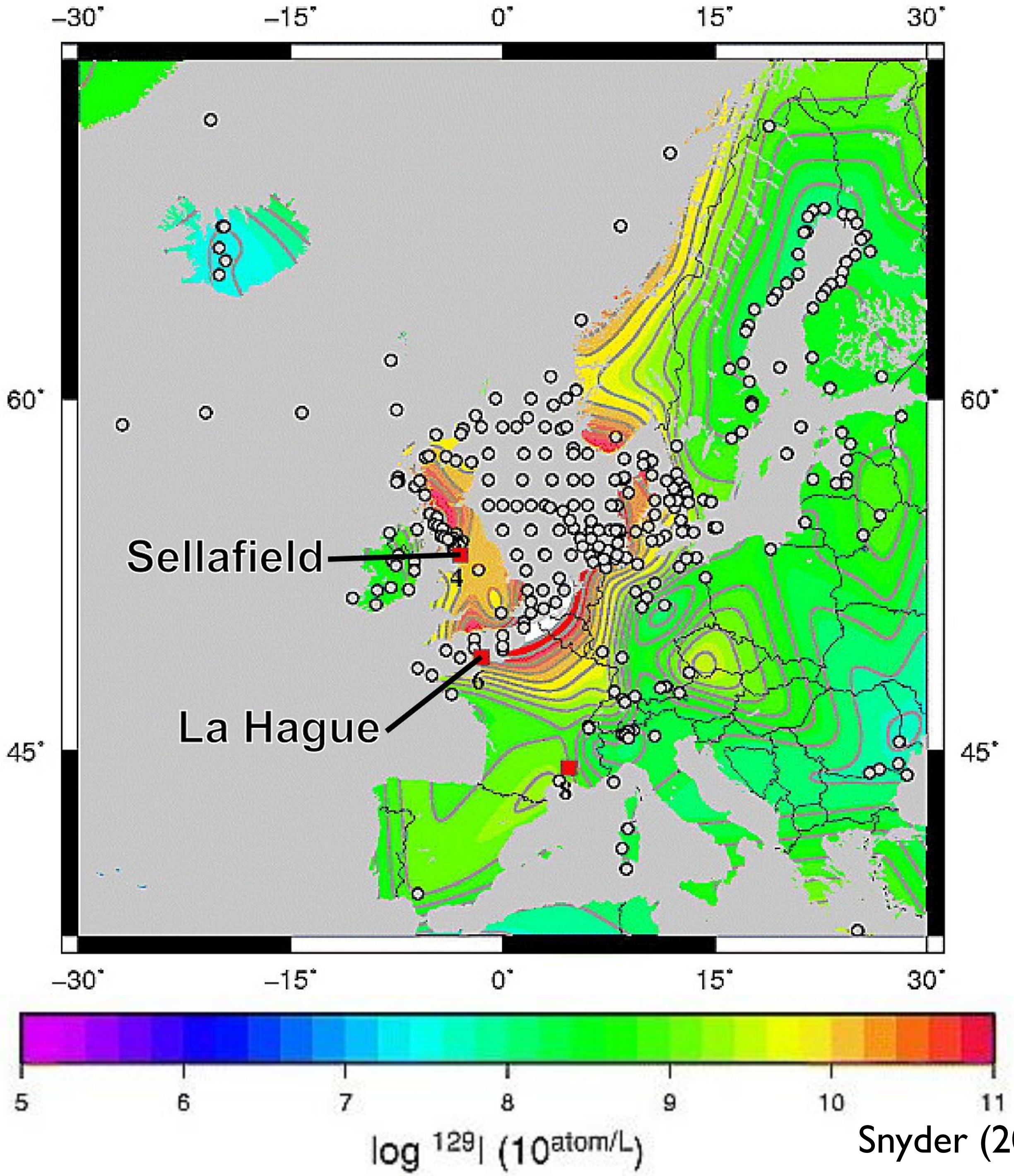
Effectively "point-like" sources

Kamleitner (2019)

PUREX PROCESS



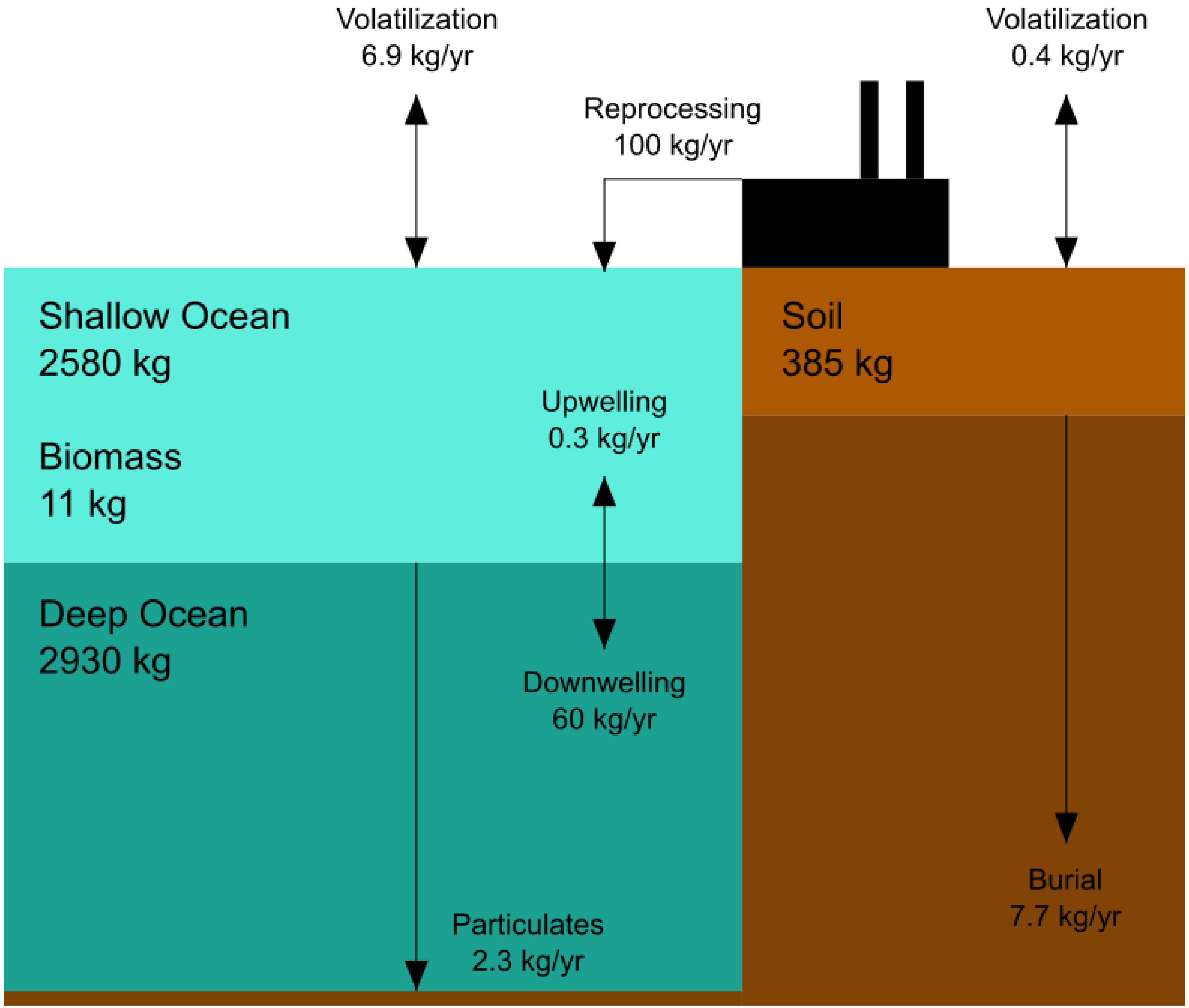
EFFECT OF ^{129}I RELEASES NEAR REPROCESSING CENTERS



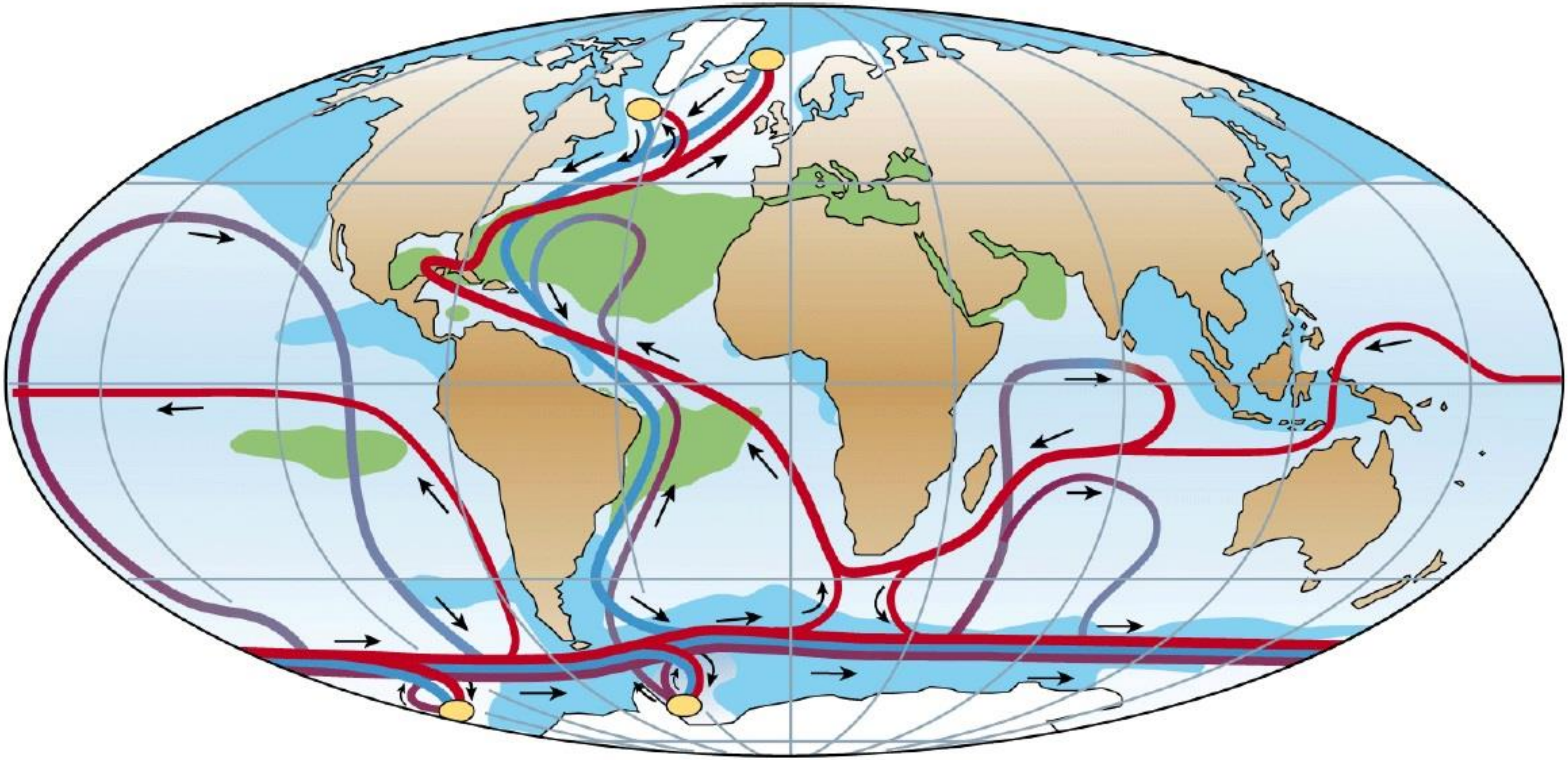
Snyder (2010)

SINKS OF ^{129}I

- Calculating the ^{129}I content of the shallow oceans (up to 100 m depth) from global ^{129}I content distribution accounts for only half of ^{129}I content
- Significant sinks
  - Deep ocean
  - Burial in soil

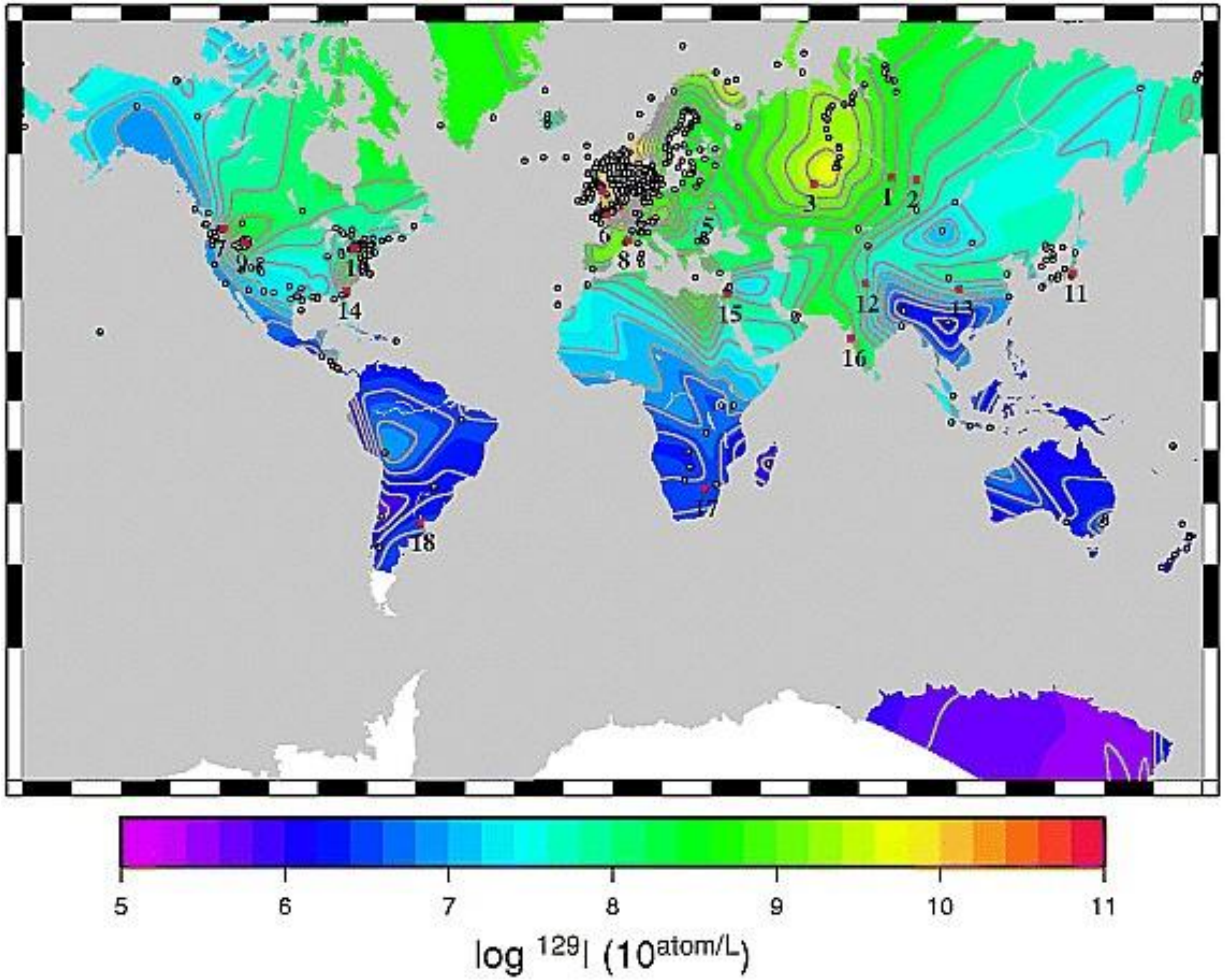


Reproduced from Snyder (2010)



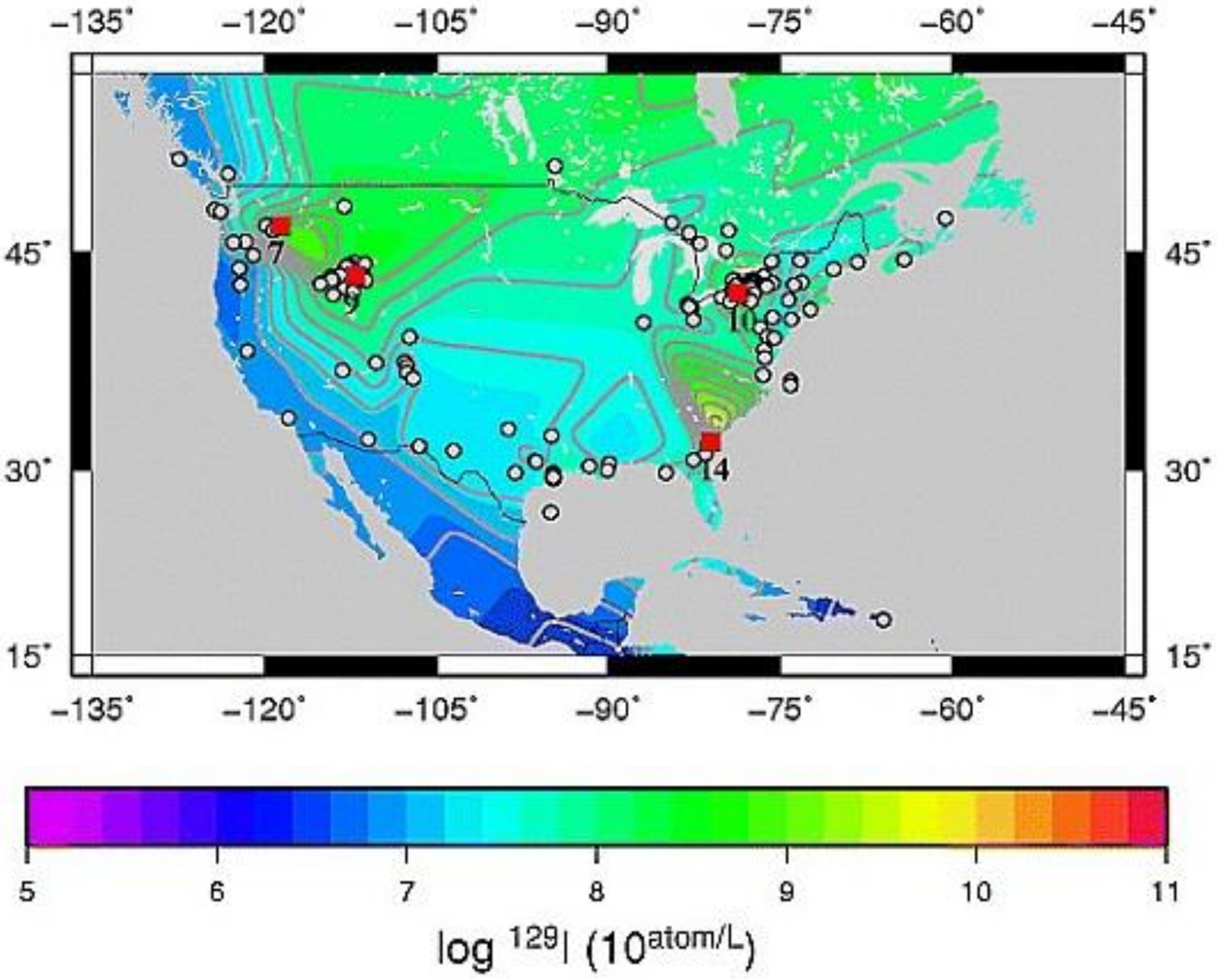
Rahmstorf (2002)

EFFECT ON GLOBAL ^{129}I WATER CONTENT



Snyder (2010)

NORTH AMERICAN ^{129}I WATER CONTENT



Snyder (2010)

NEED FOR ^{129}I MEASUREMENTS

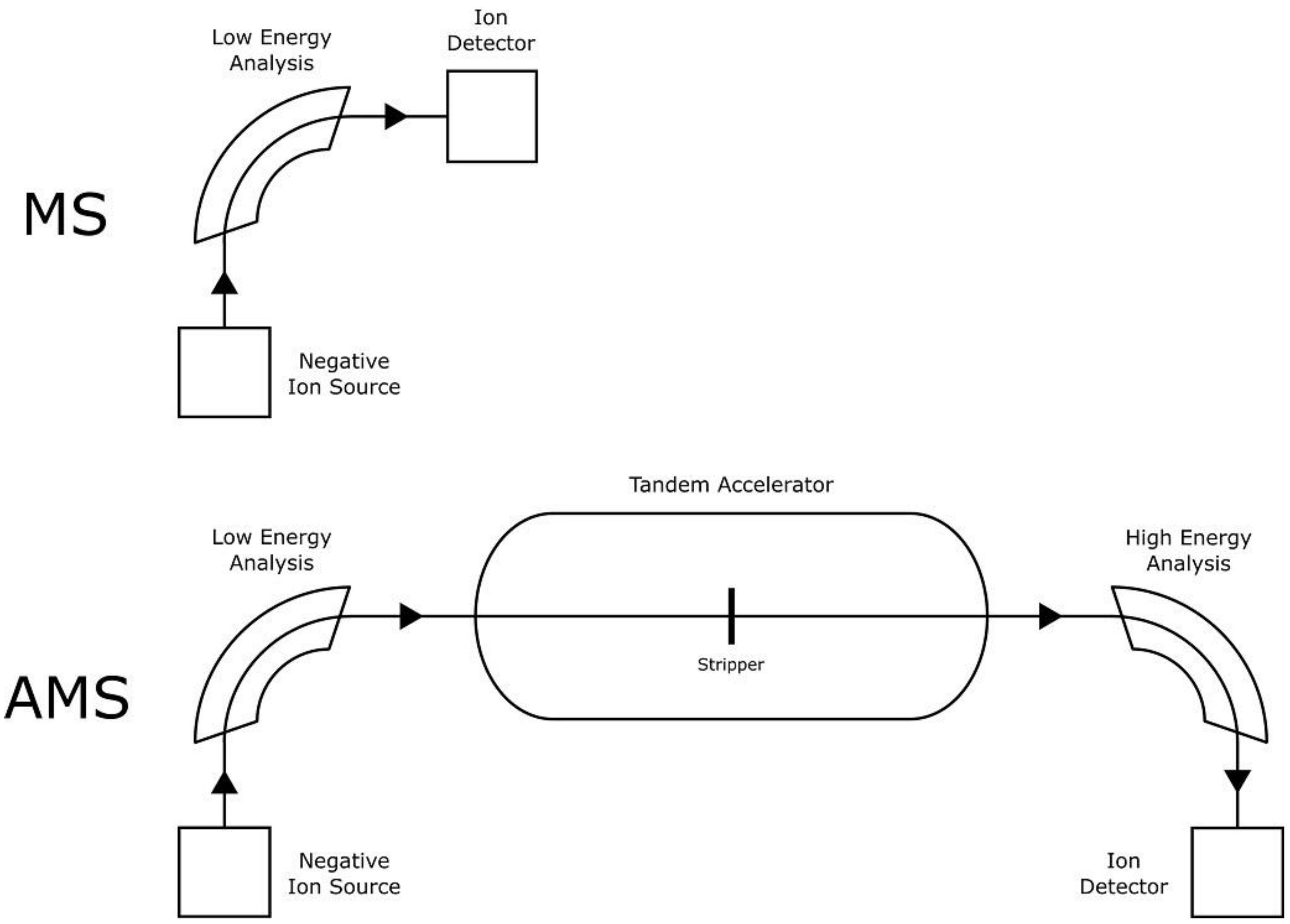
Specifically, widespread, real-time measurements are needed

- Background for identifying unknown contributions
- Identification of transport pathways
  - Also useful for prediction of ^{131}I transport
- Accumulation of ^{129}I

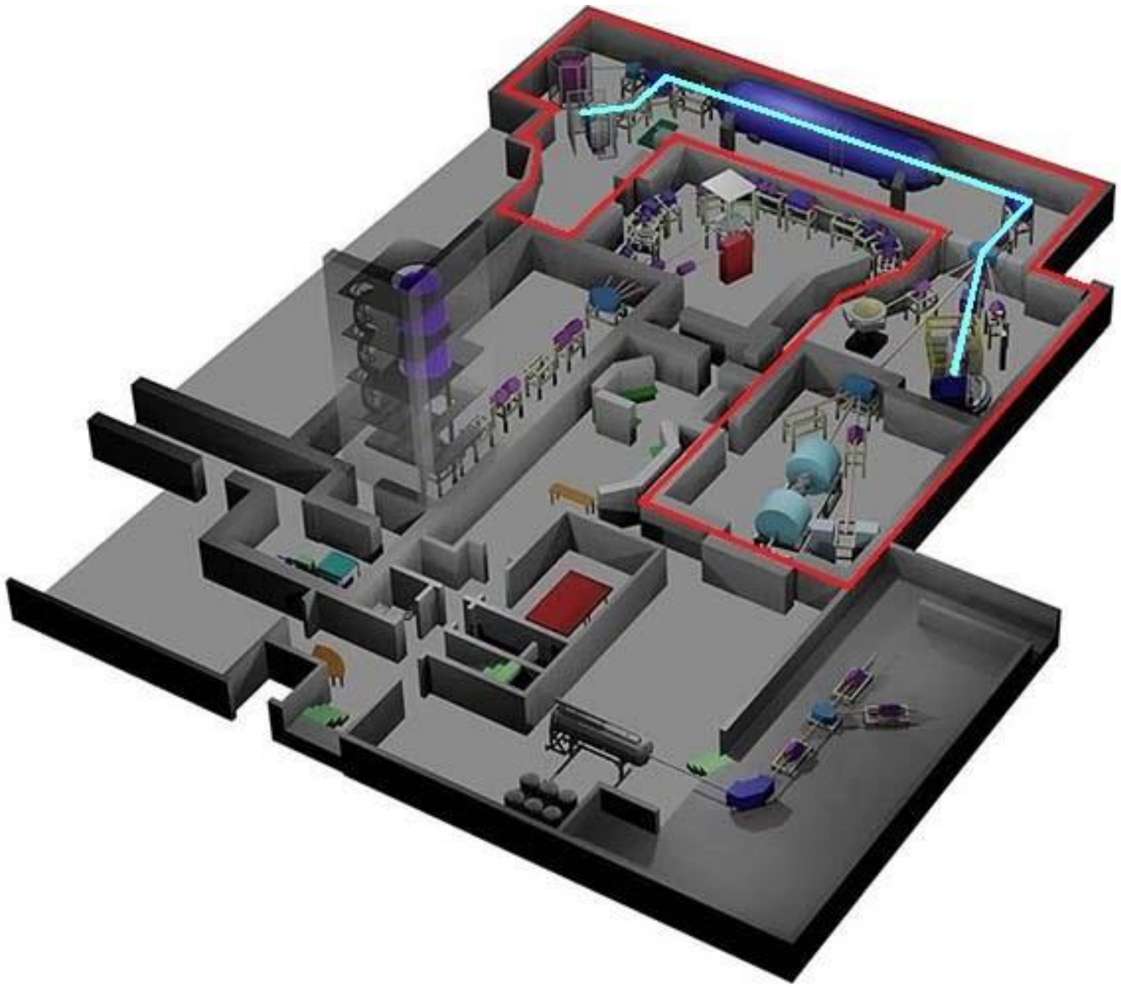
ACCELERATOR MASS SPECTROMETRY

MS VS AMS

- Both utilize:
  - Ion production
  - Low energy analysis
  - Ion detection
- MS has <u>no:</u>
  - Isobaric separation
  - Molecular discrimination

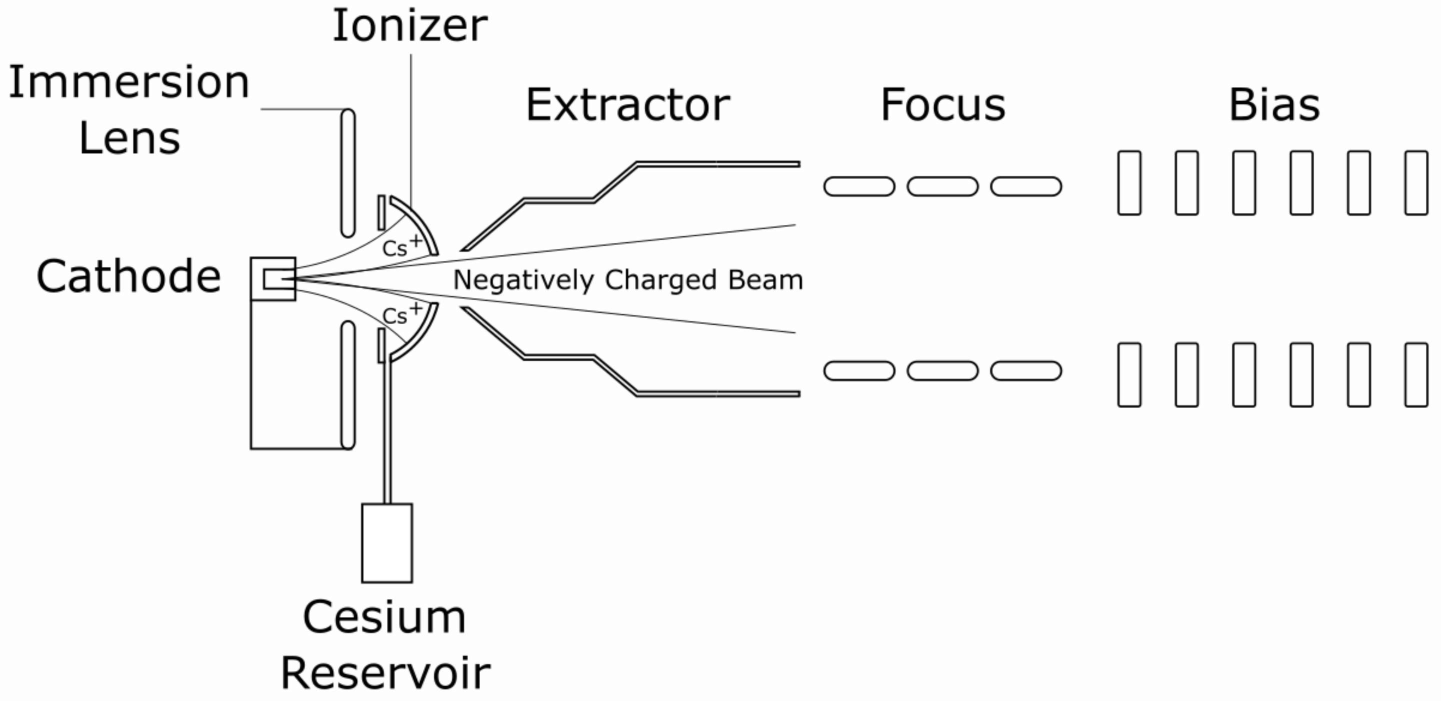


THE AMS BEAMLINE AT THE NSL



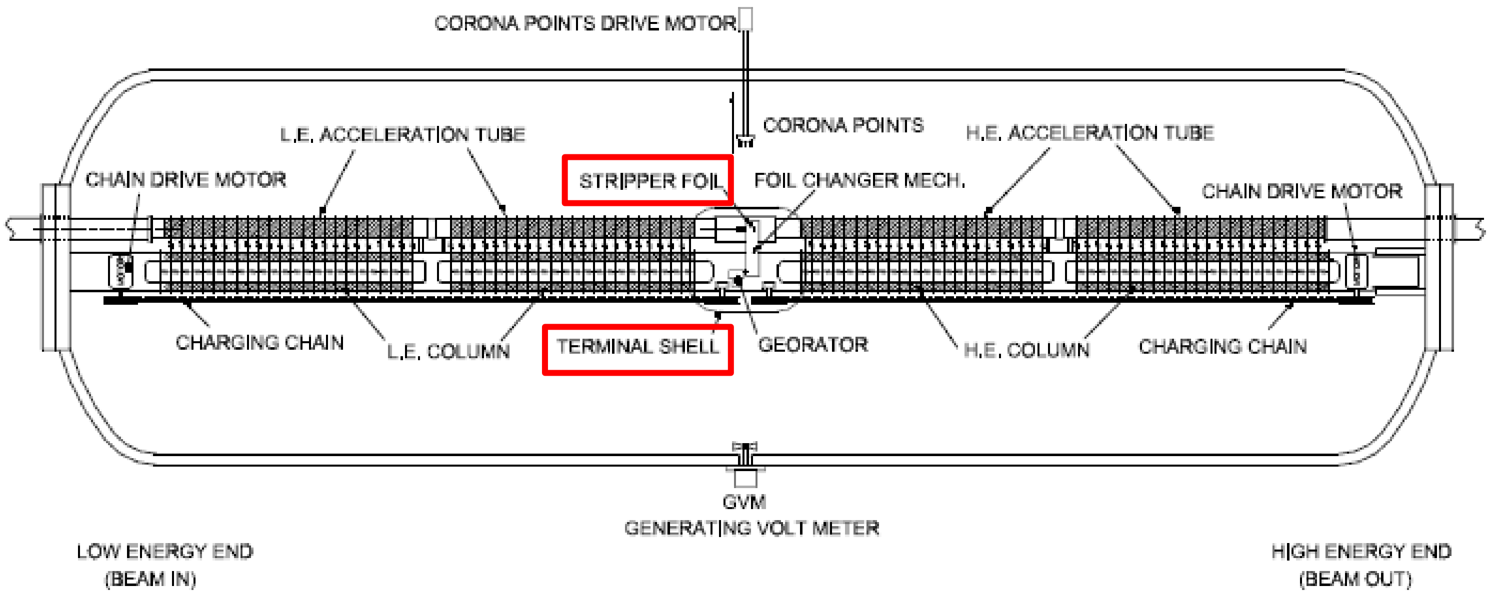
ION PRODUCTION

- Multi-Cathode Source of Negative Ions via Cesium Sputtering (MC-SNICS)
- Produces elemental and molecular ion beams of varying intensities (1 nA to > 10 μA)
- Can hold 20 or 40 samples depending on sample size



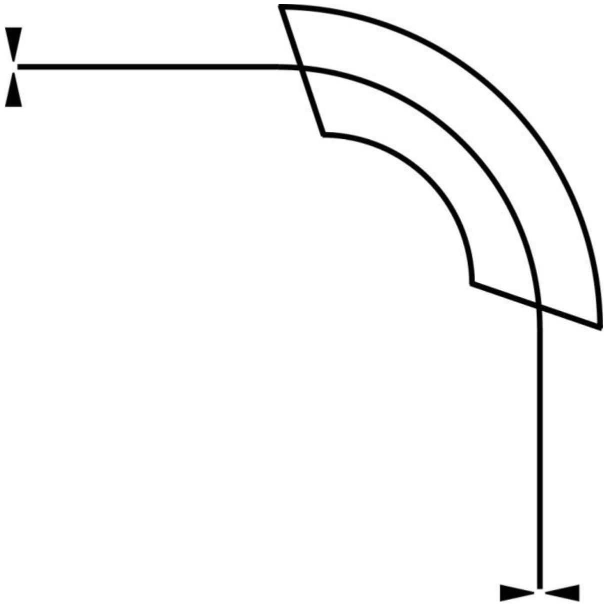
ACCELERATION

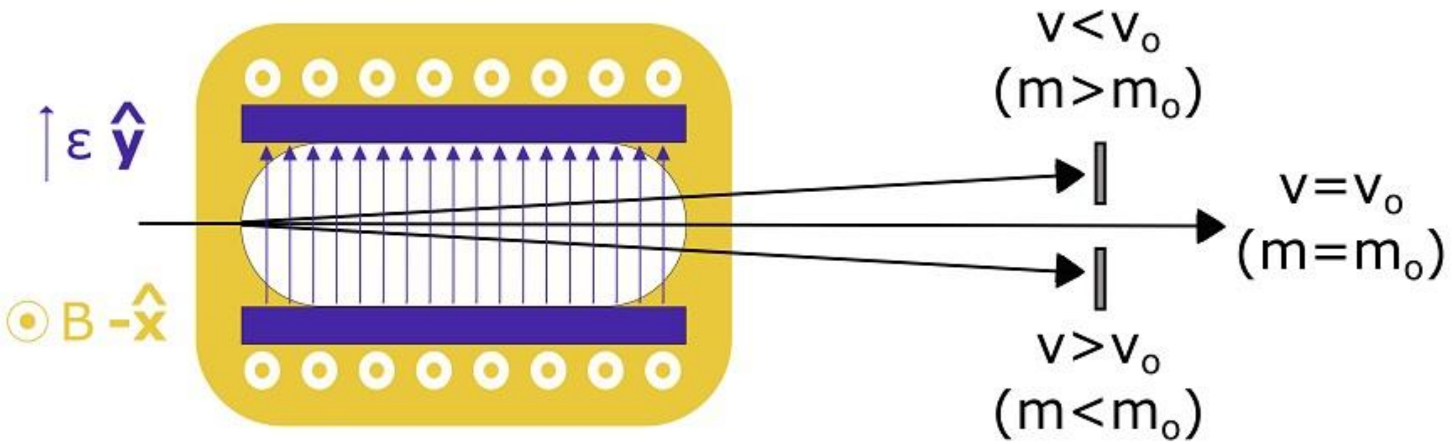
- Negative beam accelerated to terminal shell
- Stripper inside terminal strips electrons (charge state distribution), breaks up molecules
- Positive beam accelerated again



HIGH ENERGY ANALYSIS

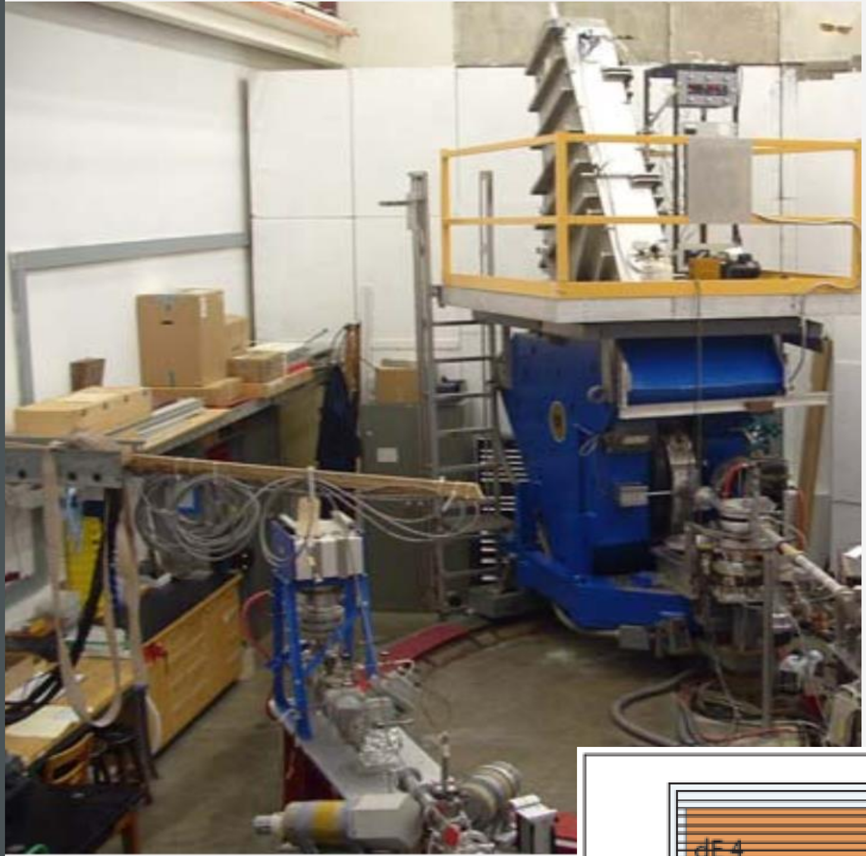
- Select energy, mass, and charge state (magnetic rigidity)
- Secondary filter (velocity) for beams of same/similar magnetic rigidity

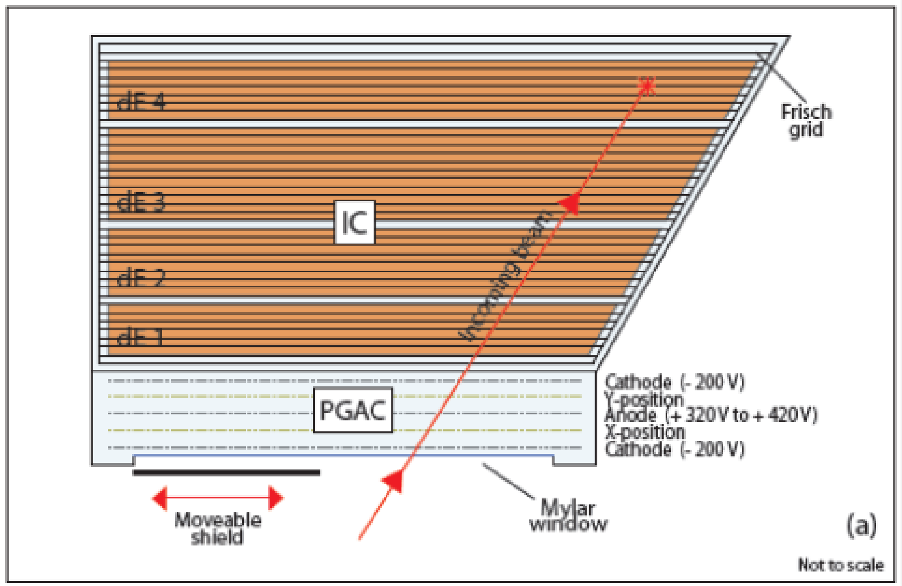


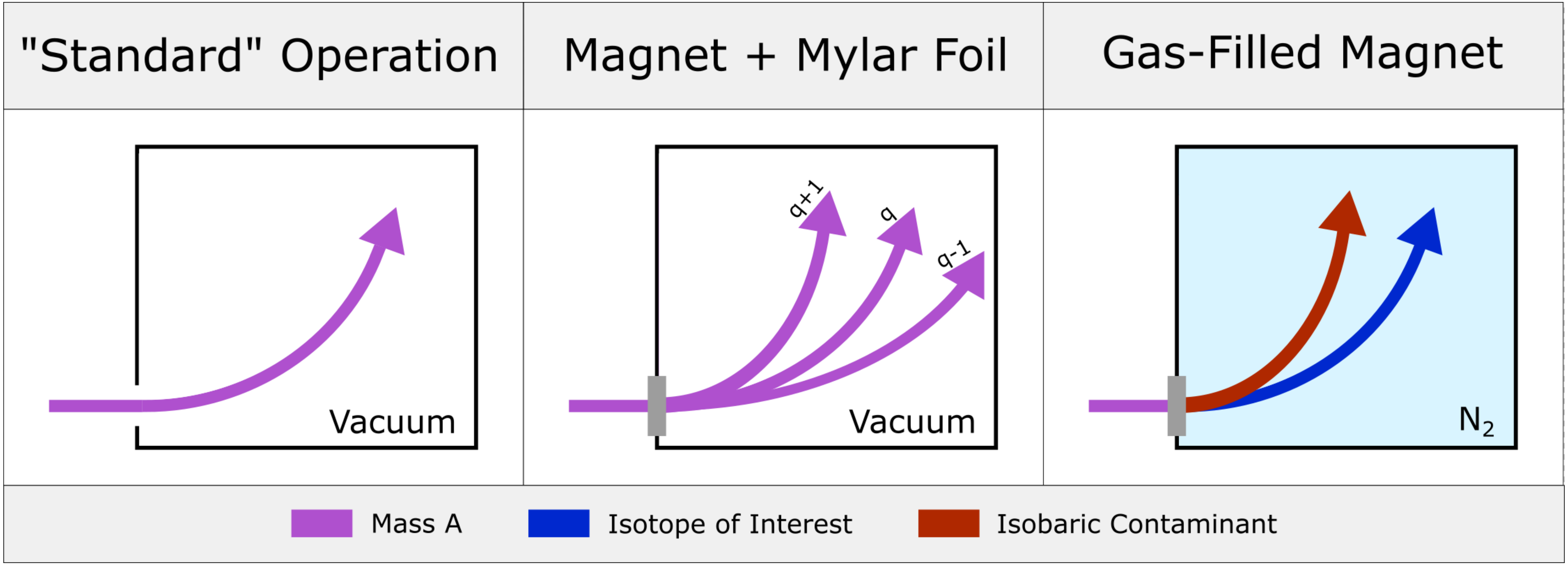


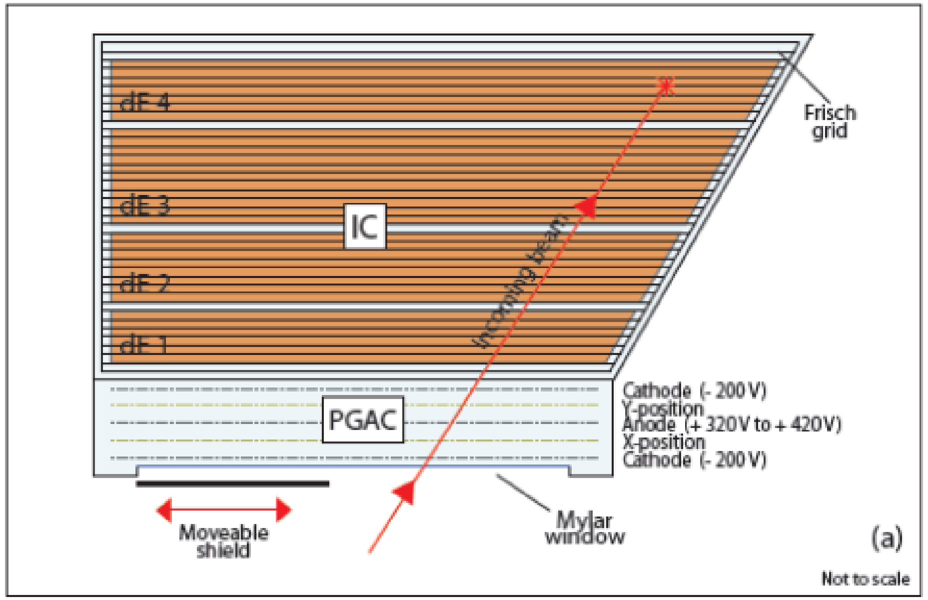
ION DETECTION

- Ions are detected by their properties (energy, energy loss)
- NSL traditionally used the Browne-Buechner spectrograph operated using the Gas-Filled Magnet technique

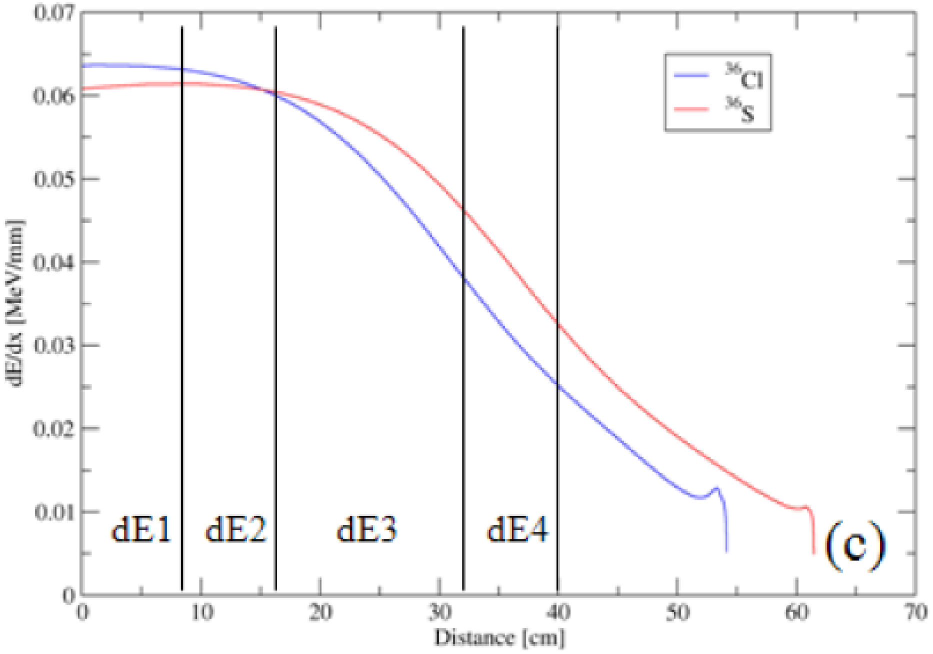




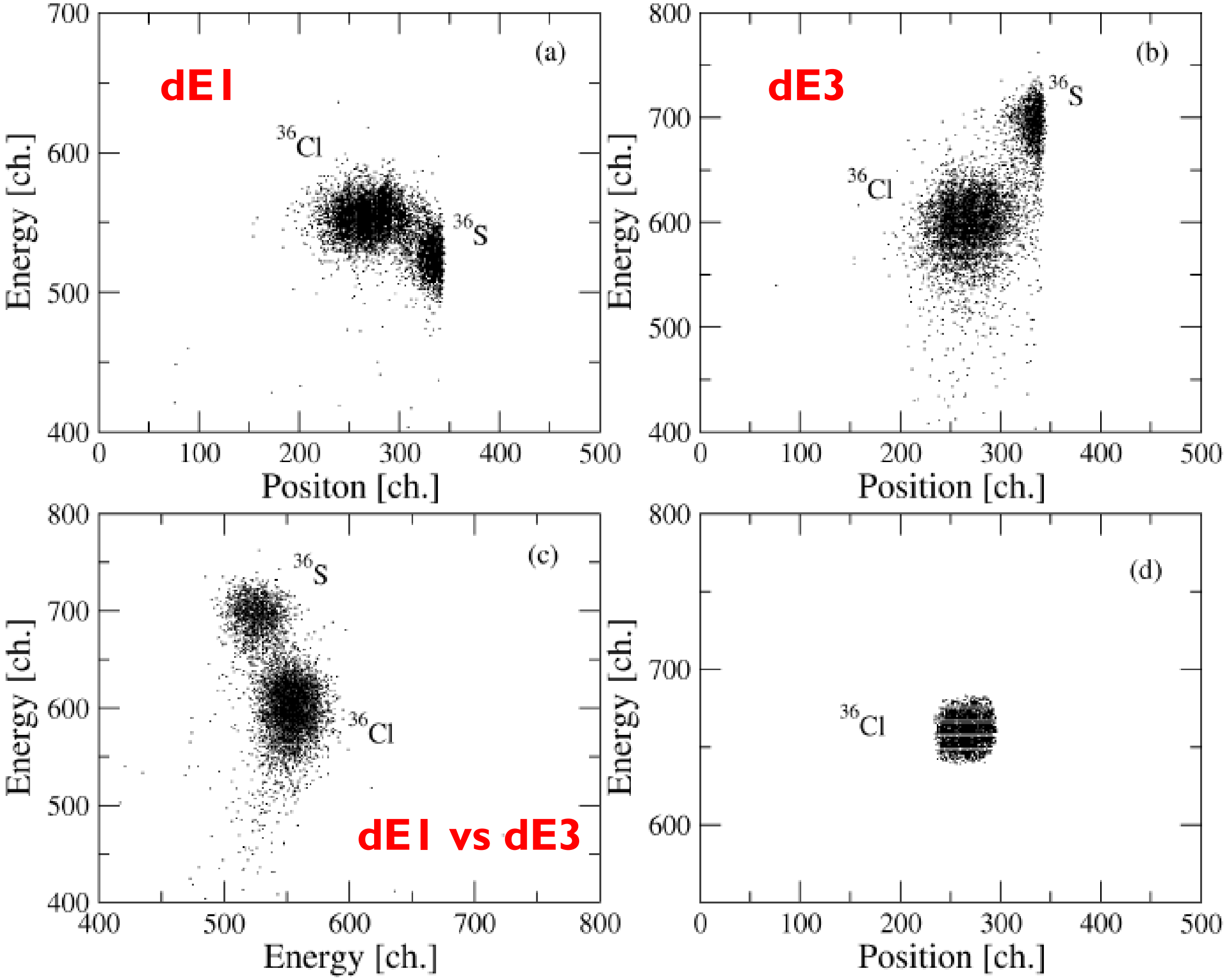

"Standard" Operation
Magnet + Mylar Foil
Gas-Filled Magnet
Vacuum
q+1
q
q-1
Vacuum
N2
Mass A
Isotope of Interest
Isobaric Contaminant



dE 4
dE 3
IC
dE 2
dE 1
Incoming beam
Frisch grid
PGAC
Cathode (- 200 V)
Y-position
Anode (+ 320 V to + 420 V)
X-position
Cathode (- 200 V)
Moveable shield
Mylar window
(a)
Not to scale



dE/dx [MeV/mm]
Distance [cm]
36Cl
36S
dE1
dE2
dE3
dE4
(c)



(a)
dE1
36Cl
36S
Energy [ch.]
Positon [ch.]
(b)
dE3
36S
36Cl
Energy [ch.]
Position [ch.]
(c)
36S
36Cl
dE1 vs dE3
Energy [ch.]
Energy [ch.]
(d)
36Cl
Energy [ch.]
Position [ch.]

AMS RADIONUCLIDES

Measured

Si Det

- ^{14}C
  - Undergraduate project for biology, art

GFM

- ^{36}Cl
  - $^{33}S(\alpha,p)^{36}Cl$, $^{34}S(^{3}He,p)^{36}Cl$ for ESS production
- ^{44}Ti
  - $^{40}Ca(\alpha,\gamma)^{44}Ti$
- ^{60}Fe
  - Verification of a longer half-life

In development

GFM

- ^{41}Ca
  - ESS production
- ^{53}Mn
  - Geophysics, terrestrial production
- ^{93}Zr
  - Nuclear waste, nuclear astrophysics

TOF

- Actinides
  - Neutron transmutation rates/reactor design

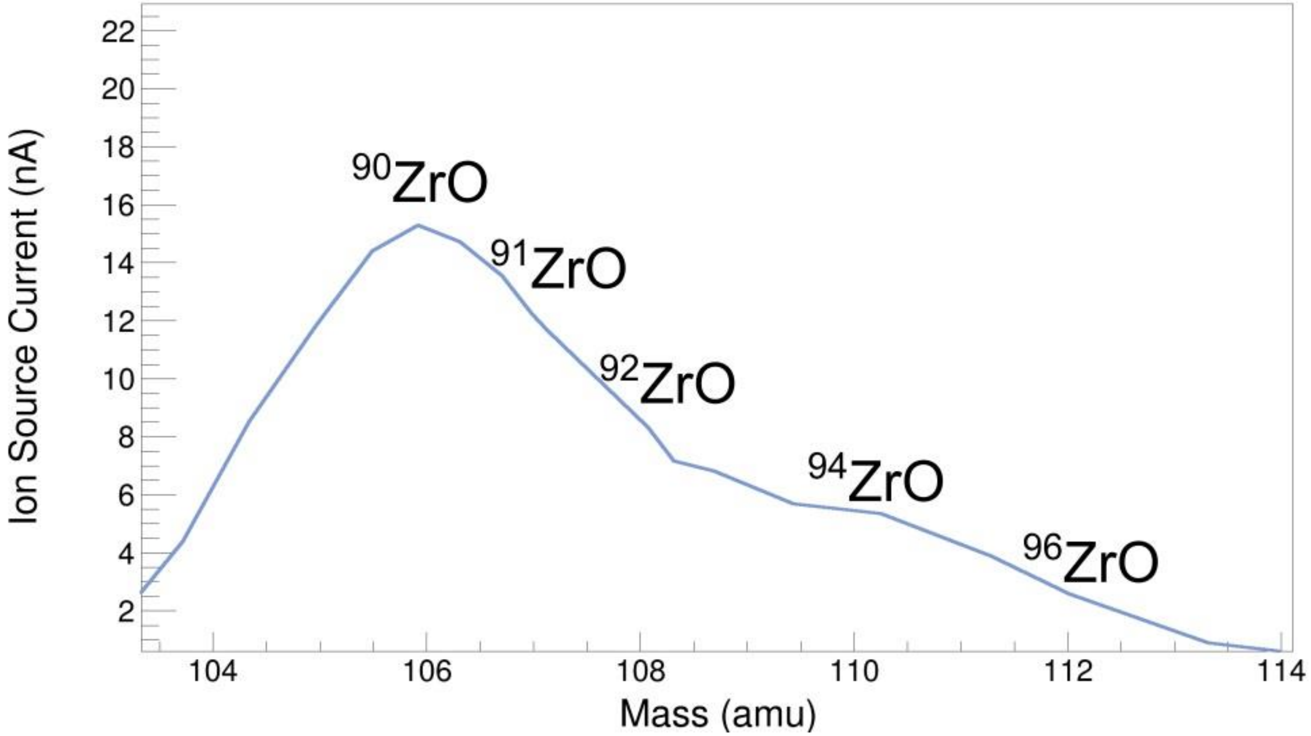
ACCELERATOR MASS SPECTROMETRY FOR ^{129}I MEASUREMENT

^{129}I ION BEAM PRODUCTION

- Isobaric contaminant: ^{129}Xe
  - Xenon cannot produce negative ions
- Stable isotopes: ^{127}I
- Sample powder: AgI + Nb (most commonly used mixture)
  - 1:5 ratio by mass
  - Copper sample holders

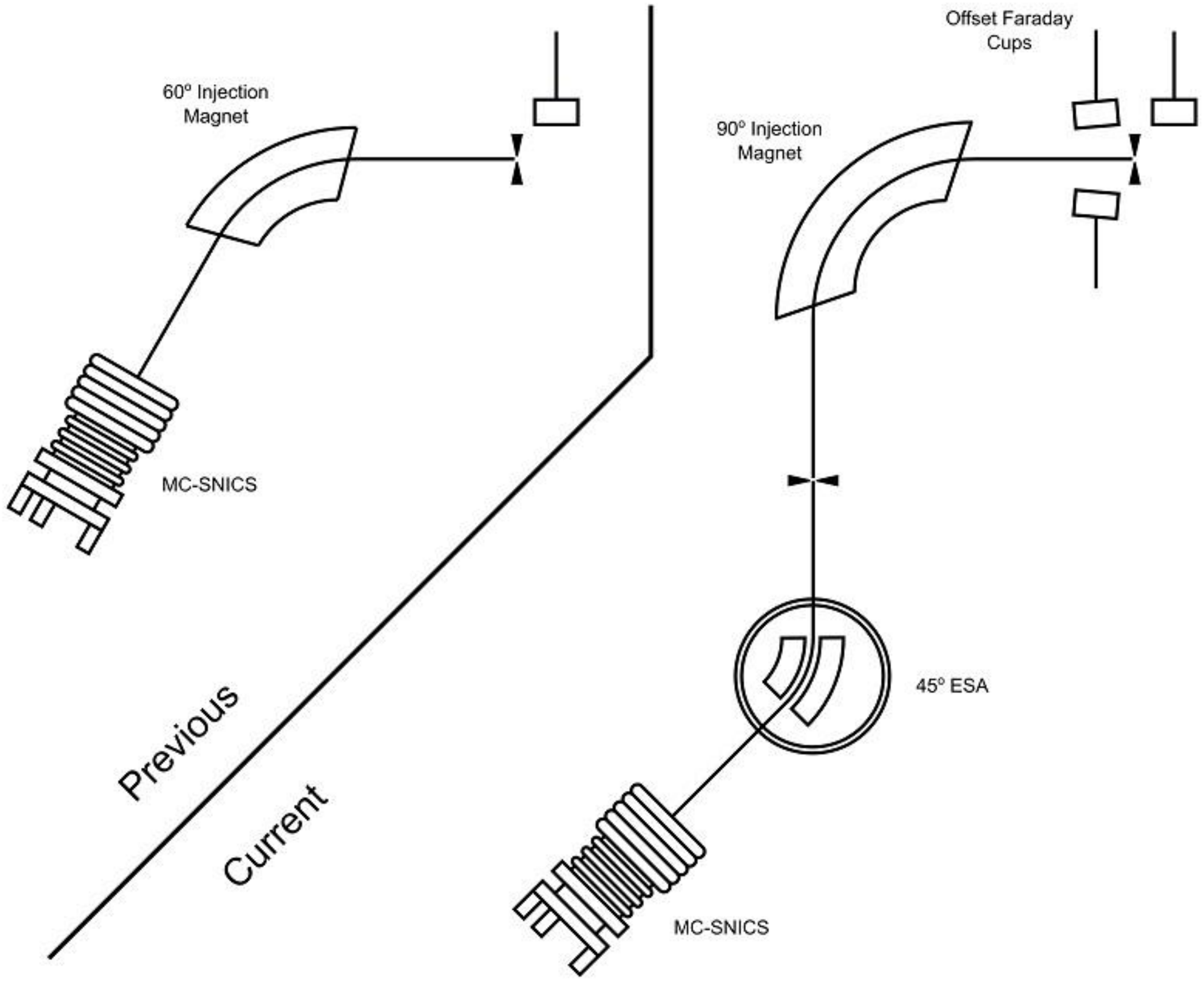
PREVIOUS LOW ENERGY INJECTION

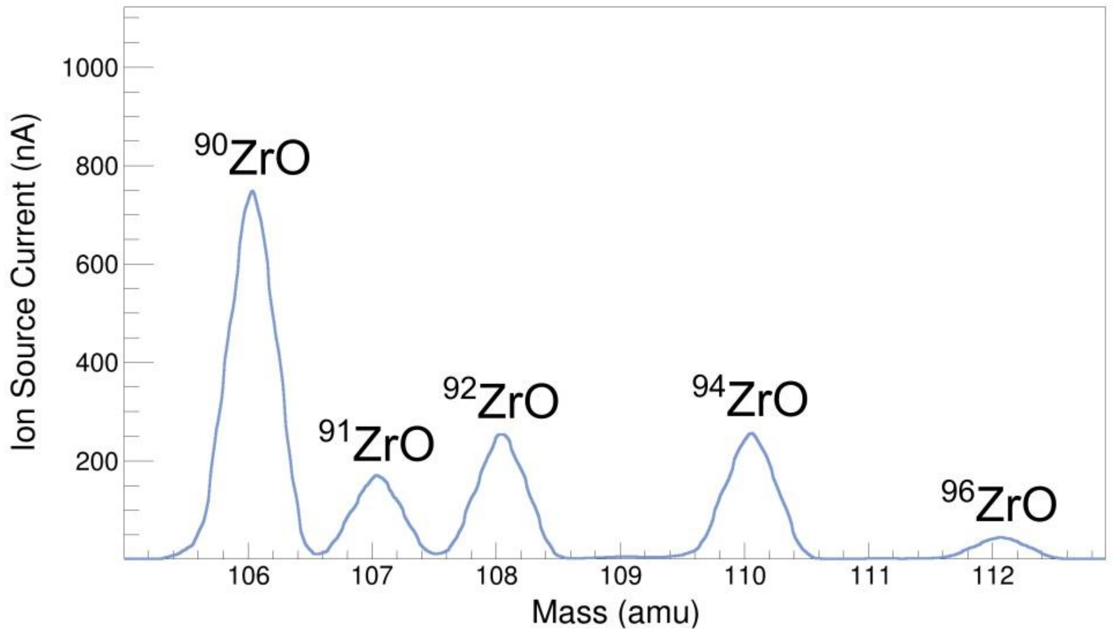
- Isotopic selectivity of the MC-SNICS proved insufficient during development of ^{93}Zr AMS
- Neighboring isotopes ^{92}Zr, ^{94}Zr not only injected past injection magnet, but all the way to the AMS beamline
- This poses a problem to the separation of ^{129}I from ^{127}I



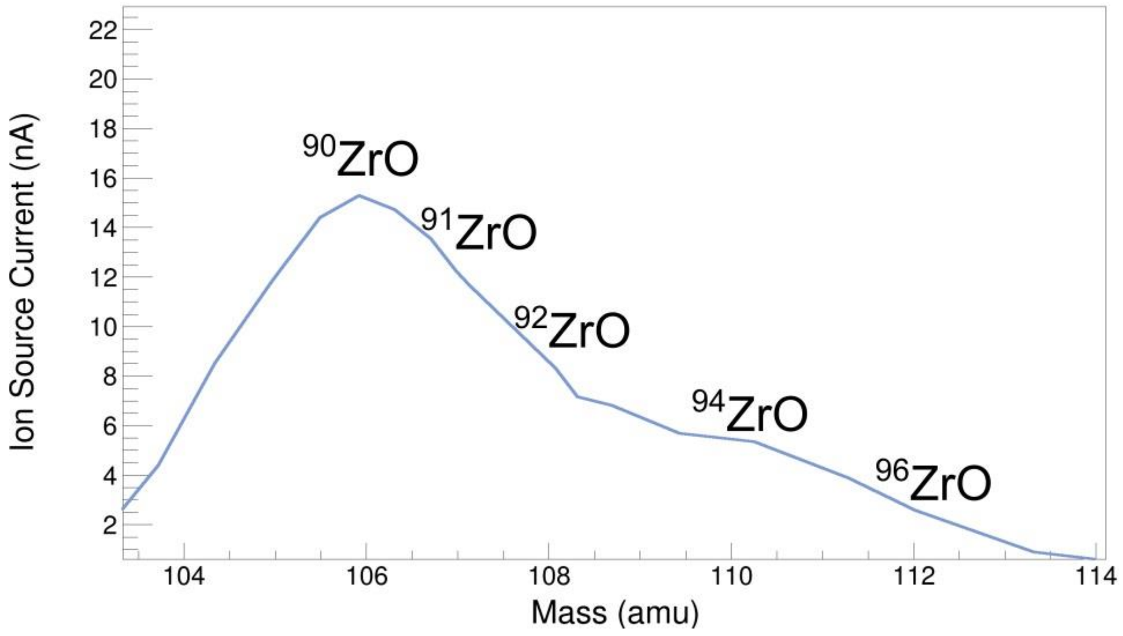
UPGRADE: NSF MAJOR RESEARCH INSTRUMENTATION GRANT

- In 2013, an MRI application for injection upgrade was approved, installed in 2016
- Added a 45° electrostatic analyzer for energy selection
- A 90° injection magnet adds improved separation and double focusing
- Improved optics included in installation plans





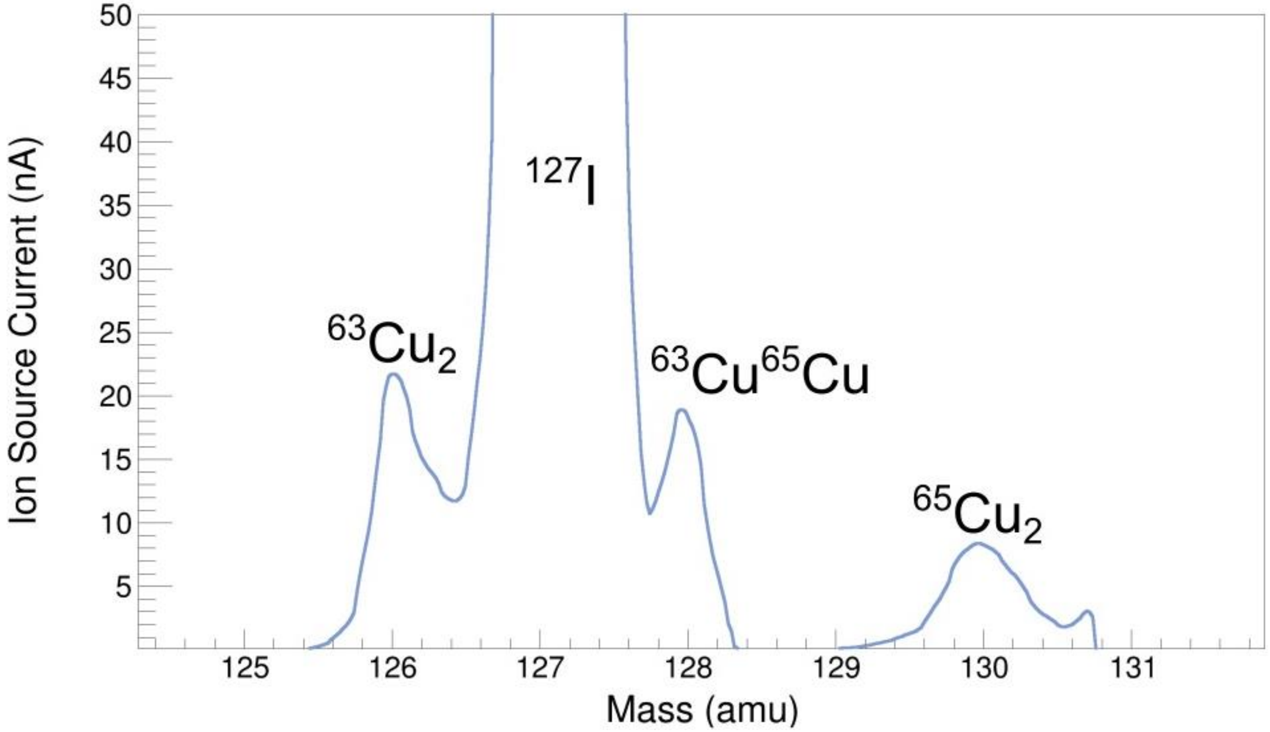
^{90}ZrO
^{91}ZrO
^{92}ZrO
^{94}ZrO
^{96}ZrO
Ion Source Current (nA)
Mass (amu)



^{90}ZrO
^{91}ZrO
^{92}ZrO
^{94}ZrO
^{96}ZrO
Ion Source Current (nA)
Mass (amu)

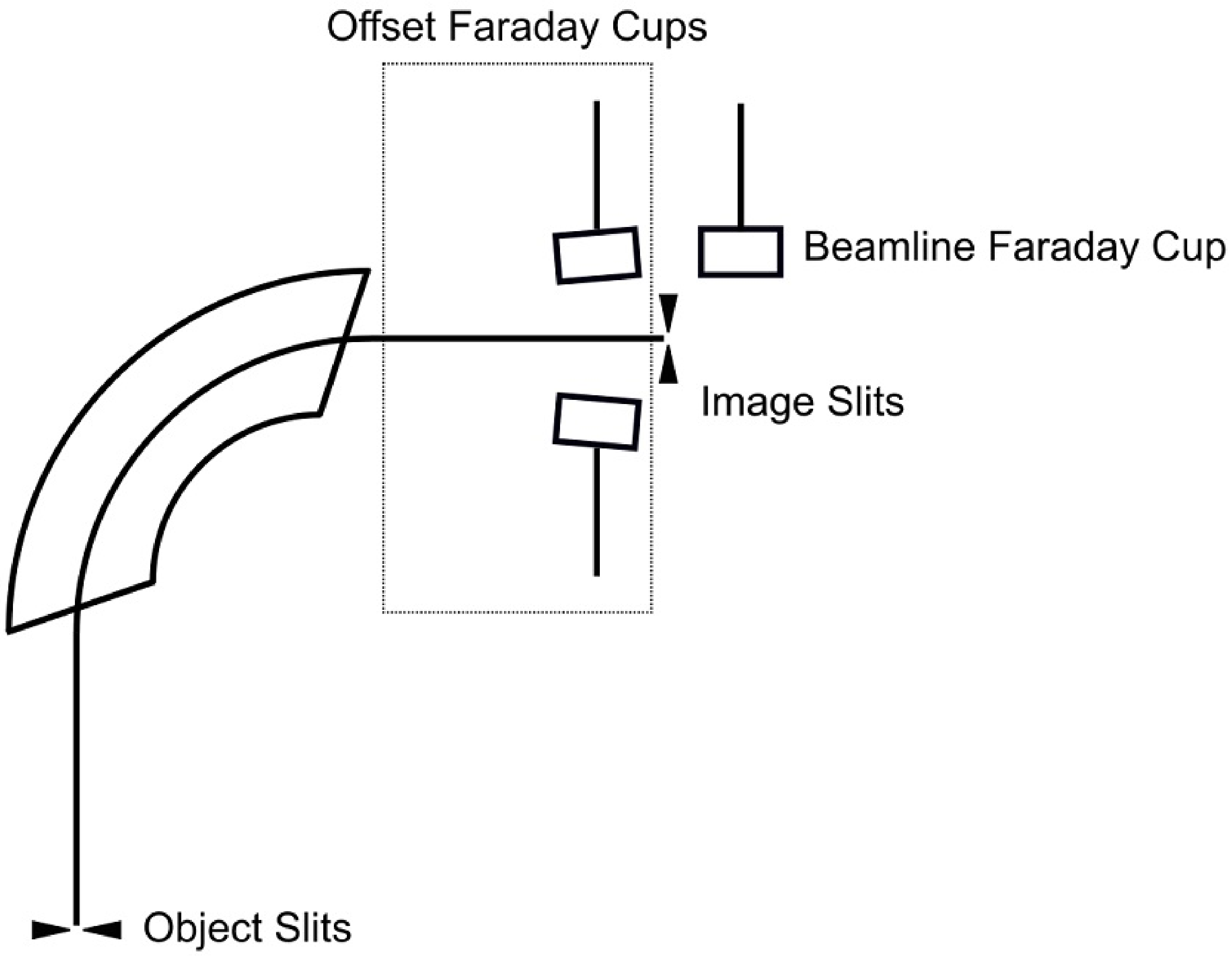
^{127}I INJECTION

- Separation of ^{127}I from ^{129}I easily achievable through manipulation of the injection slits
- Copper dimers appear in the spectrum, but will be broken up in the accelerator

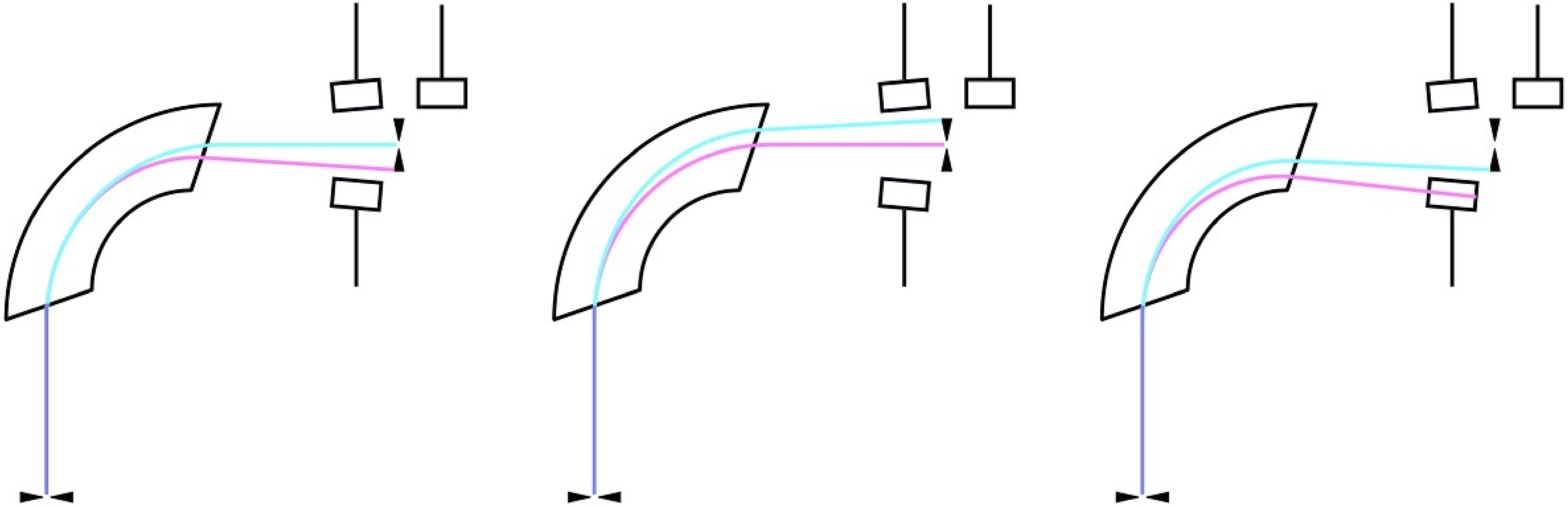


MID-RUN CURRENT MONITORING

- Offset Faraday cups allow for measuring stable beam currents
- Improves precision of measurements by accurately monitoring source output versus radionuclide counts
- However, some isotopes (^{127}I, for example) are not bent enough away from the radionuclide trajectory
  - Magnet Bias, Fast switching

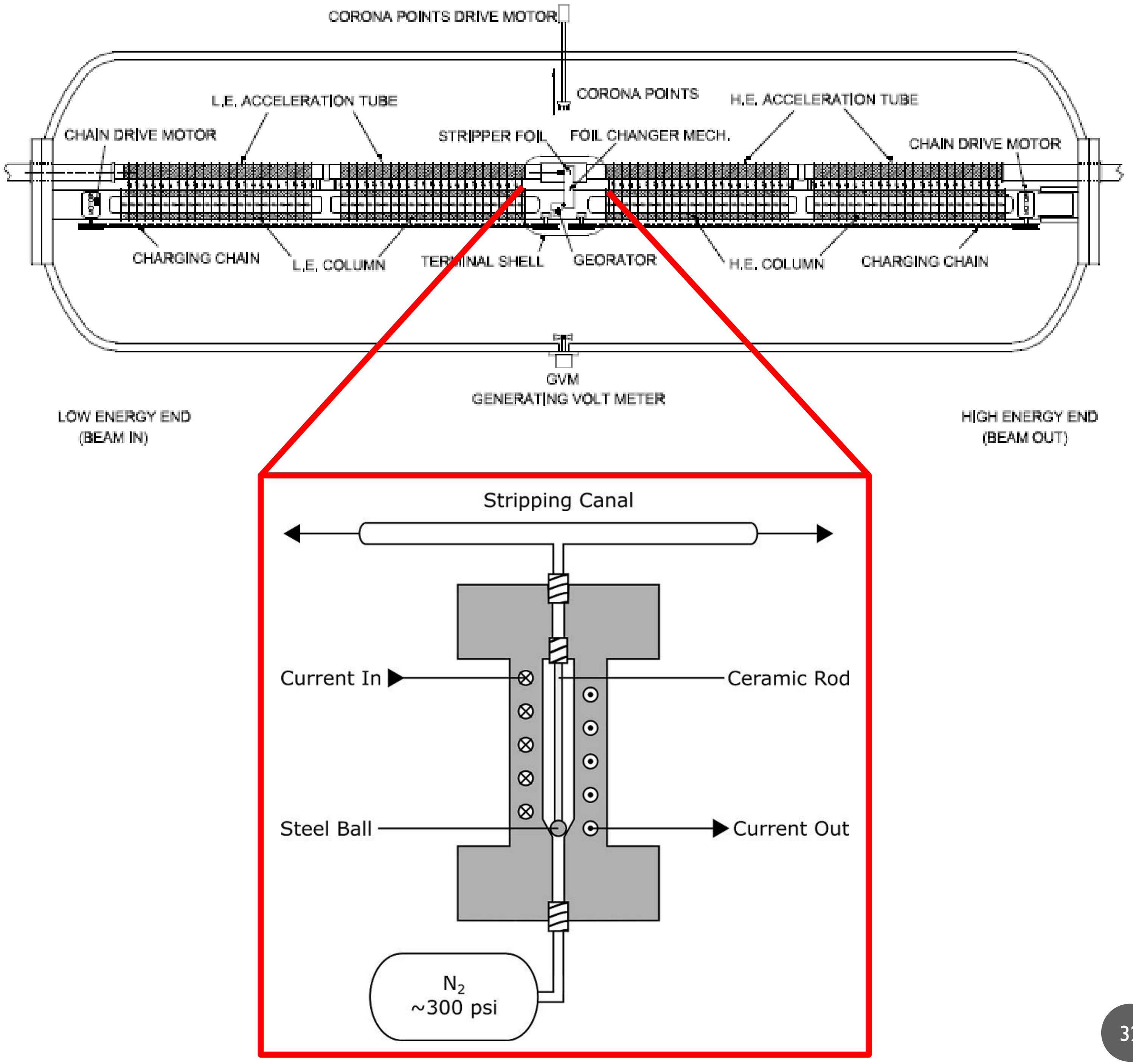


127I 129I



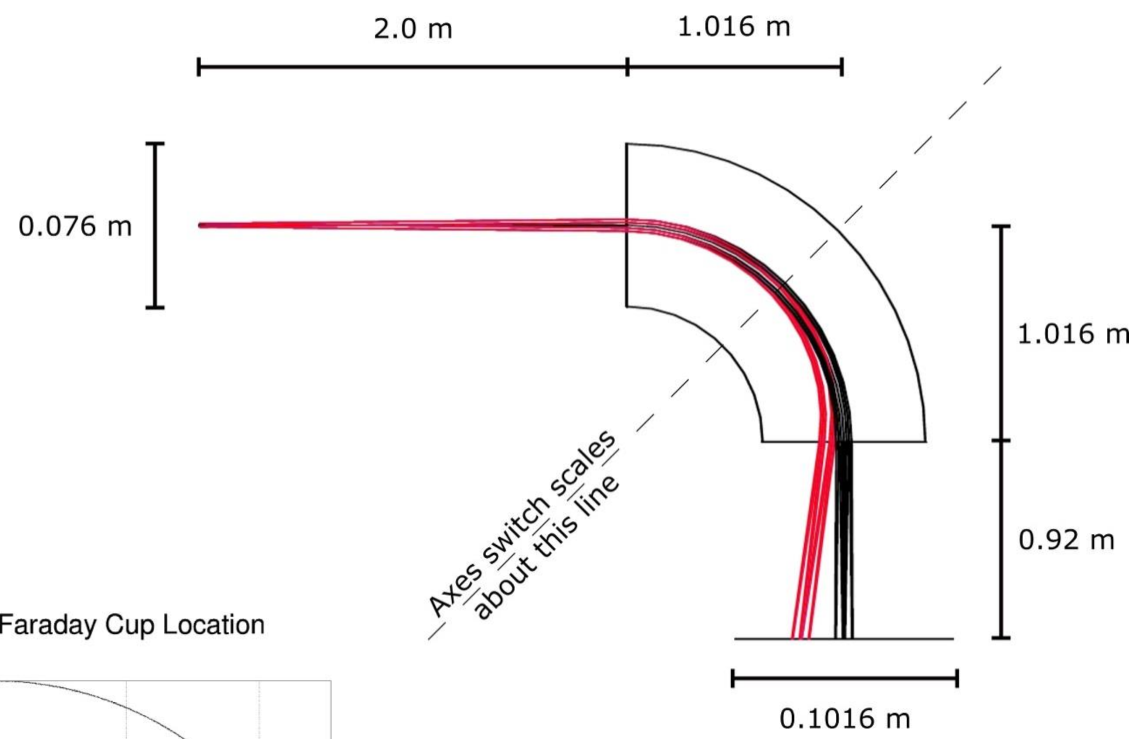
ACCELERATION: GAS STRIPPING

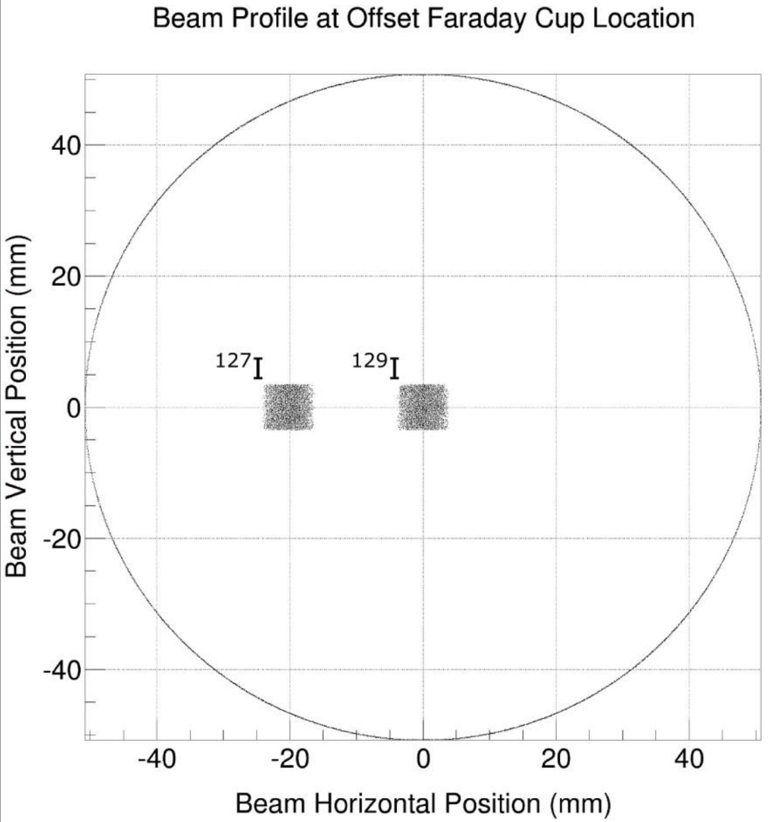
- Gas stripping doesn't degrade with time, and reduces straggling
  - Produces lower mean charge states
- Currently N_2 gas, but other gases could be explored
  - not easily changed, requires ~1 week downtime

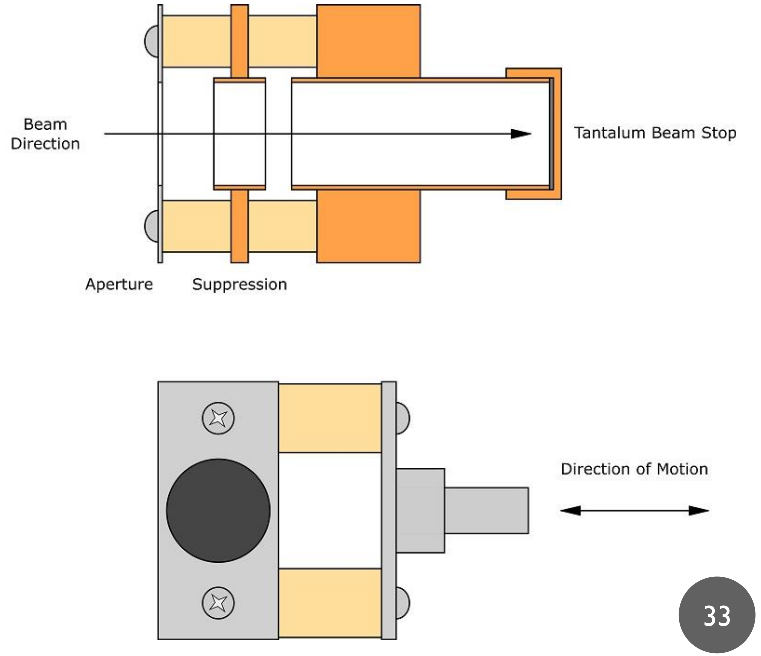


HIGH ENERGY ANALYSIS: OFFSET FARADAY CUP

- Fast switching allows injection of stable beams during cycle
  - Can measure beam transmission through the accelerator to monitor system stability
- Not included in the MRI, simulated with COSY Infinity using typical beam parameters for design and implementation in the lab
- Also useful for other isotopes (^{36}Cl, ^{41}Ca, ^{60}Fe)

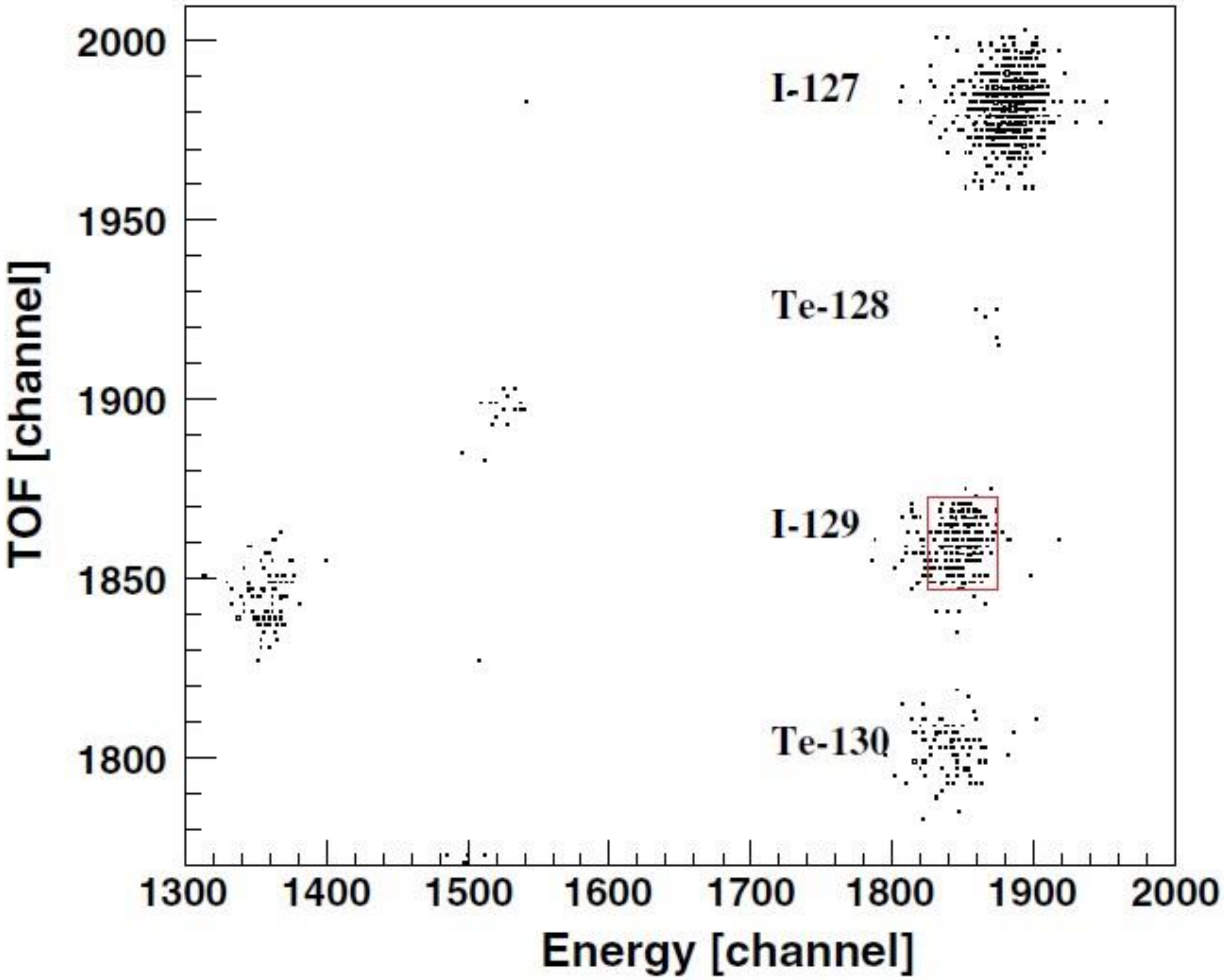




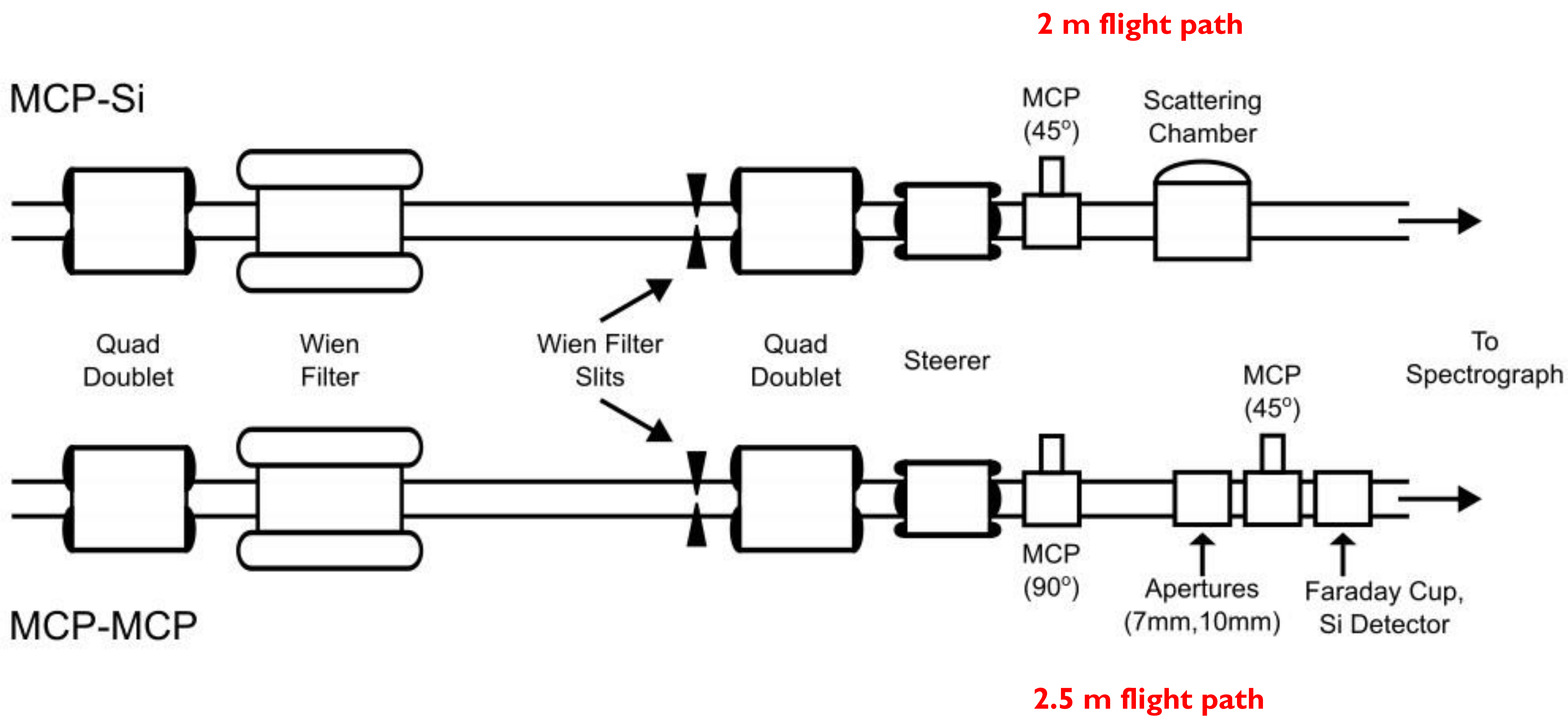


^{129}I ION DETECTION

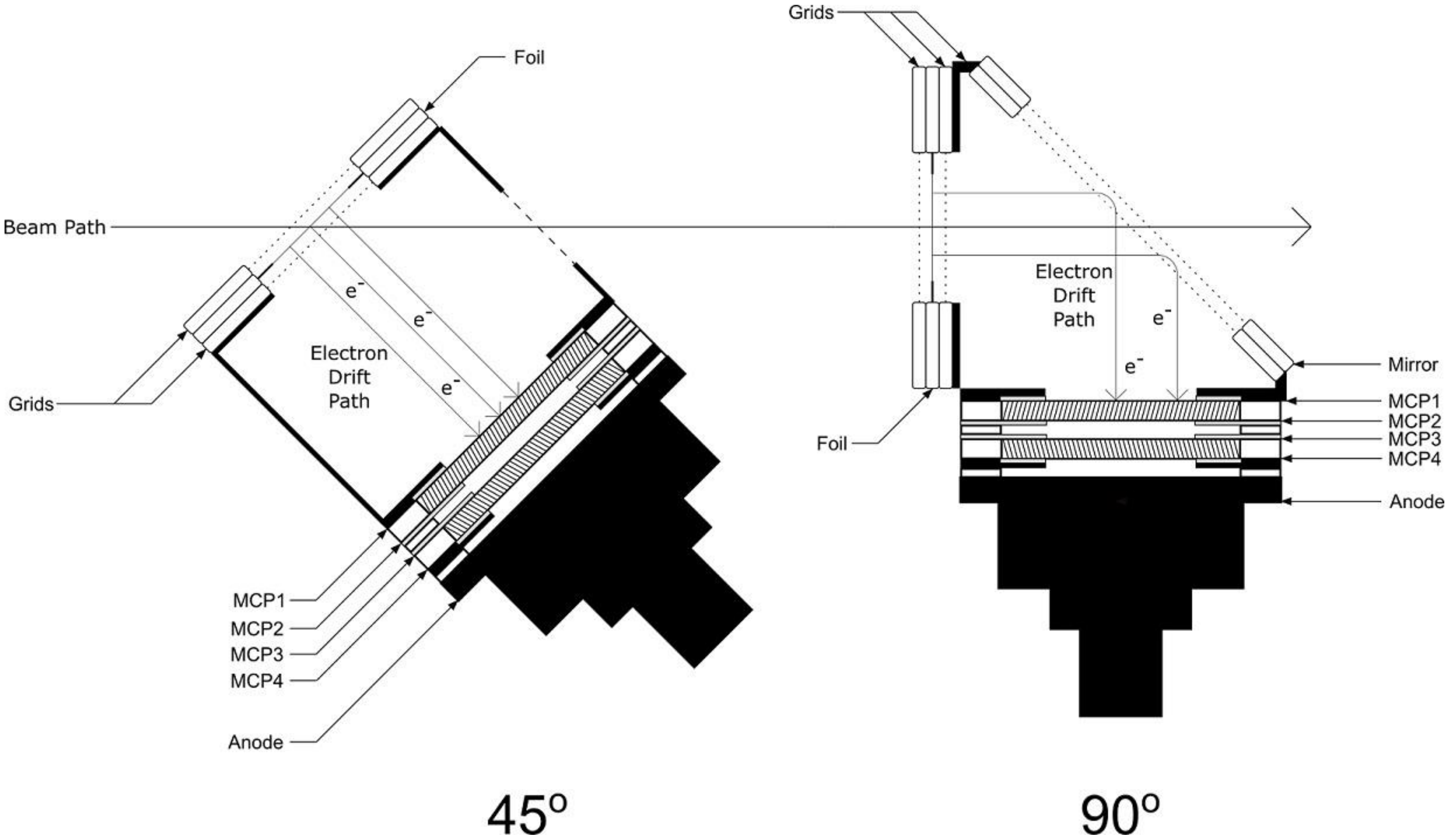
- The ^{129}Xe isobar isn't created
  - The contamination comes from ^{127}I tailing into the injection
- Energy separation isn't sufficient to resolve ^{129}I from ^{127}I
  - Time-of-flight (TOF) is required using MCPs, with energy measurement as a second identification parameter



Reithmeier (2005)



2 m flight path
MCP-Si
MCP
(45°)
Scattering
Chamber
Quad
Doublet
Wien
Filter
Wien Filter
Slits
Quad
Doublet
Steerer
To
Spectrograph
MCP
(45°)
MCP
(90°)
Apertures
(7mm,10mm)
Faraday Cup,
Si Detector
MCP-MCP
2.5 m flight path



Foil
Beam Path
Grids
Electron
Drift
Path
e-
e-
e-
MCP1
MCP2
MCP3
MCP4
Anode
45°
Grids
Foil
Electron
Drift
Path
e-
e-
Mirror
MCP1
MCP2
MCP3
MCP4
Anode
90°

*all ^{127}I spectra

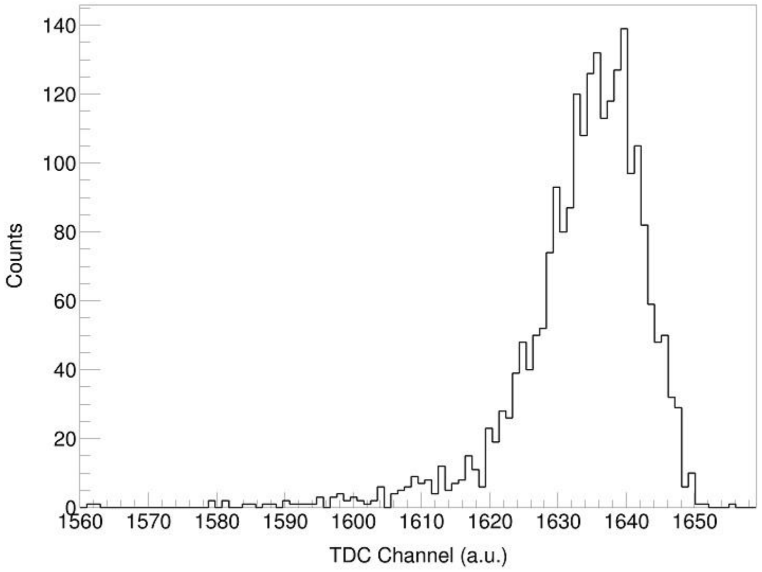
FIRST TOF DEVELOPMENT: MCP-SI

E_{129} = 57.6 MeV, q = 8+
ΔTOF = ~1.7 ns

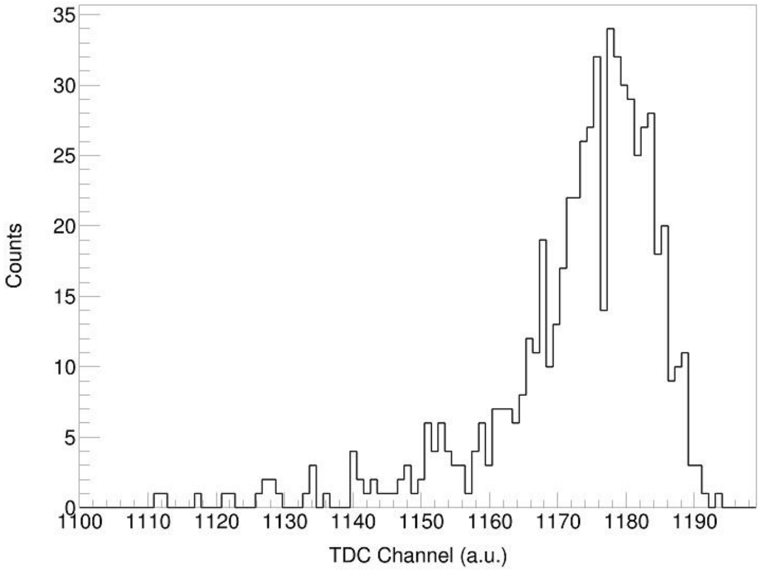
E_{129} = 78.8 MeV, q = 9+
ΔTOF = ~1.4 ns

MCP-Si

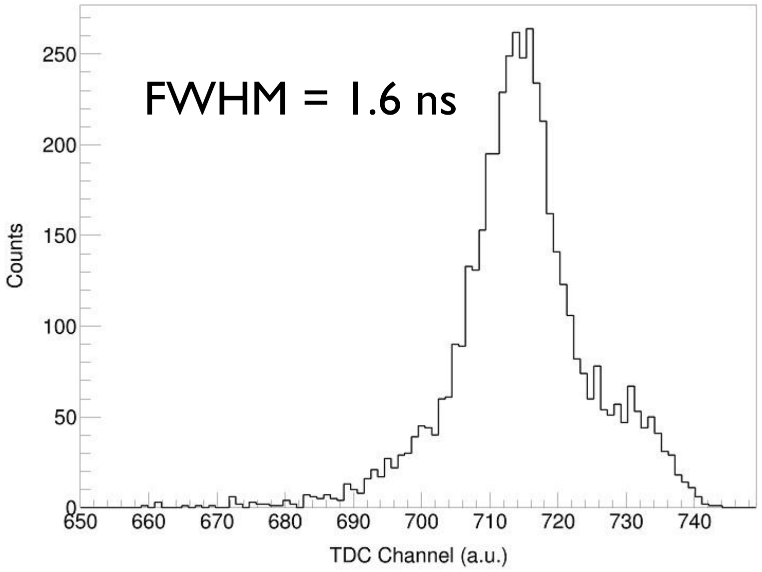
FWHM = 1.9 ns



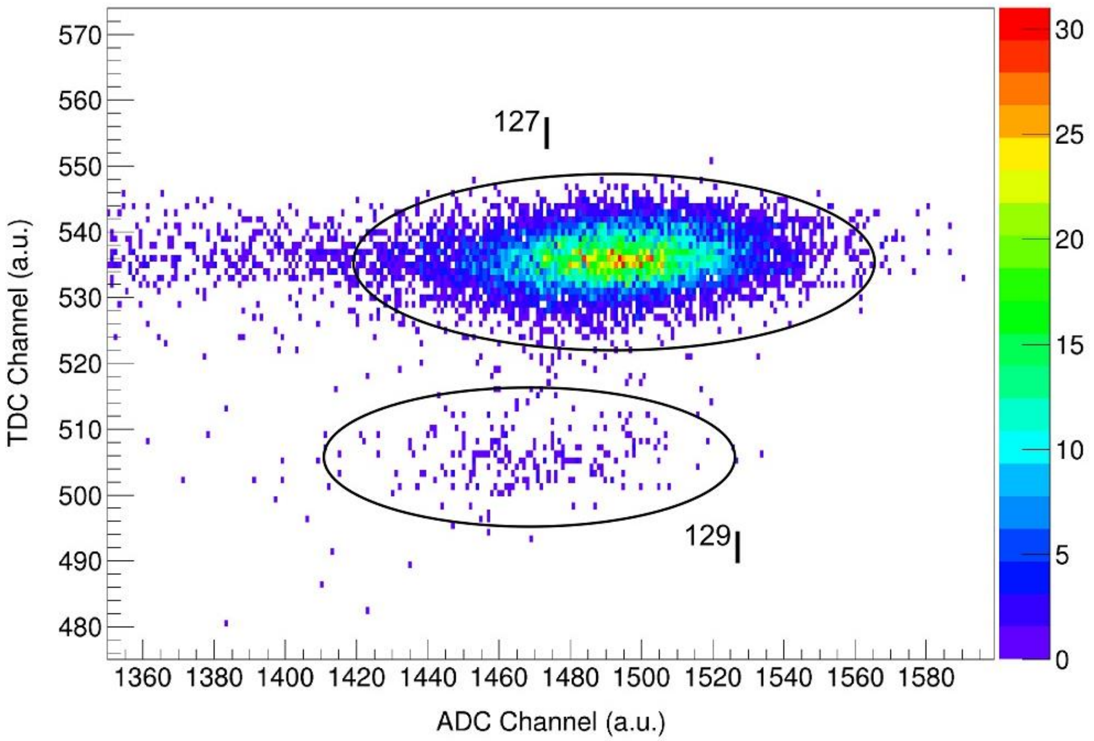
MCP-PIN



MCP-Si



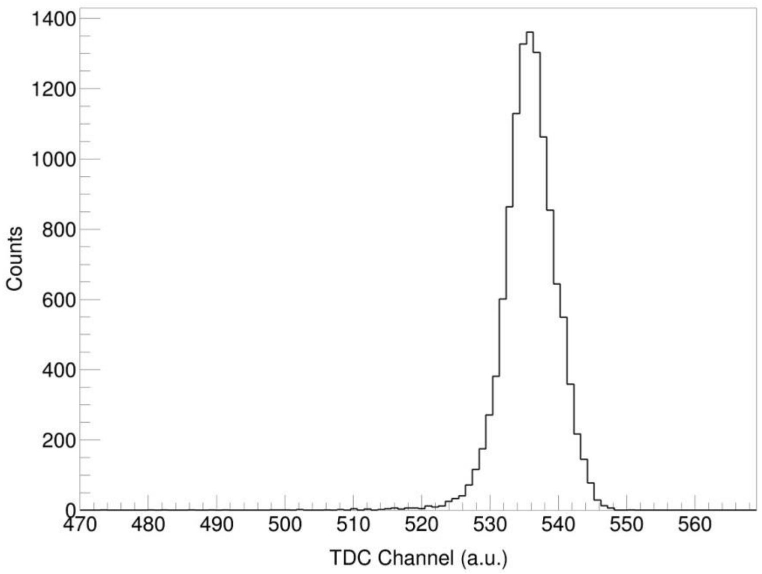
SECOND TOF DEVELOPMENT: MCP-MCP



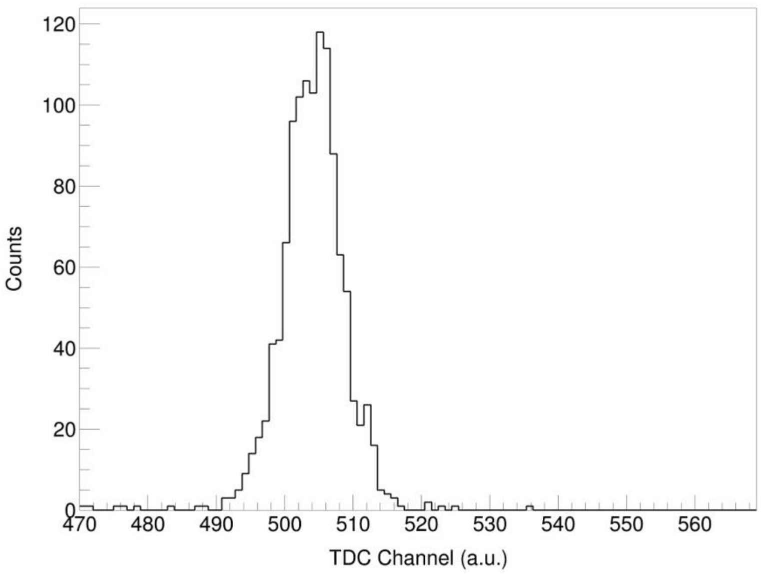
E_{129} = 78.8 MeV, q = 9+
ΔTOF = ~1.8 ns

FWHM = 870 ns

^{127}I

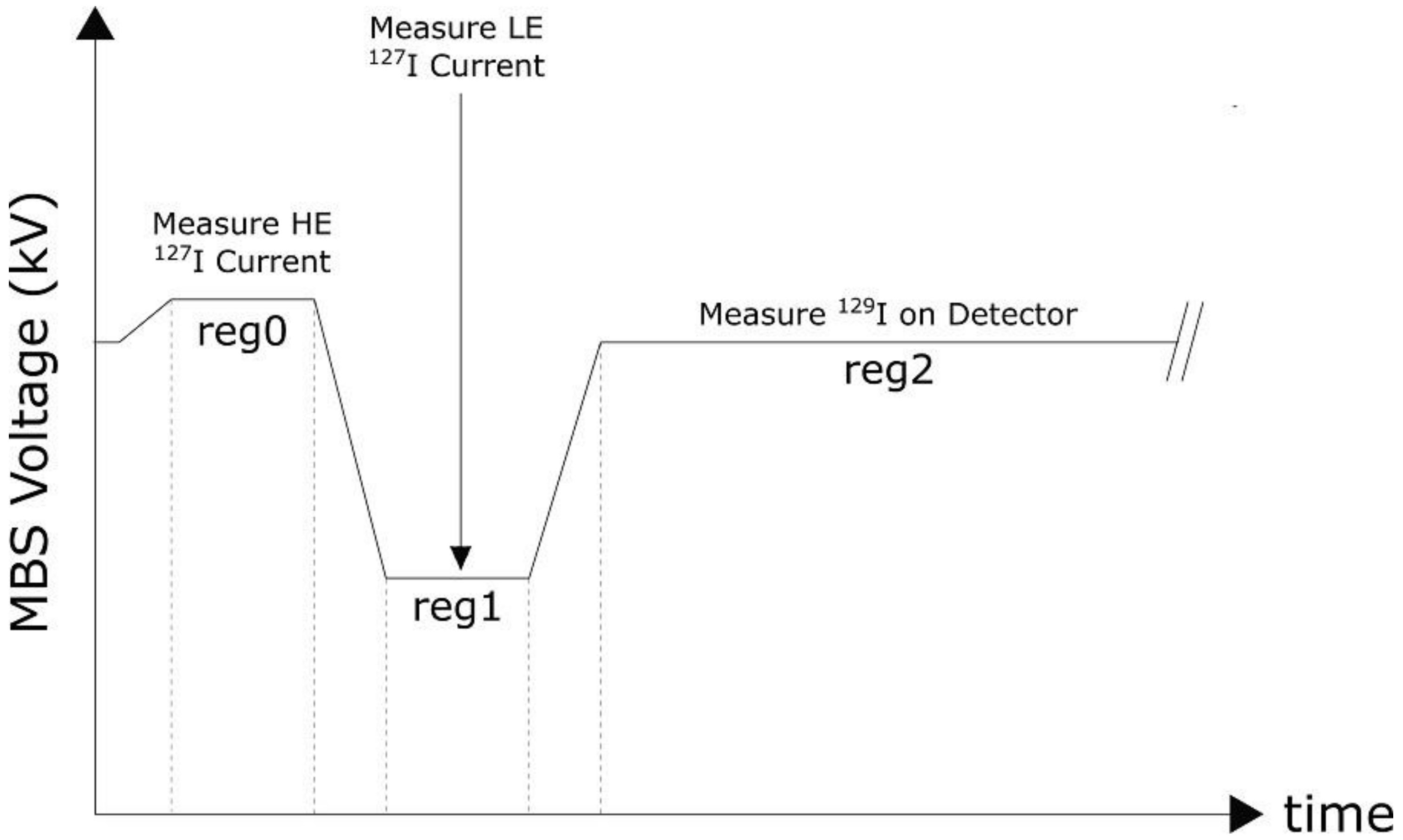


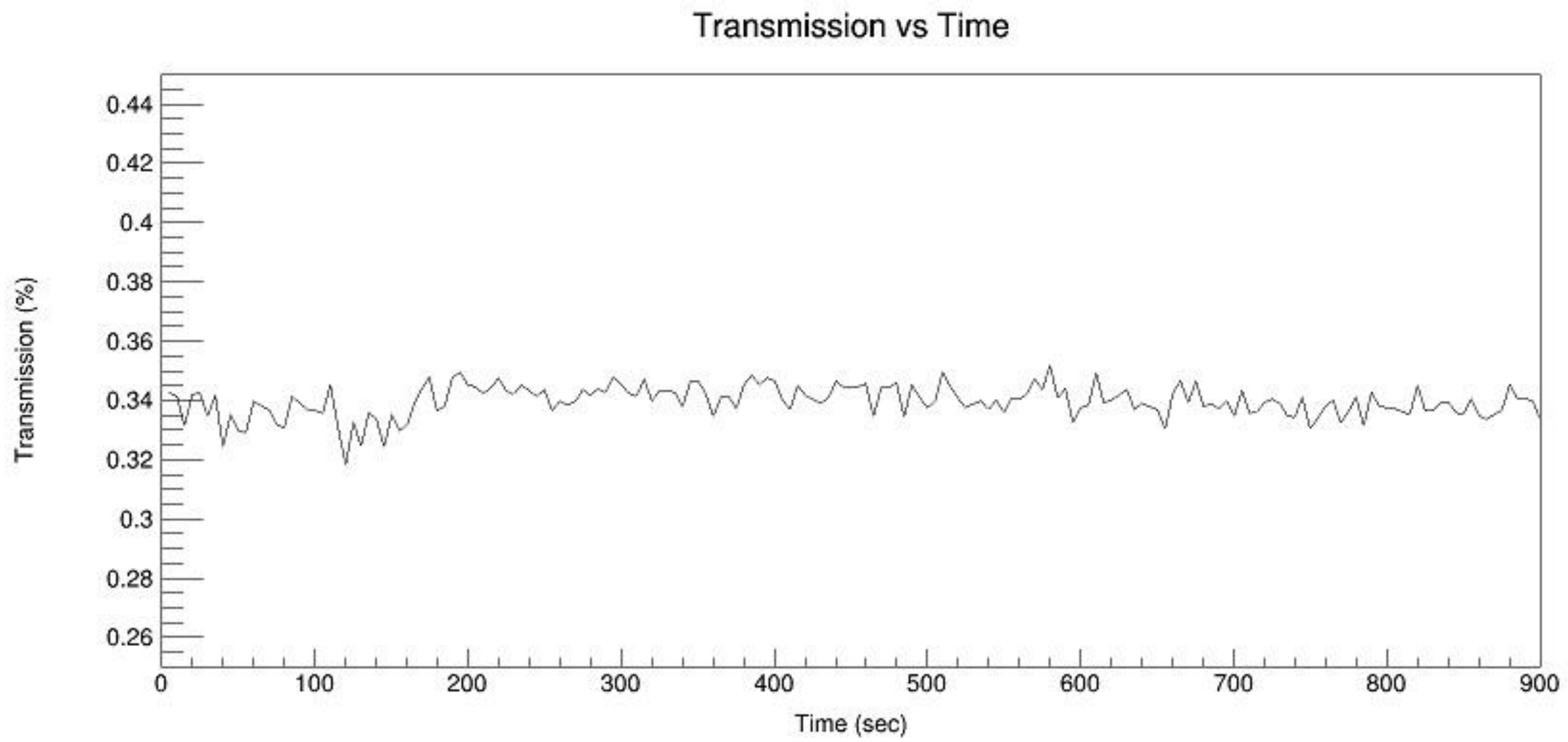
^{129}I



FAST SWITCHING SEQUENCE

- Measure currents for ~100 μs
- Measure radionuclide for ~90,000 μs
- MBS gate can veto counts that show up during non-radionuclide states

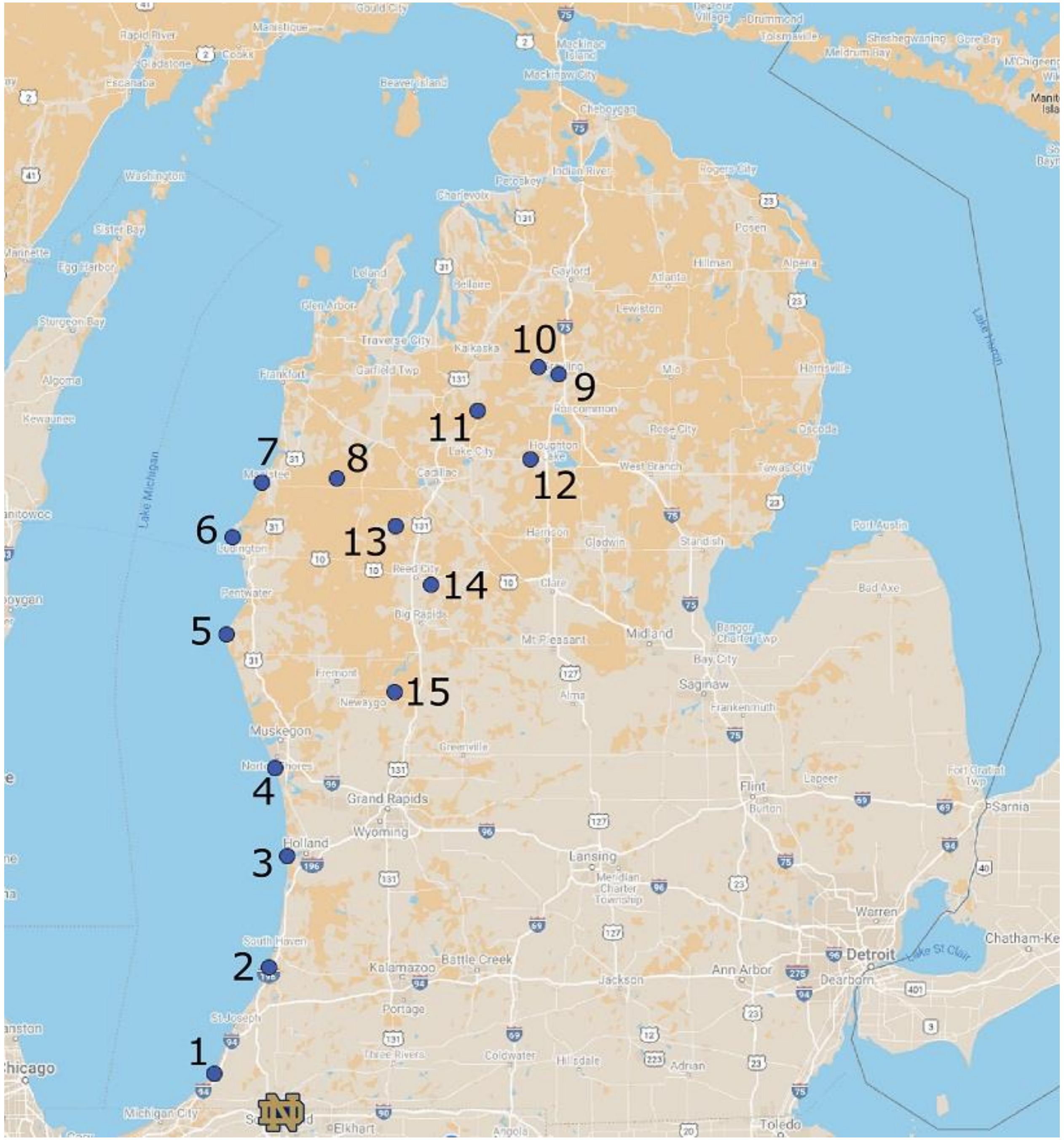


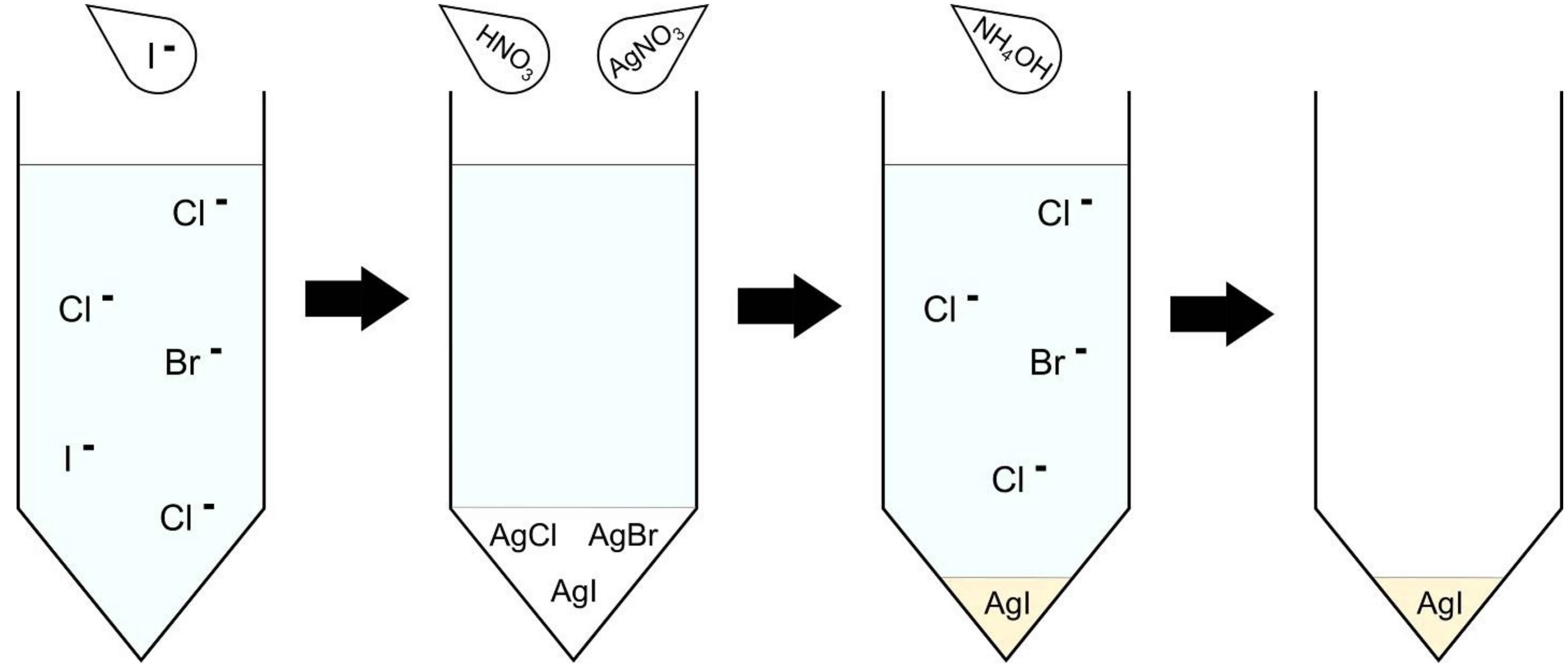

Transmission vs Time
Transmission (%)
0.44
0.42
0.4
0.38
0.36
0.34
0.32
0.3
0.28
0.26
0
100
200
300
400
500
600
700
800
900
Time (sec)

SAMPLE COLLECTION AND CHEMISTRY

SAMPLING LOCATIONS

- 15 samples
  - 7 Lake Michigan samples
  - 4 rivers
- 250 mL filtered water collected





Stable iodine is added via a carrier solution to the water sample to fix the $^{129}I/I$ ratio

Nitric acid is added for the precipitation of AgI (and AgCl, AgBr) using $AgNO_3$

Ammonium hydroxide is added to return AgCl, AgBr to solution, leaving AgI precipitate

Solution is decanted, leaving only AgI to be dried overnight in an oven

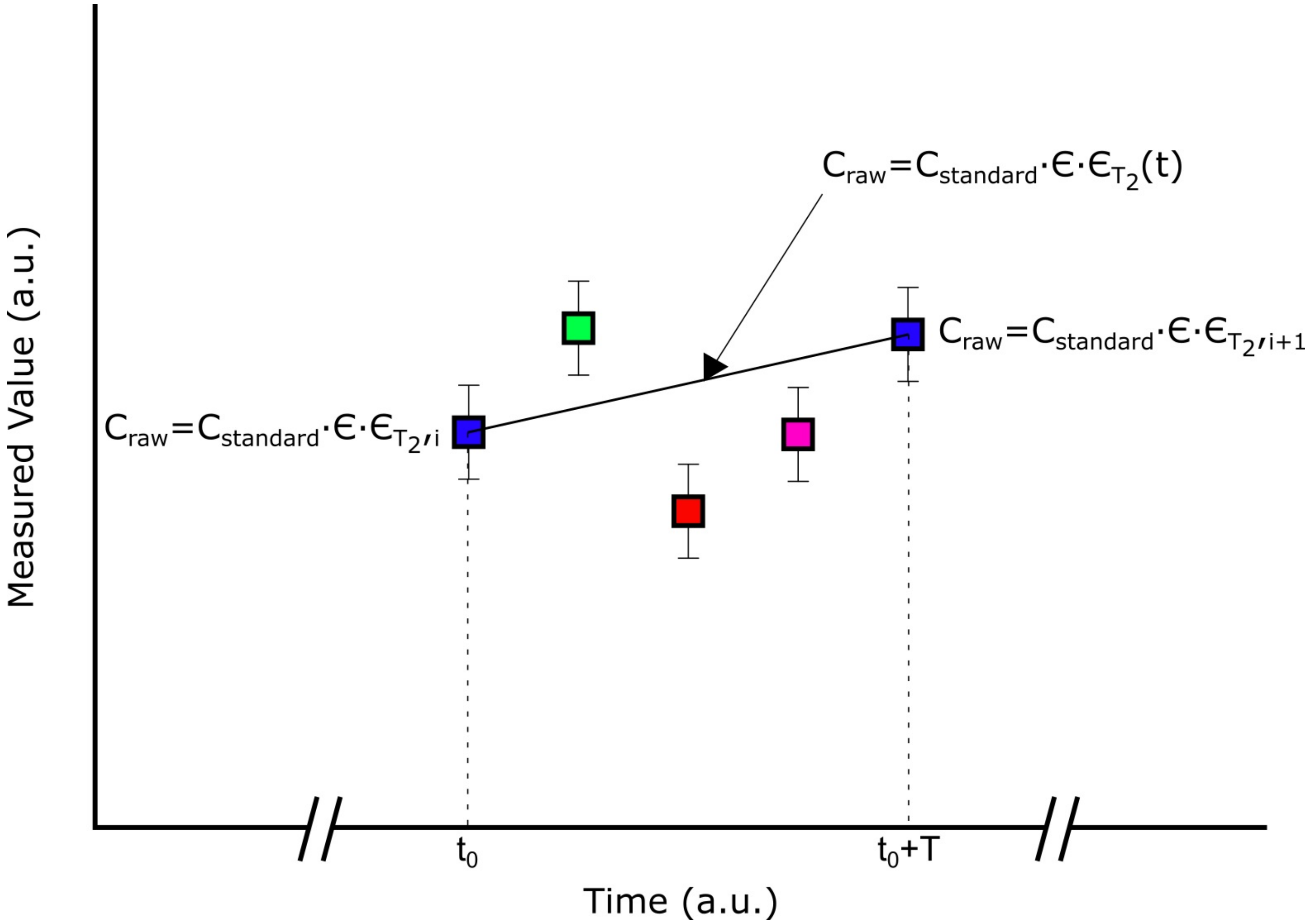
^{129}I MEASUREMENT AND RESULTS

EXPERIMENTAL PROCEDURE

- Measurement of AMS standards for system validation
- Measurement cycle for water samples
  - Standards – system variation
  - Blanks – background level
  - Samples
- Sample concentration correction
- Establish ^{129}I content of sample water

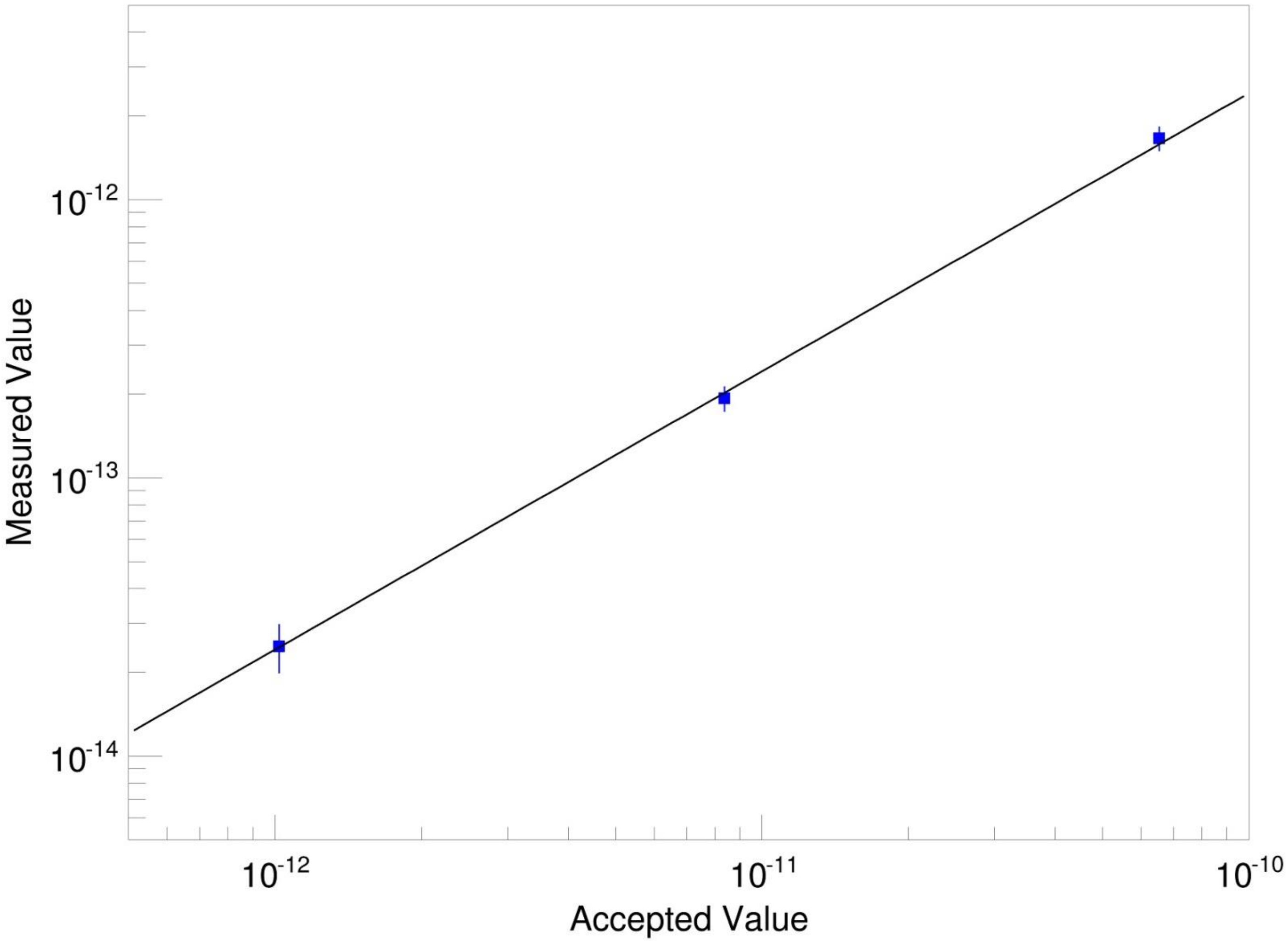
AMS STANDARD CORRECTION

- Absolute measurements are difficult to perform without accurately characterizing the system from start to finish
- In place of absolute measurements, AMS perform measurements relative to AMS standards (known isotopic ratio)
- Standard measurements absorb different efficiencies into a correction factor

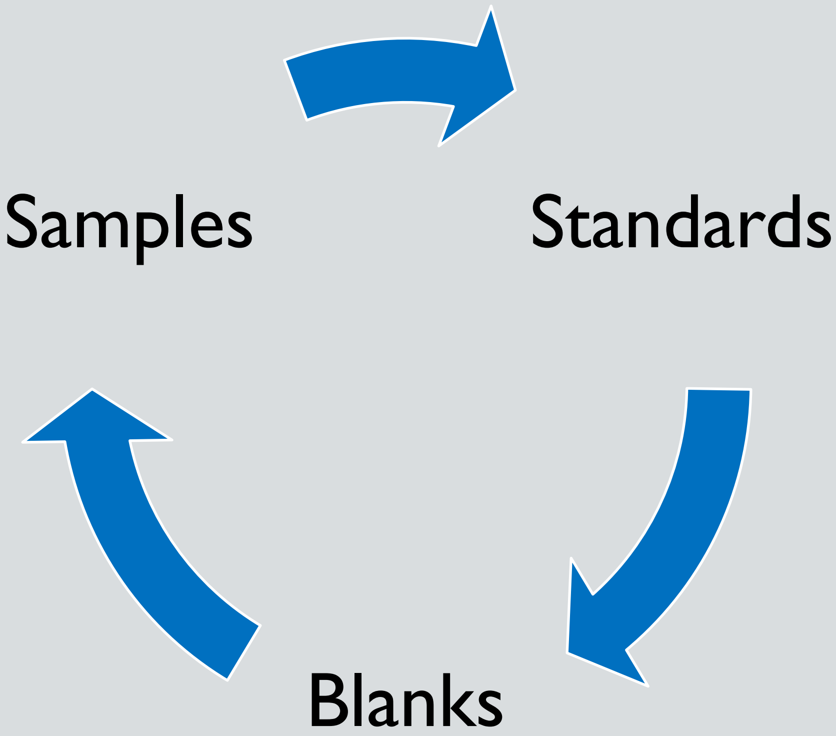


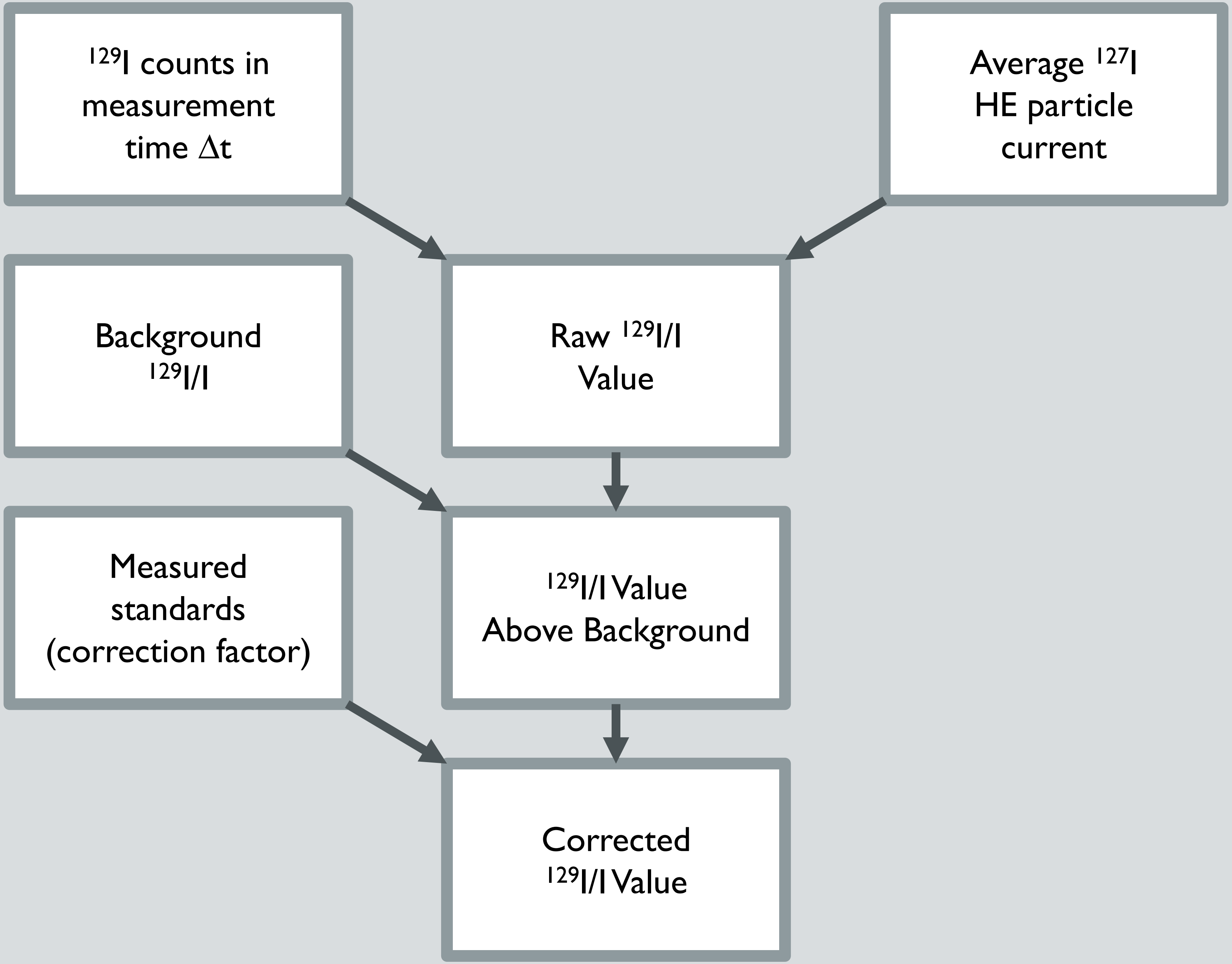
MEASUREMENT OF STANDARDS

- Confirming the measured ratio of standards matches the nominal ratio demonstrates that the system can accurately measure a range of isotopic concentrations
  - The three standards show a linear trend, with a slope of 0.0241 and consistent with an x-intercept of 0
    - Suggests ~2.4% of expected ^{129}I counts are being detected



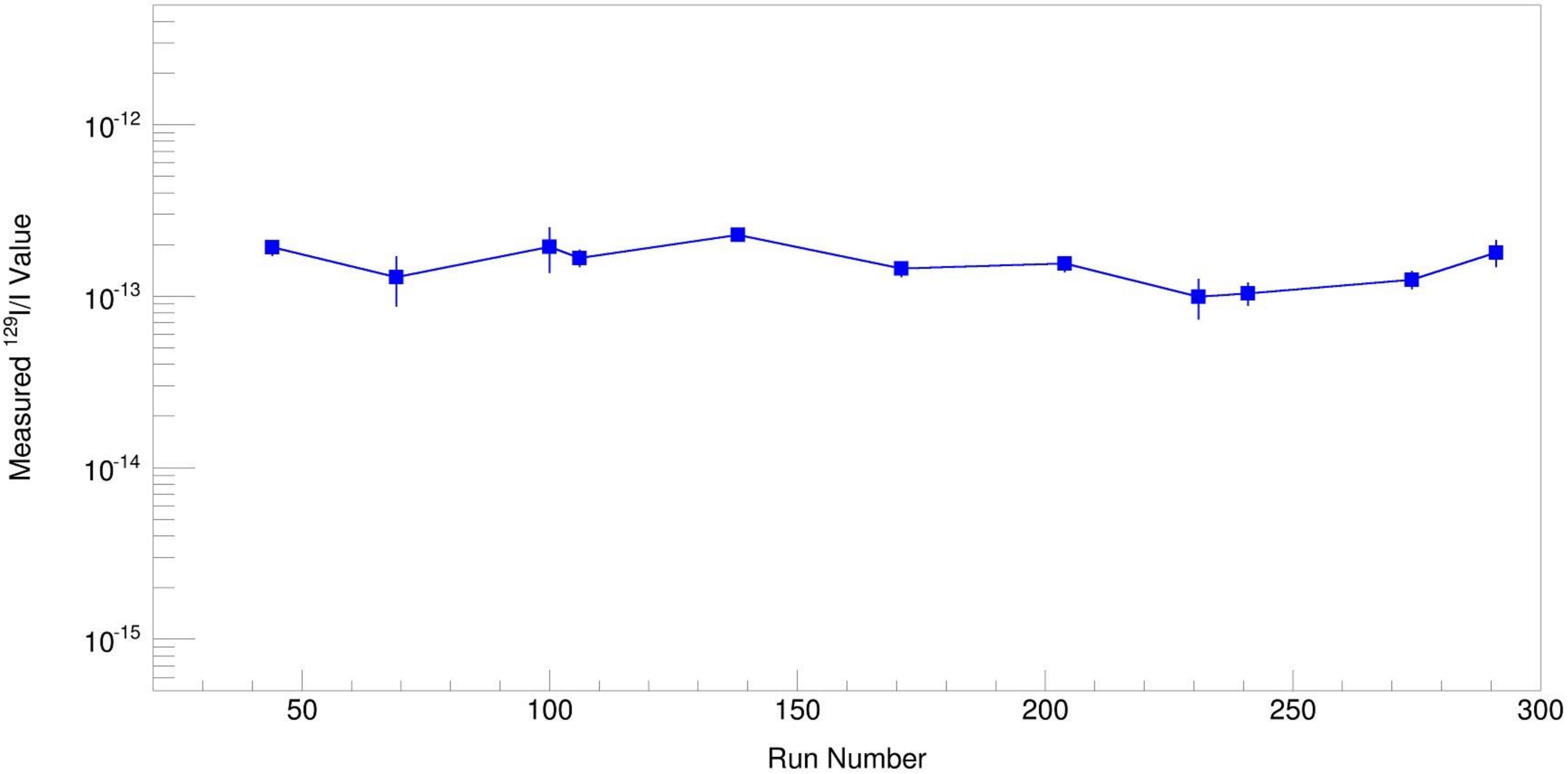
SAMPLE MEASUREMENT





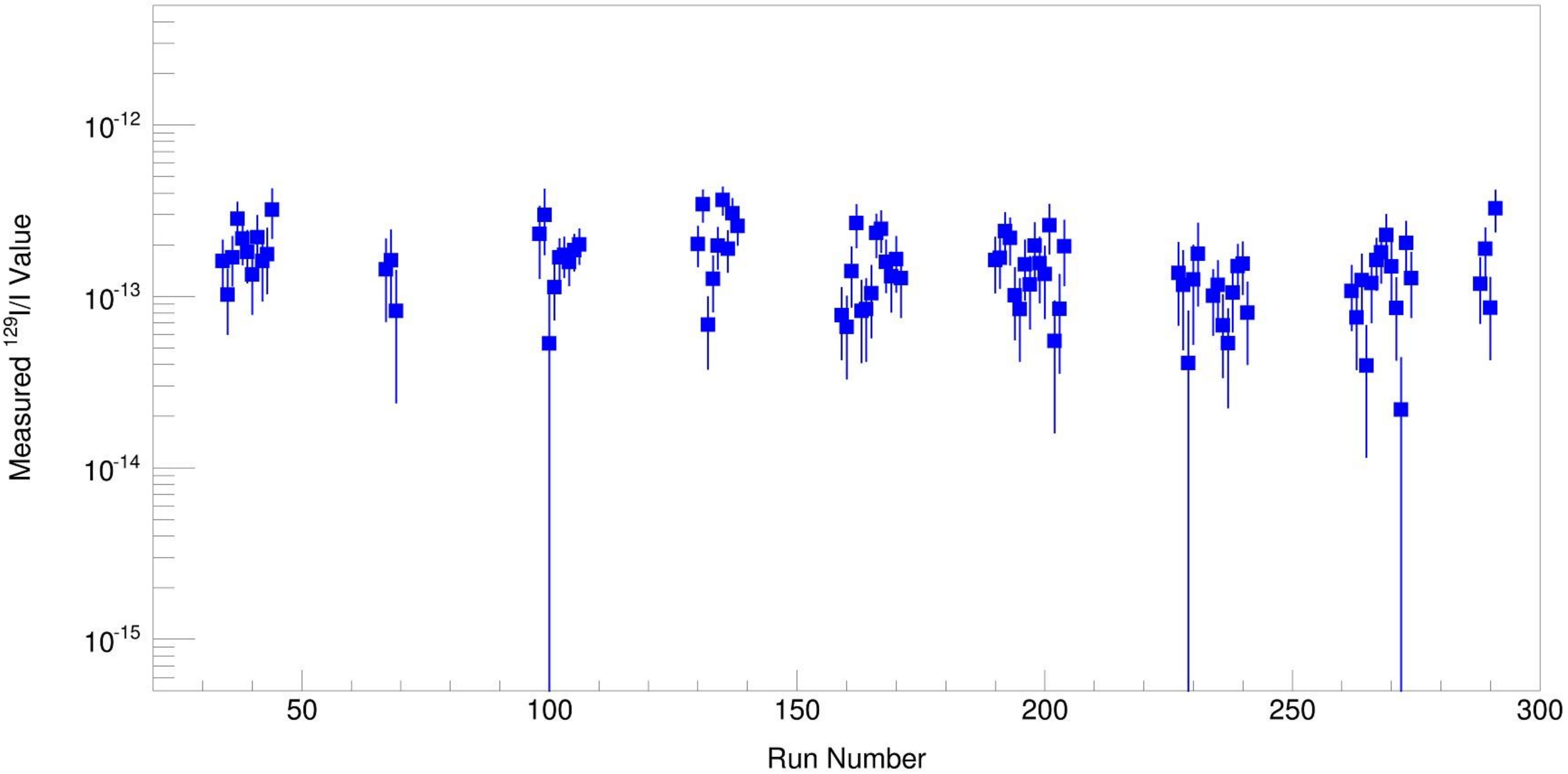
^{129}I counts in measurement time Δt
Average ^{127}I HE particle current
Background ^{129}I/I
Raw ^{129}I/I Value
Measured standards (correction factor)
^{129}I/I Value Above Background
Corrected ^{129}I/I Value

Standards

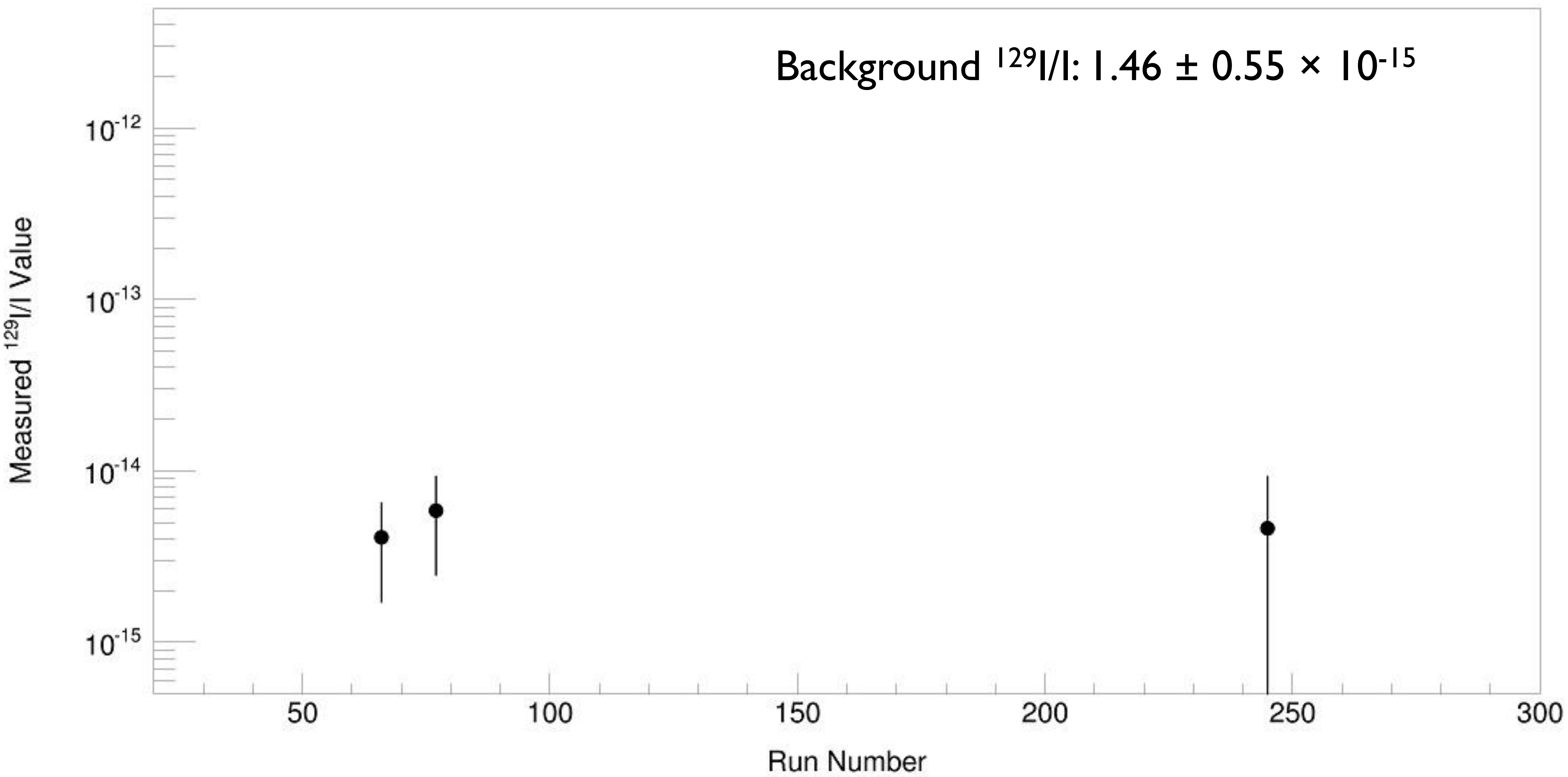


Measured ^{129}I/I Value
10^{-12}
10^{-13}
10^{-14}
10^{-15}
50
100
150
200
250
300
Run Number

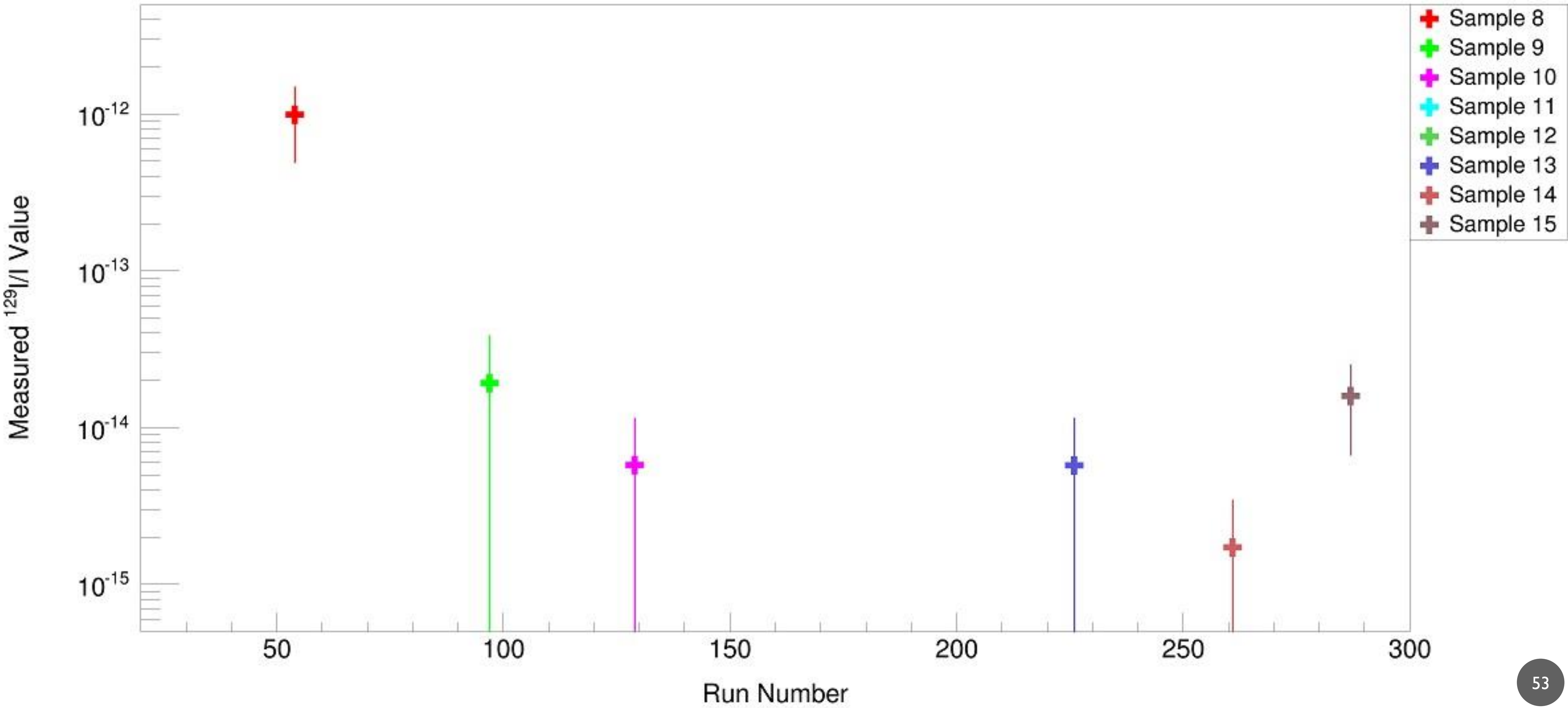
Standards by run



Blanks

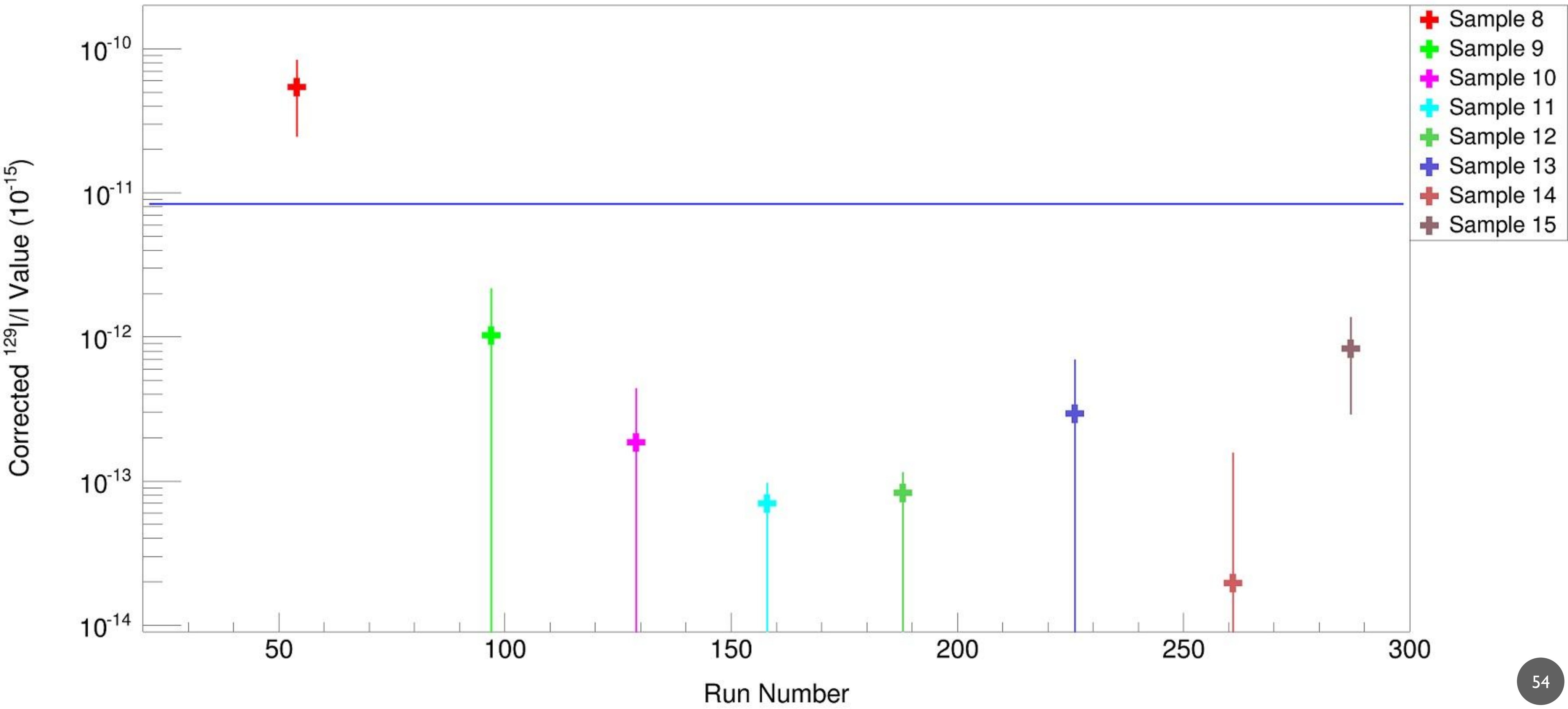

Background ^{129}I/I: 1.46 ± 0.55 × 10^{-15}
Measured ^{129}I/I Value
10^{-12}
10^{-13}
10^{-14}
10^{-15}
50
100
150
200
250
300
Run Number

Samples



Measured ^{129}I/I Value
10^{-12}
10^{-13}
10^{-14}
10^{-15}
50
100
150
200
250
300
Run Number
Sample 8
Sample 9
Sample 10
Sample 11
Sample 12
Sample 13
Sample 14
Sample 15

Samples, corrected

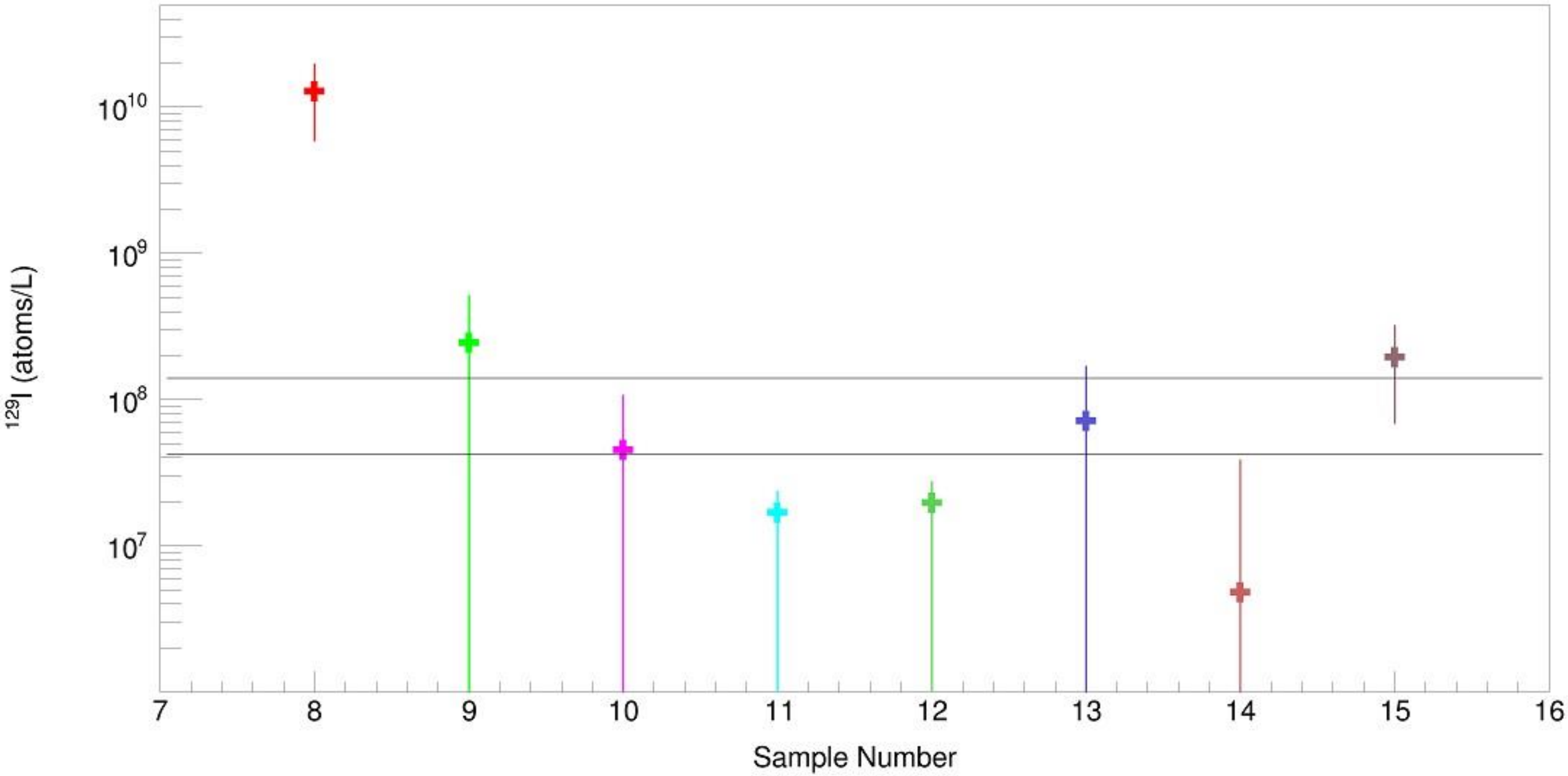


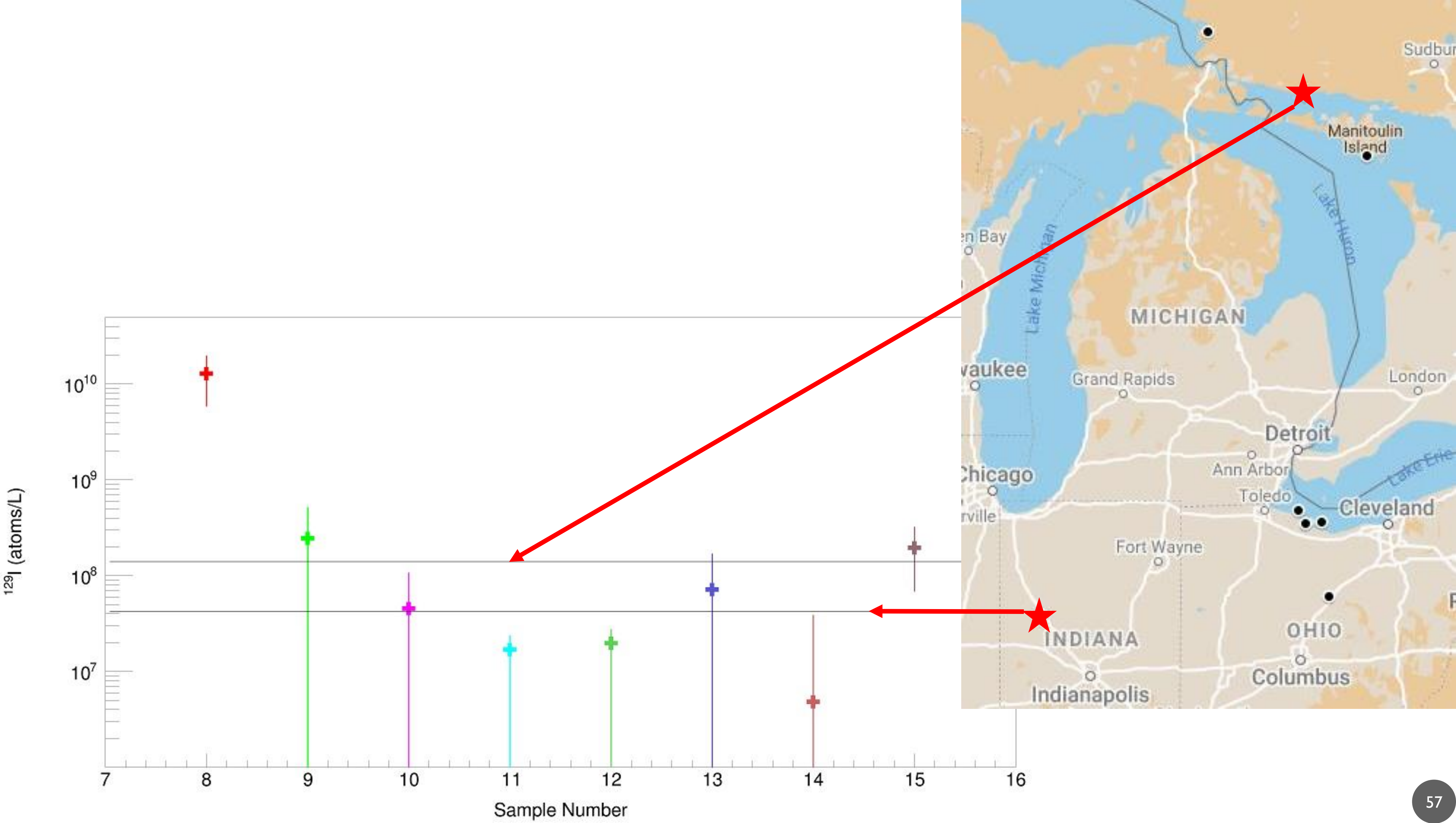
^{129}I CONTENT PER UNIT VOLUME FOR THE MEASURED SAMPLES

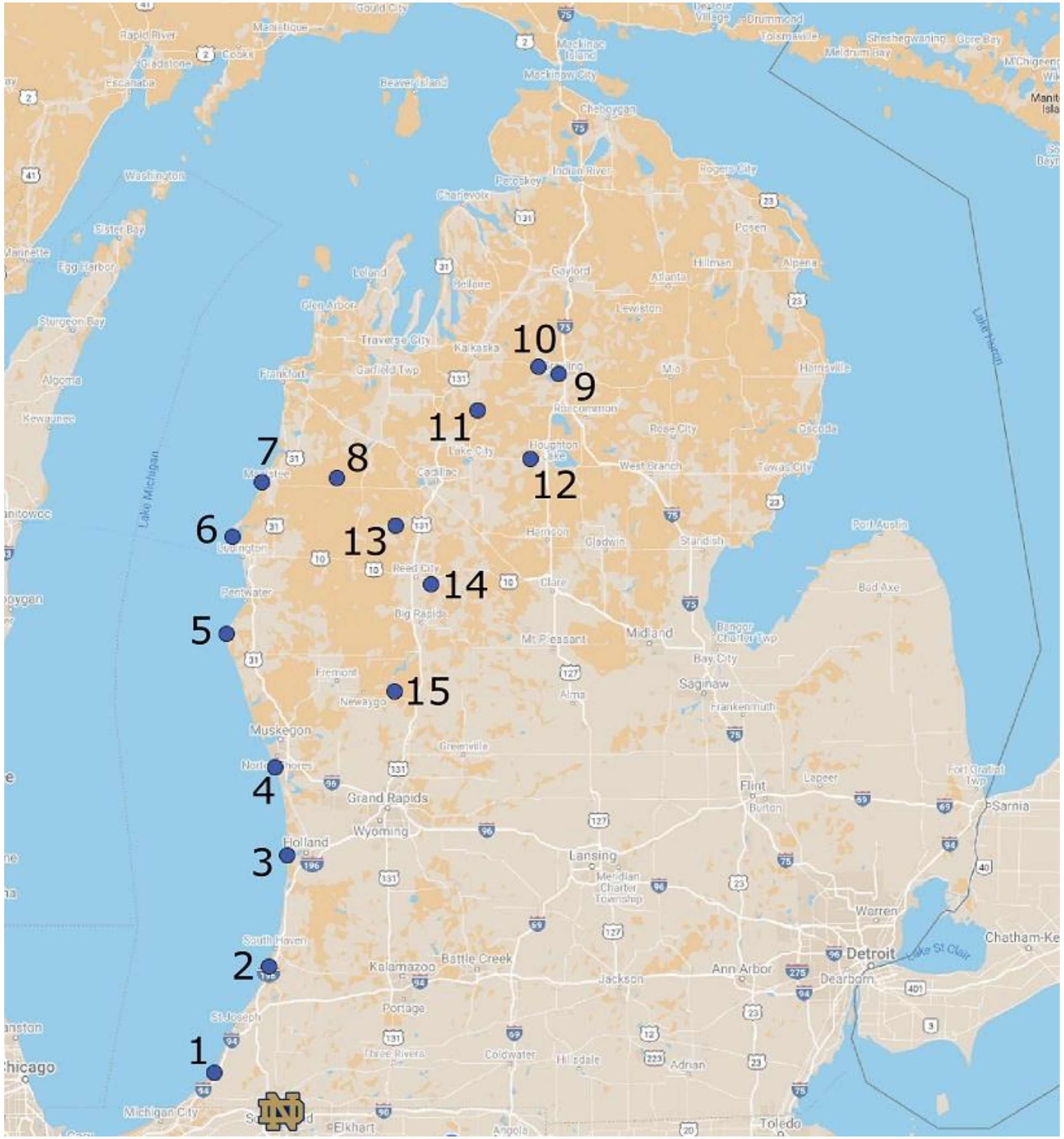
Sample	N_{127} (10^{18})	^{129}I/I (10^{-15})	N_{129} (10^{6})	Volume (mL)	^{129}I $(10^{6}$ atoms/L)
8	9.49	53868.81 ± 28971.57	511.02 ± 274.84	40.00 ± 0.50	12775.55 ± 6872.77
9	9.52	1023.91 ± 1125.10	9.75 ± 10.71	40.00 ± 0.50	243.72 ± 267.82
10-2	9.73	185.60 ± 250.37	1.81 ± 2.44	40.00 ± 0.50	45.16 ± 60.92
11	9.70	<69.61 ± 26.67[a]	<0.67 ± 0.26[a]	40.00 ± 0.50	<16.87 ± 6.47[a]
12	9.51	<82.44 ± 31.61[a]	<0.78 ± 0.30[a]	40.00 ± 0.50	<19.60 ± 7.52[a]
13	9.75	292.20 ± 394.97	2.85 ± 3.85	40.00 ± 0.50	71.26 ± 96.32
14	9.84	19.56 ± 135.84	0.19 ± 1.34	40.00 ± 0.50	4.81 ± 33.42
15	9.39	828.43 ± 533.78	7.78 ± 5.01	40.00 ± 0.50	194.57 ± 125.39

[a] Upper limit

Samples, ^{129}I content in water




^{129}I (atoms/L)
10^{10}
10^9
10^8
10^7
7
8
9
10
11
12
13
14
15
16
Sample Number
Sudbury
Manitoulin Island
Lake Huron
Lake Michigan
MICHIGAN
Green Bay
Milwaukee
Grand Rapids
London
Detroit
Ann Arbor
Chicago
Toledo
Lake Erie
Cleveland
Merrillville
Fort Wayne
INDIANA
OHIO
Columbus
Indianapolis


10
9
11
8
7
12
6
13
14
5
15
4
3
2
1
Traverse City
Muskegon
Grand Rapids
Holland
Lansing
Kalamazoo
Battle Creek
Flint
Saginaw
Detroit

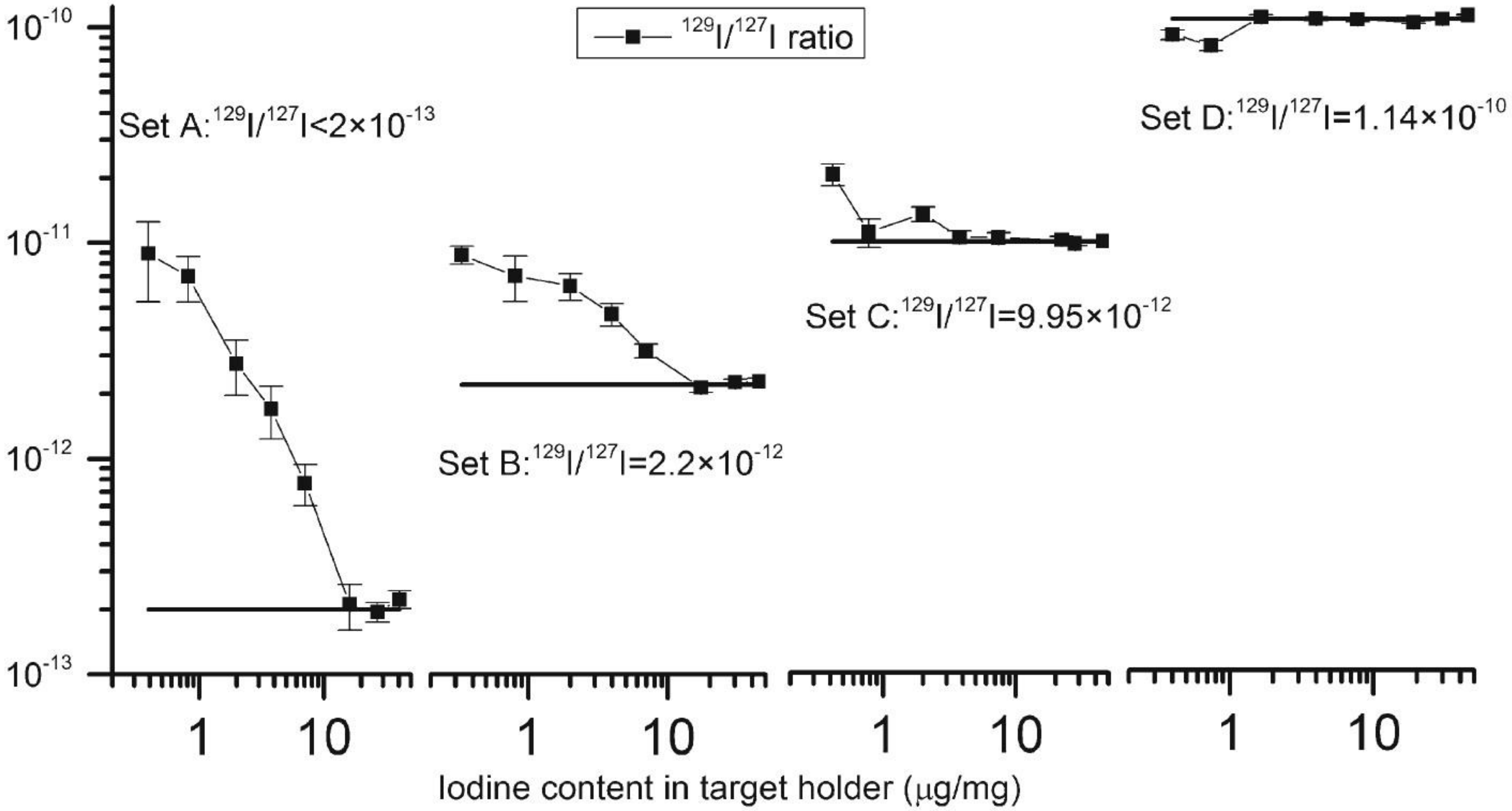

Manitoulin Island
Lake Michigan
Lake Huron
MICHIGAN
Grand Rapids
London
Detroit
Ann Arbor
Toledo
Cleveland
Chicago
Fort Wayne
INDIANA
OHIO
Columbus
Indianapolis

ONGOING/FUTURE WORK

PLANNED ^{129}I DEVELOPMENT

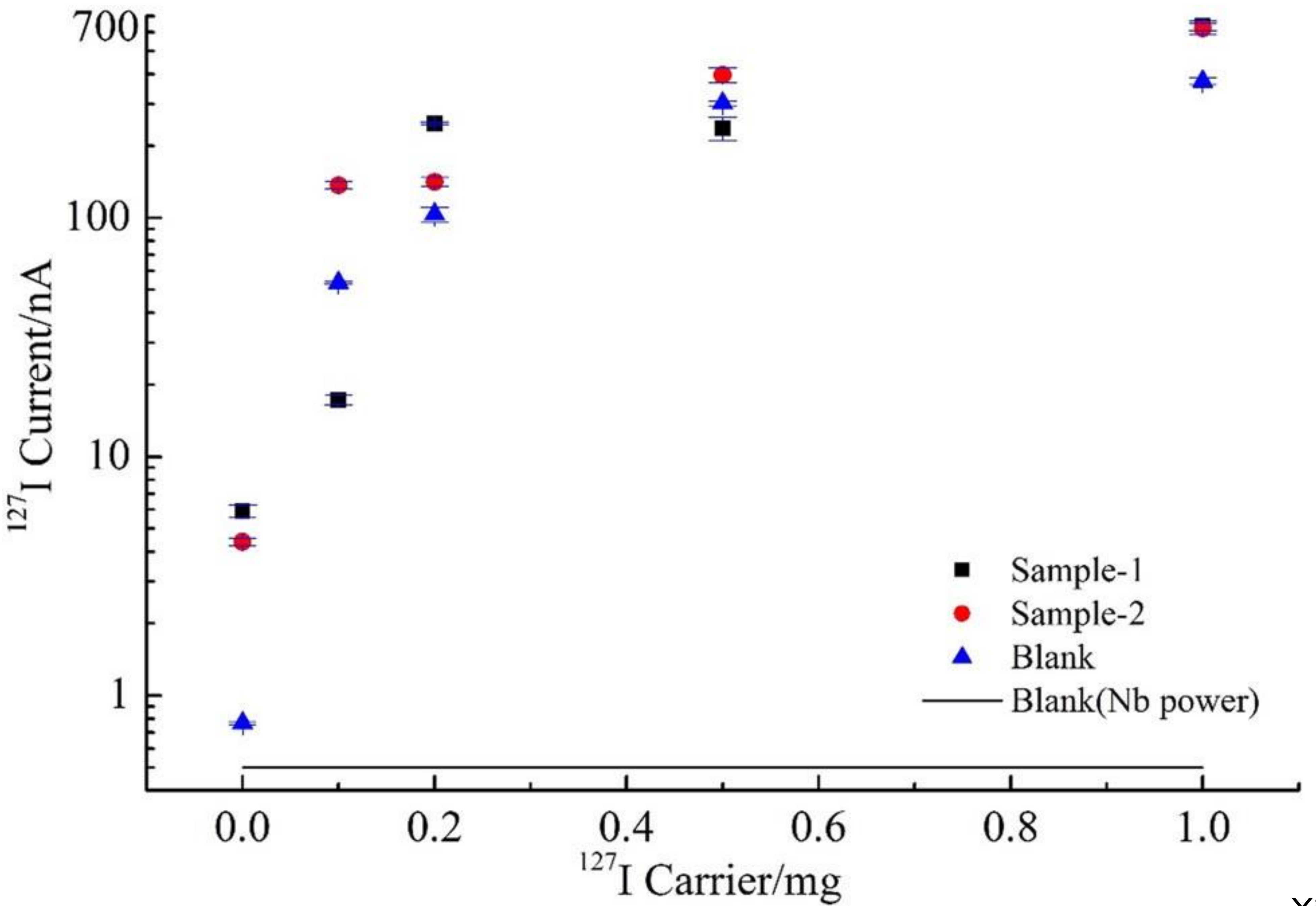
- Experimental
  - **Coprecipitation with AgCl**
  - Widening injection, analysis magnet slits

- Technical
  - Recirculating gas stripping
  - Retractable ionization chamber
  - FN regulation using HE offset cup



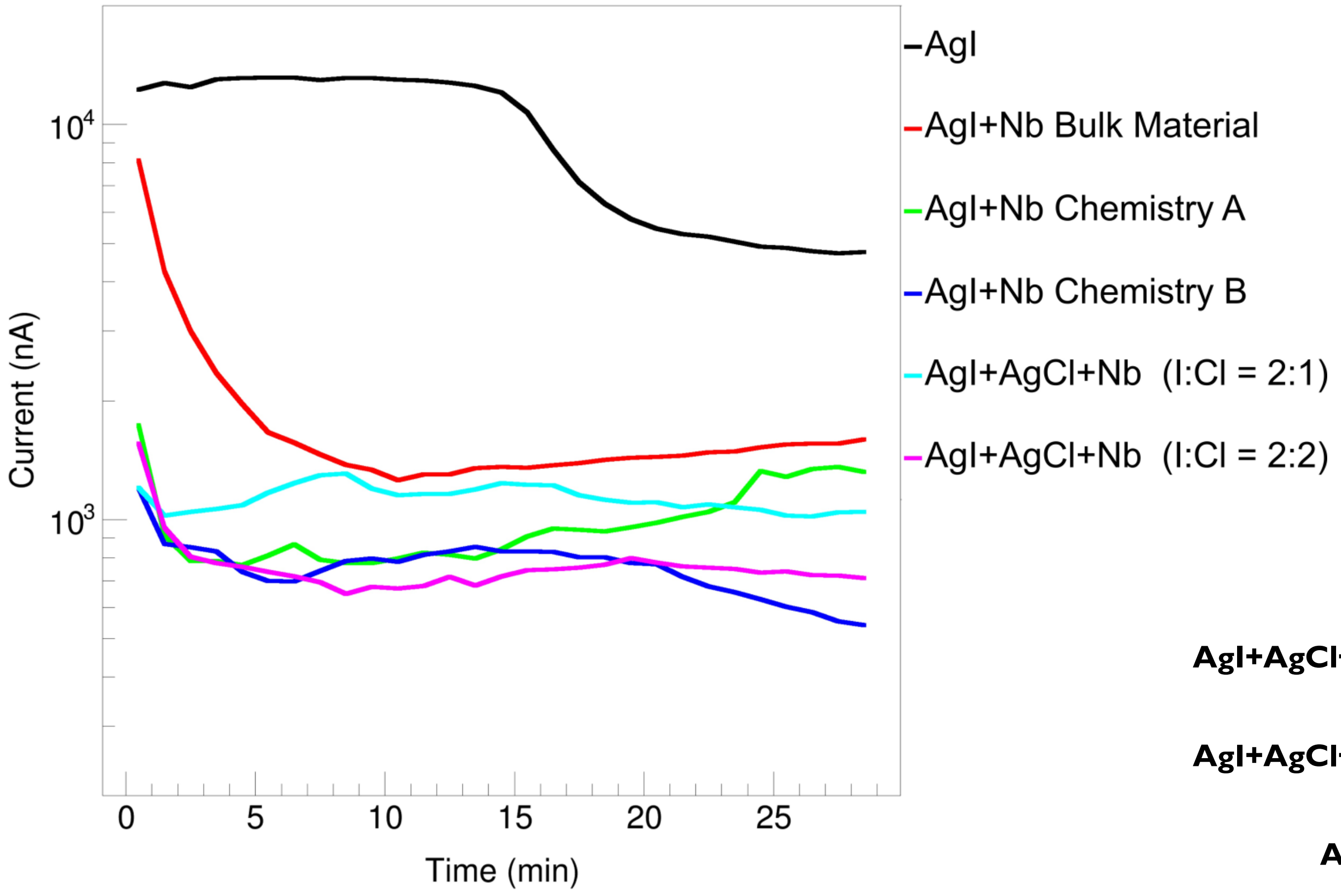
$^{129}I/^{127}I$ ratio
Set A: $^{129}I/^{127}I<2\times10^{-13}$
Set B: $^{129}I/^{127}I=2.2\times10^{-12}$
Set C: $^{129}I/^{127}I=9.95\times10^{-12}$
Set D: $^{129}I/^{127}I=1.14\times10^{-10}$
10^{-10}
10^{-11}
10^{-12}
10^{-13}
1
10
Iodine content in target holder (μg/mg)

Liu (2015)

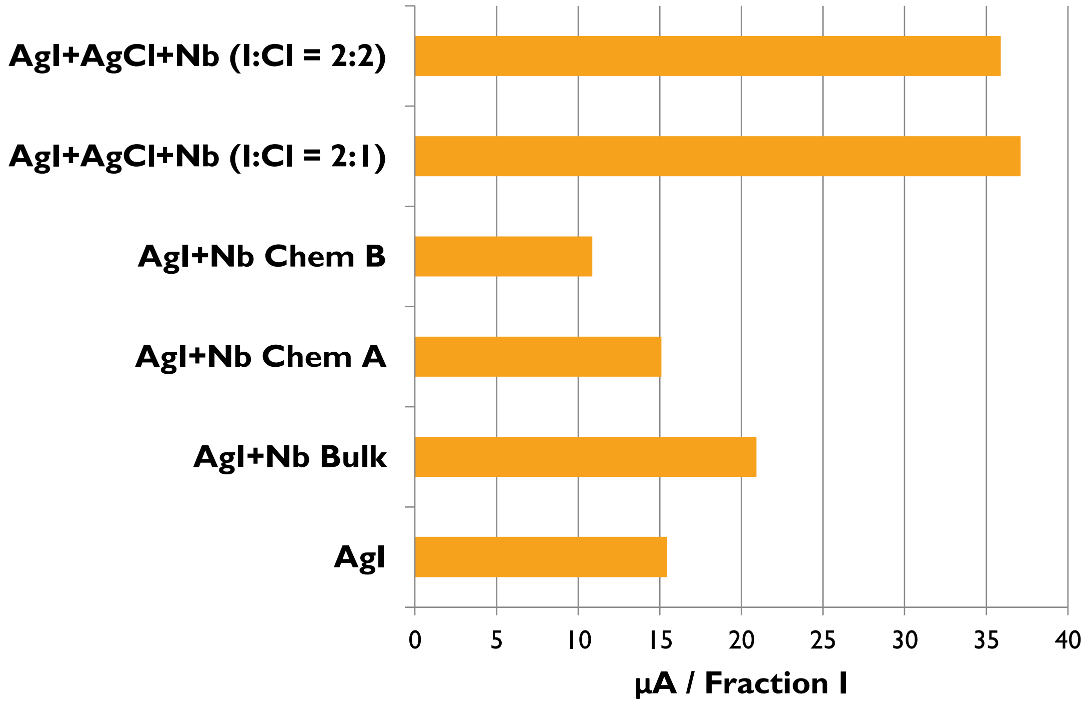


700
100
10
1
^{127}I Current/nA
0.0
0.2
0.4
0.6
0.8
1.0
^{127}I Carrier/mg
Sample-1
Sample-2
Blank
Blank(Nb power)

Xing (2017)



Current (nA)
10^4
10^3
0
5
10
15
20
25
Time (min)
AgI
AgI+Nb Bulk Material
AgI+Nb Chemistry A
AgI+Nb Chemistry B
AgI+AgCl+Nb (I:Cl = 2:1)
AgI+AgCl+Nb (I:Cl = 2:2)

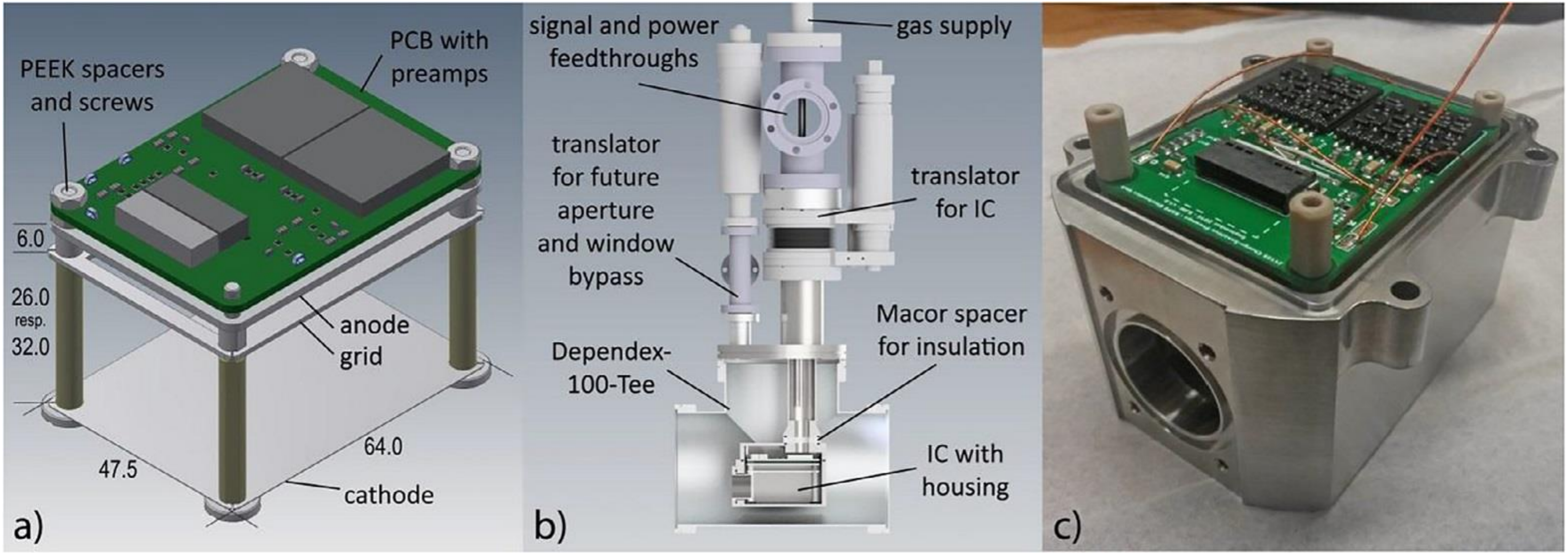


AgI+AgCl+Nb (I:Cl = 2:2)
AgI+AgCl+Nb (I:Cl = 2:1)
AgI+Nb Chem B
AgI+Nb Chem A
AgI+Nb Bulk
AgI
0
5
10
15
20
25
30
35
40
µA / Fraction I

PLANNED ^{129}I DEVELOPMENT

- Experimental
  - Coprecipitation with AgCl
  - Widening injection, analysis magnet slits

- Technical
  - Recirculating gas stripping
  - **Retractable ionization chamber**
  - FN regulation using HE offset cup

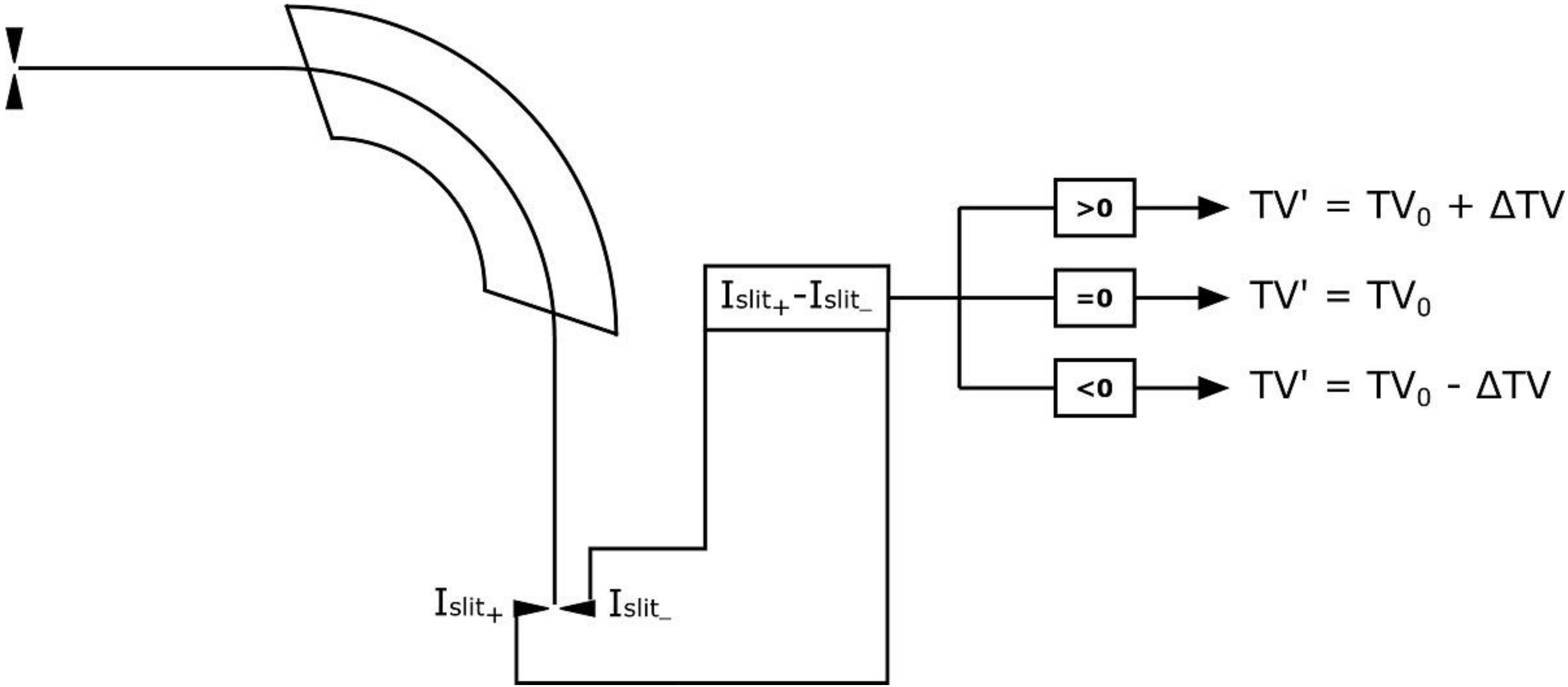


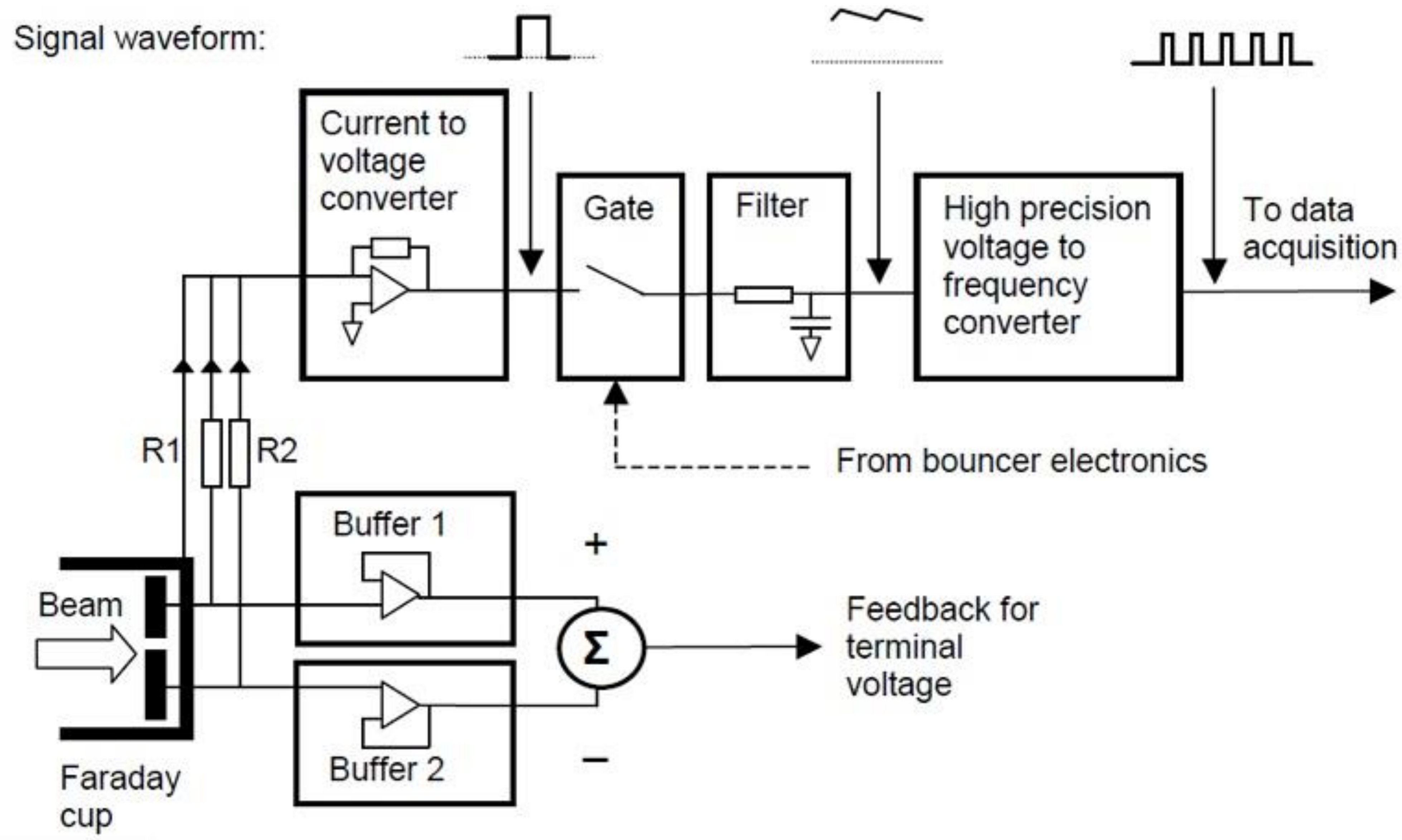
Martschini (2019)

PLANNED ^{129}I DEVELOPMENT

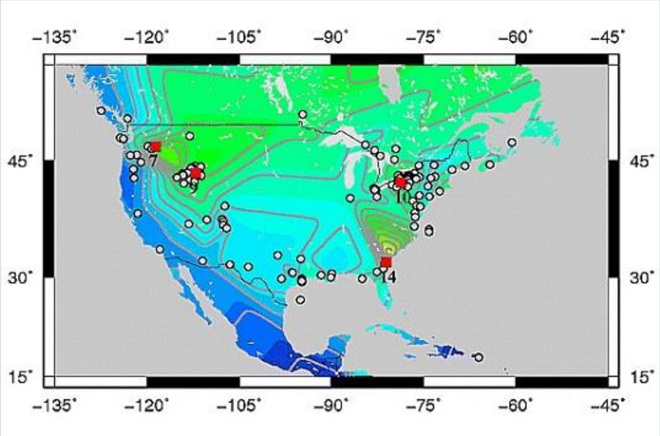
- Experimental
  - Coprecipitation with AgCl
  - Widening injection, analysis magnet slits

- Technical
  - Recirculating gas stripping
  - Retractable ionization chamber
  - **FN regulation using HE offset cup**

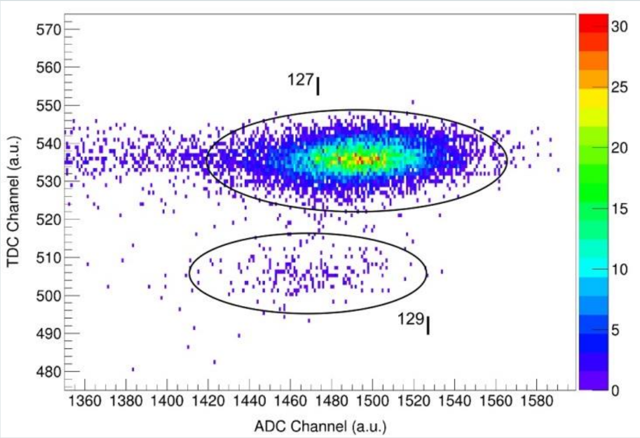

$I_{slit+} - I_{slit-}$
>0
=0
<0
$TV' = TV_0 + \Delta TV$
$TV' = TV_0$
$TV' = TV_0 - \Delta TV$
I_{slit+}
I_{slit-}


Signal waveform:
Current to voltage converter
Gate
Filter
High precision voltage to frequency converter
To data acquisition
R1
R2
From bouncer electronics
Buffer 1
+
Beam
Σ
Feedback for terminal voltage
Buffer 2
−
Faraday cup

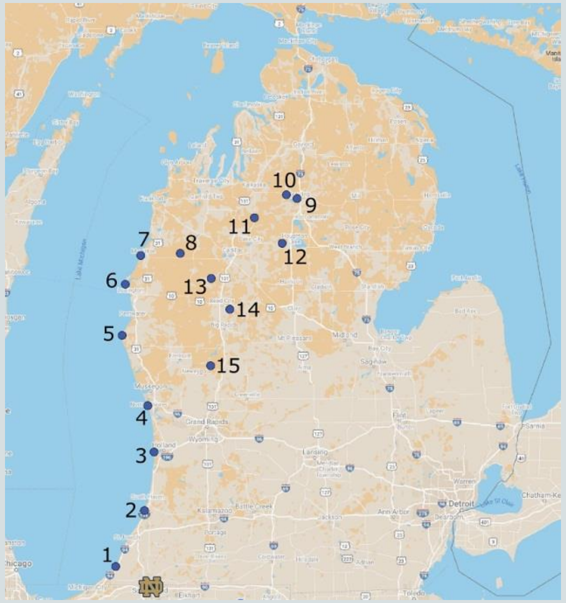
CONCLUSIONS



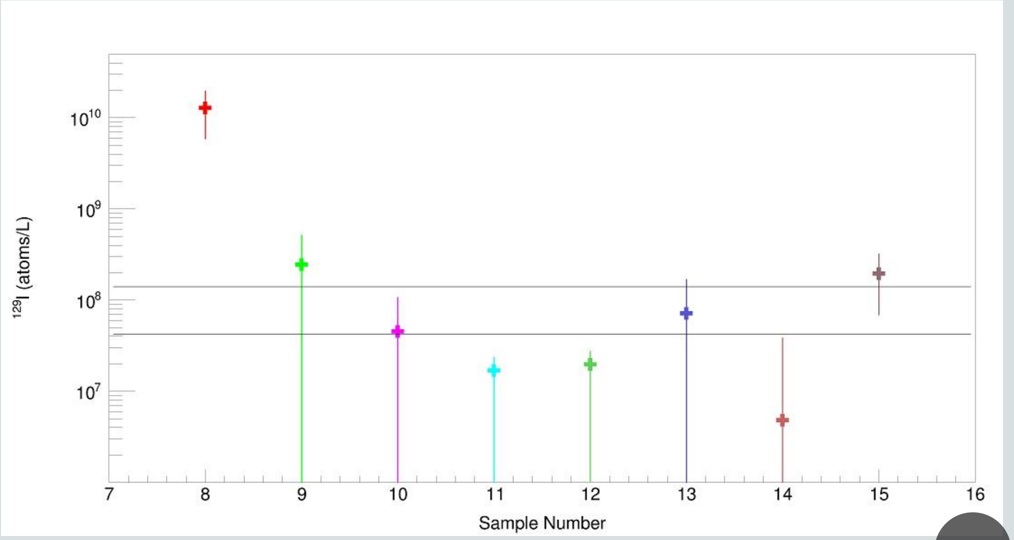
- ^{129}I is a useful environmental tracer because of its point-like sources and ability to move through the environment
  - Lack of measurements limit its potential



- The AMS group now has the capability of measuring ^{129}I and separate its contaminant, ^{127}I



- 15 samples were collected and processed to determine their ^{129}I content
  - 8 were measured because of time constraints, low statistics



- Several different methods for increasing the ^{129}I signal have been planned for future work prior to further measurements and a subsequent publication

ACKNOWLEDGMENTS

AMS Group

Tyler Anderson

Lauren Callahan

Adam Clark

Austin Nelson

Philippe Collon

NSL

Dan Robertson

Ed Stech

Brad Mulder

Haley Borsodi

Brian Huge

Experimental Shifters

Drew Blankstein

Dan Burdette

Samuel Henderson

Craig Reingold

Sabrina Strauss

PRIME Lab

Greg Chmiel

Tom Woodruff

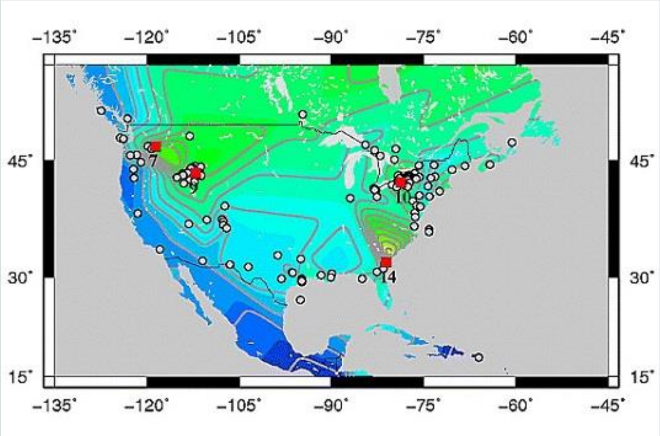
Marc Caffee

Others

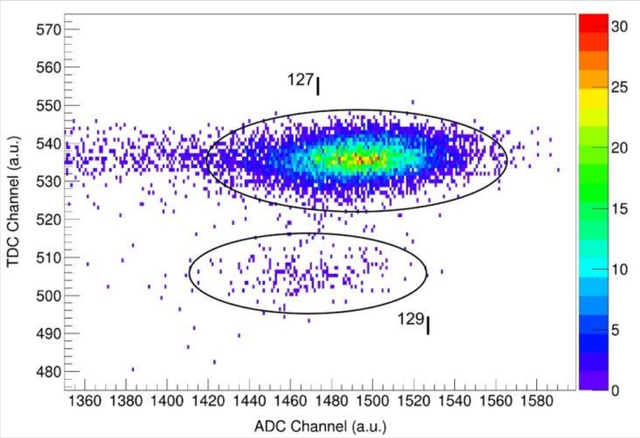
Carol Skulski

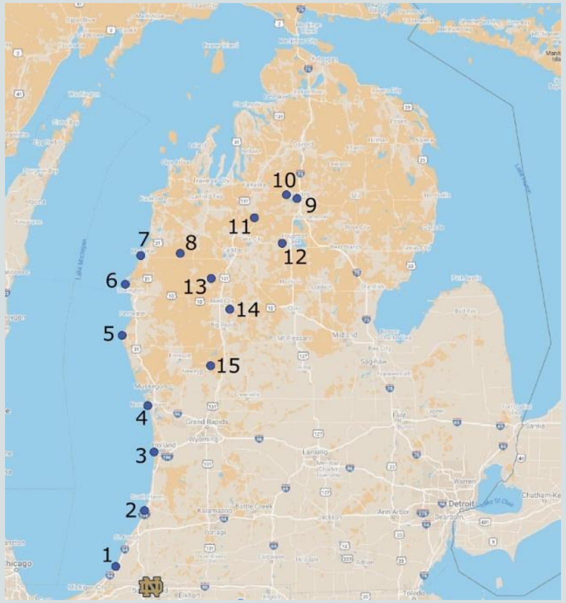
Mike Skulski

CONCLUSIONS



- ^{129}I is a useful environmental tracer because of its point-like sources and ability to move through the environment
  - Lack of measurements limit its potential
- The AMS group now has the capability of measuring ^{129}I and separate its contaminant, ^{127}I





- 15 samples were collected and processed to determine their ^{129}I content
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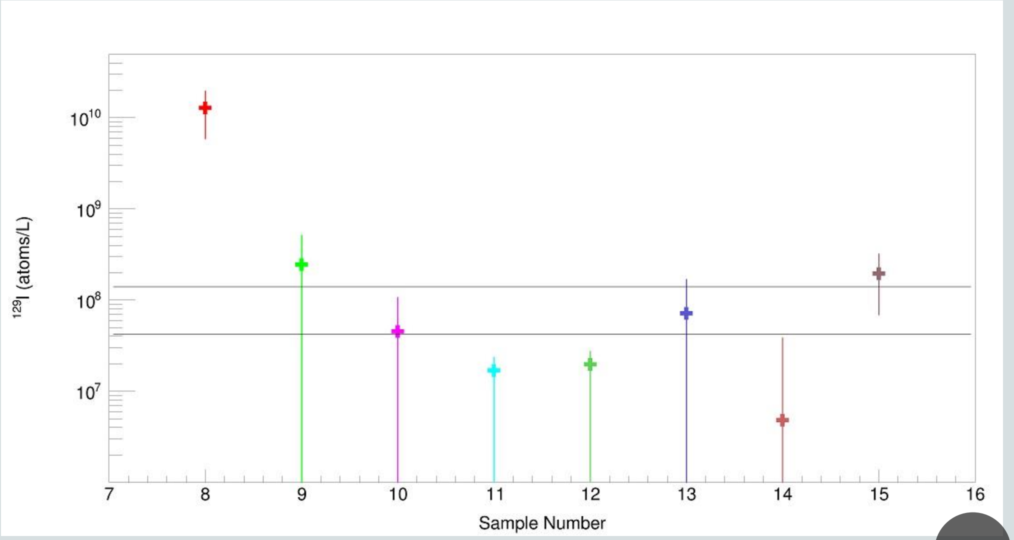
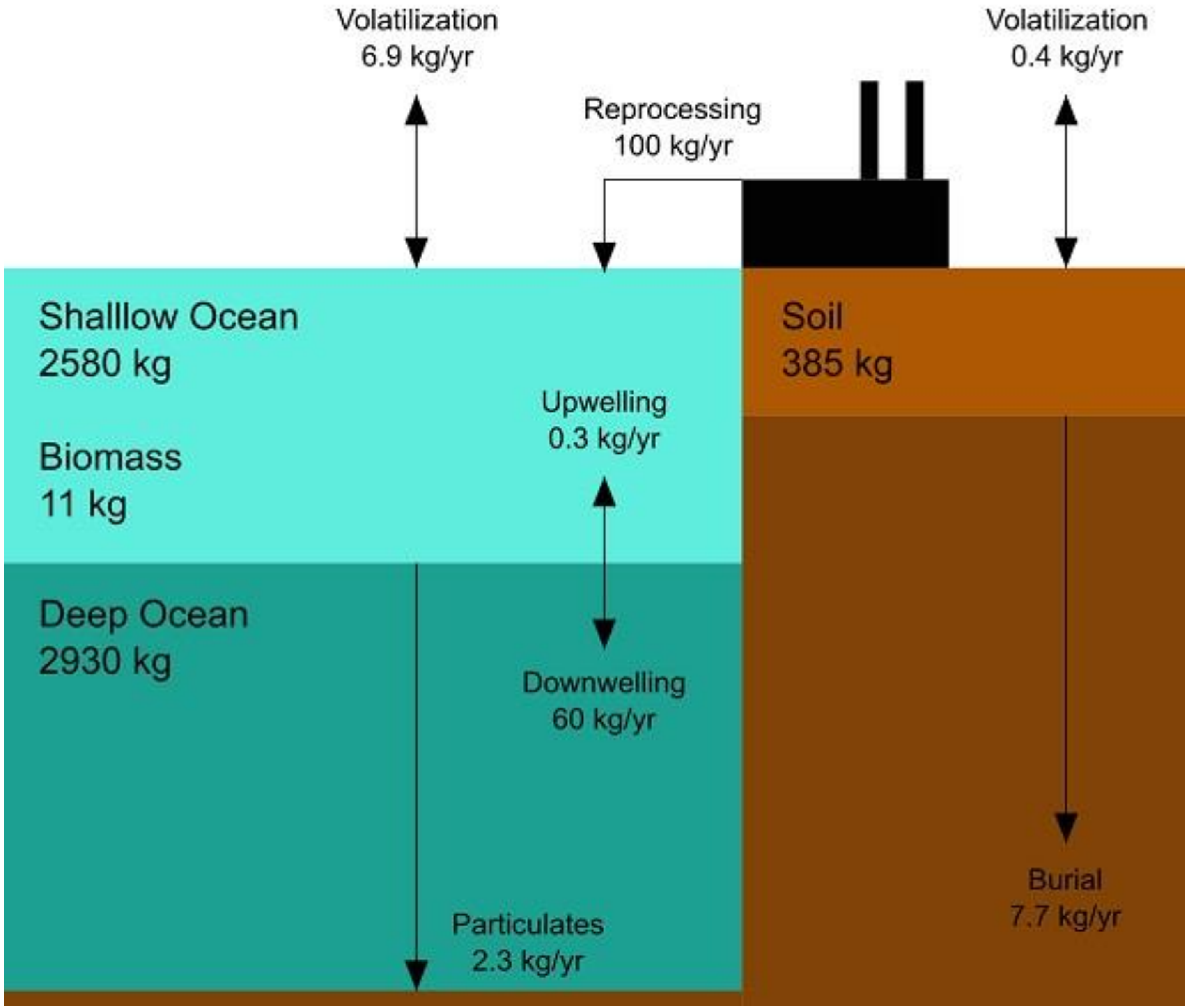


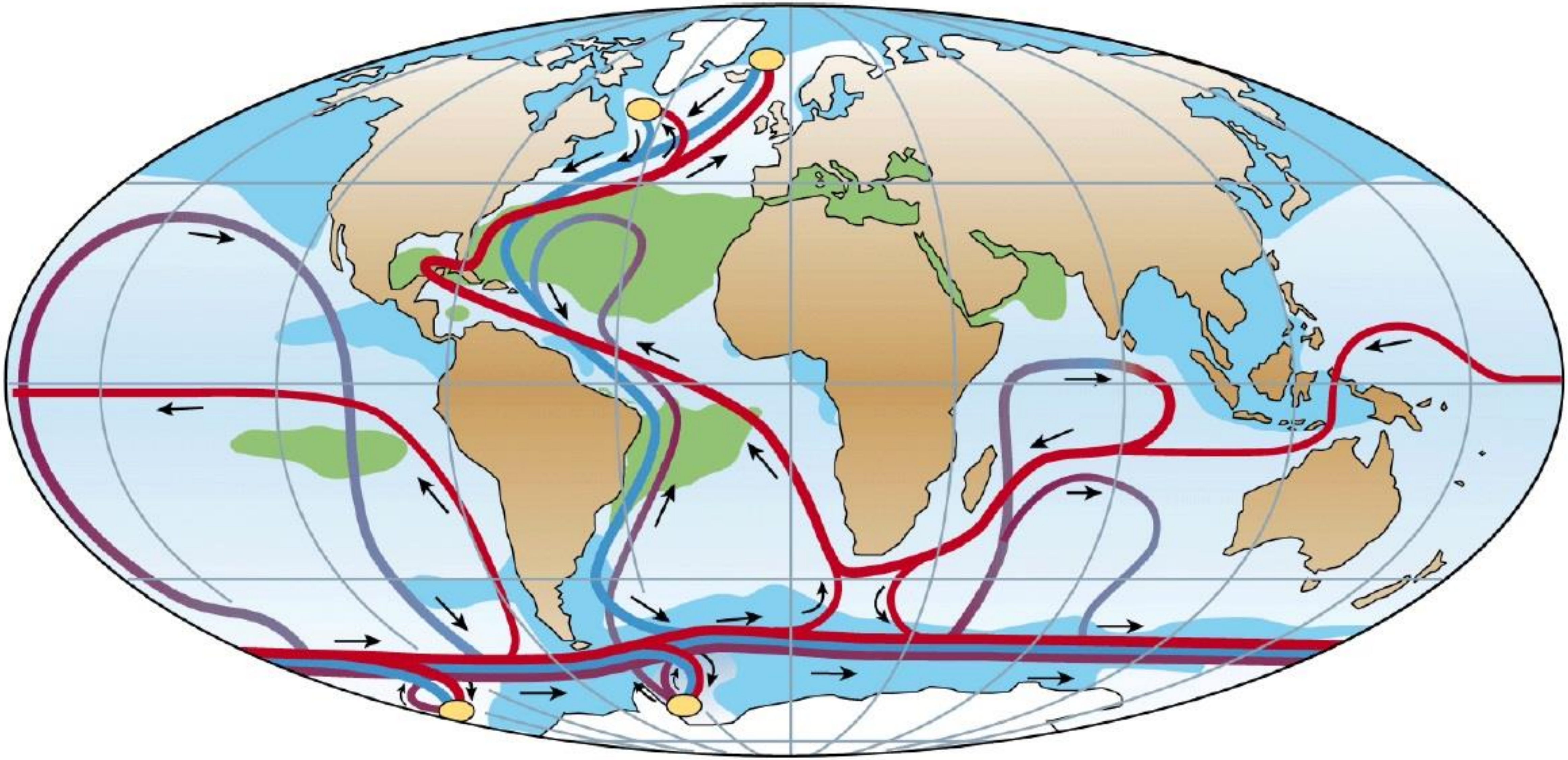
BACKUP SLIDES

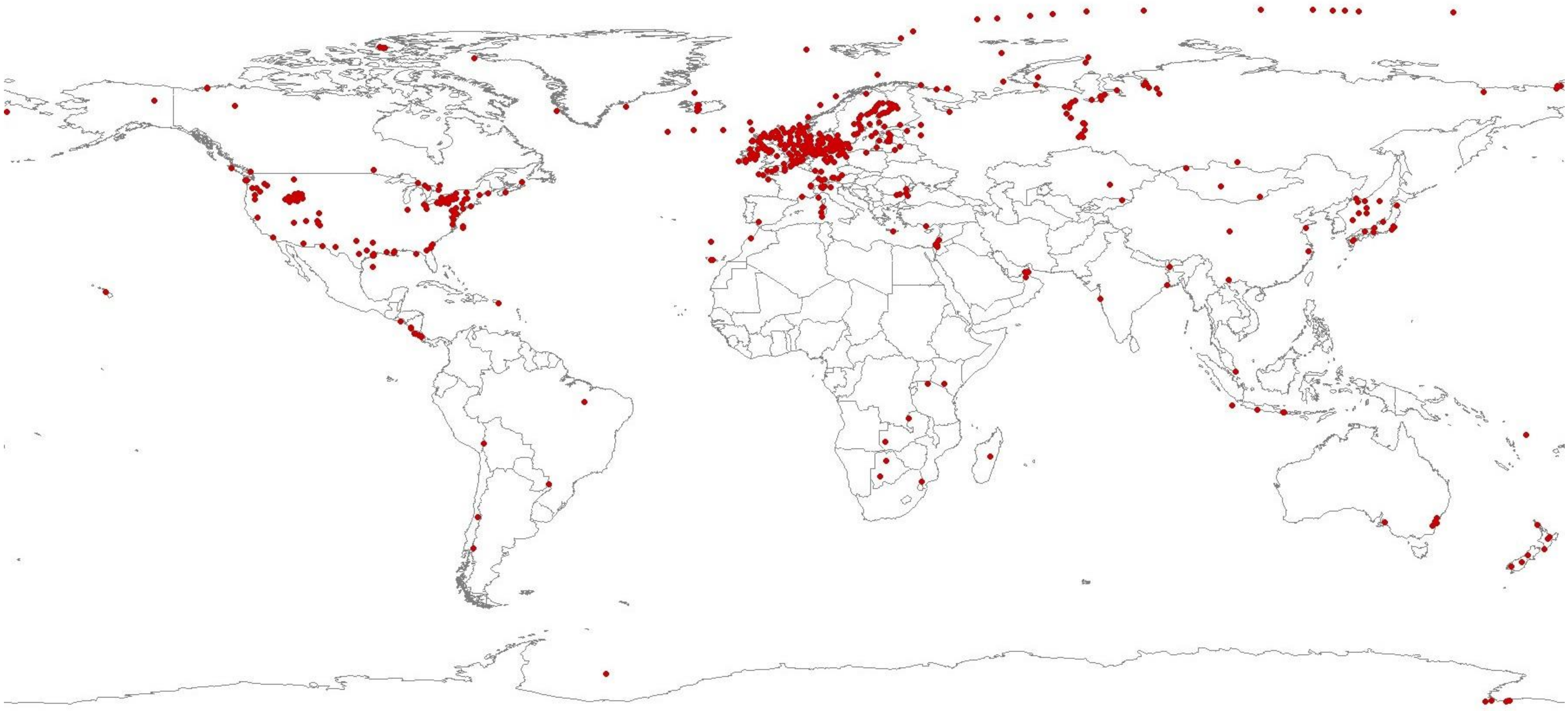
SINKS OF ^{129}I

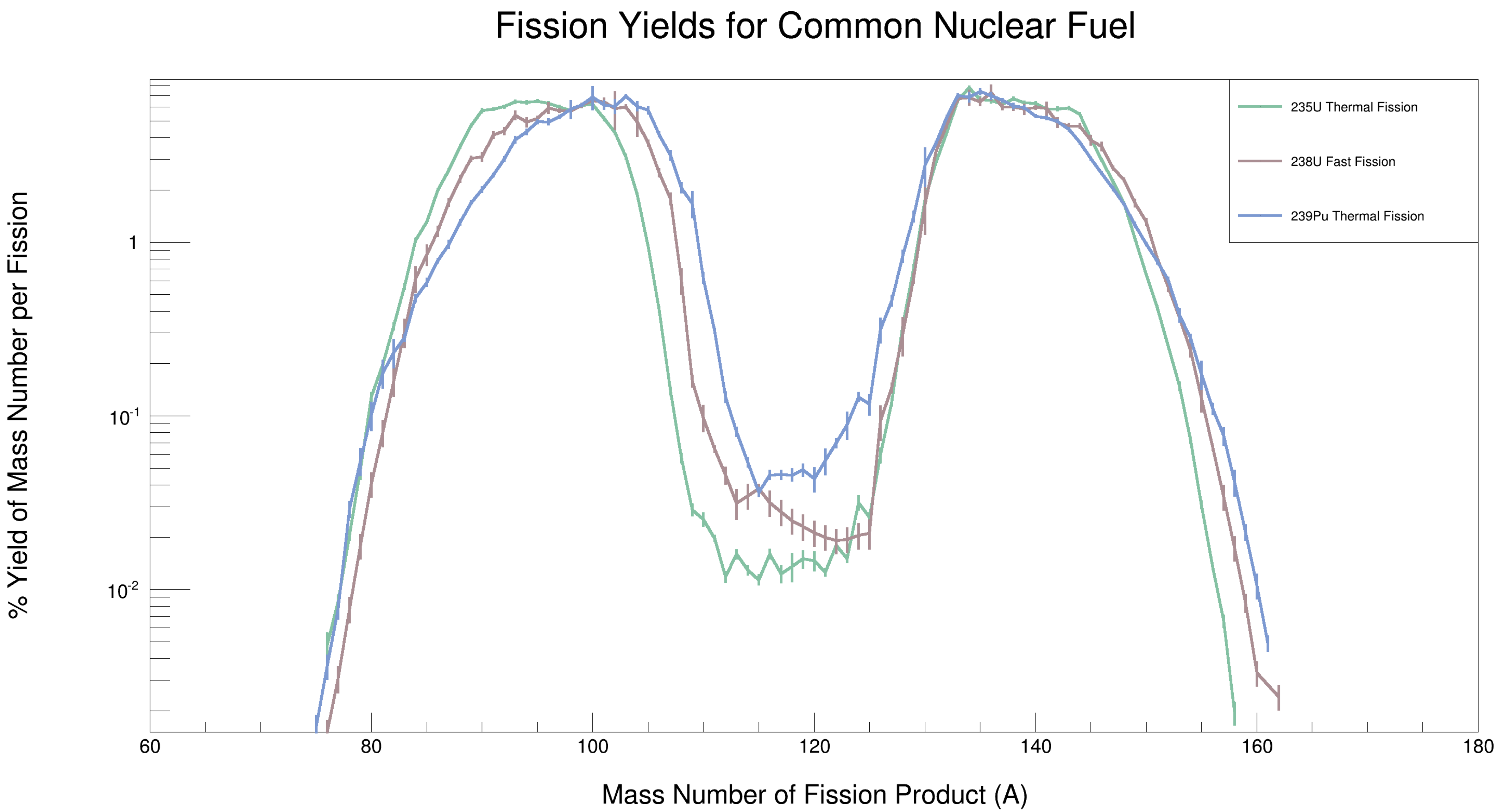
- Calculating the ^{129}I content of the shallow oceans (up to 100 m depth) from global ^{129}I content distribution accounts for only half of ^{129}I content

- Significant sinks
  - Deep ocean
  - Burial in soil

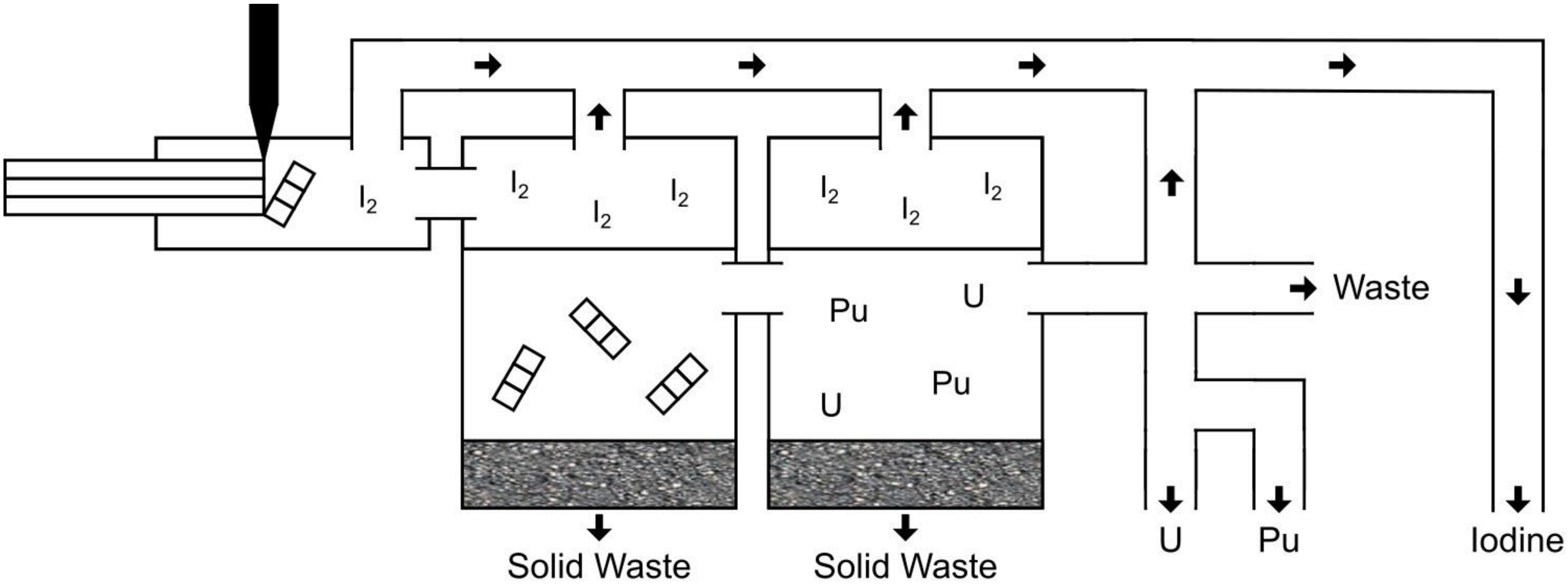


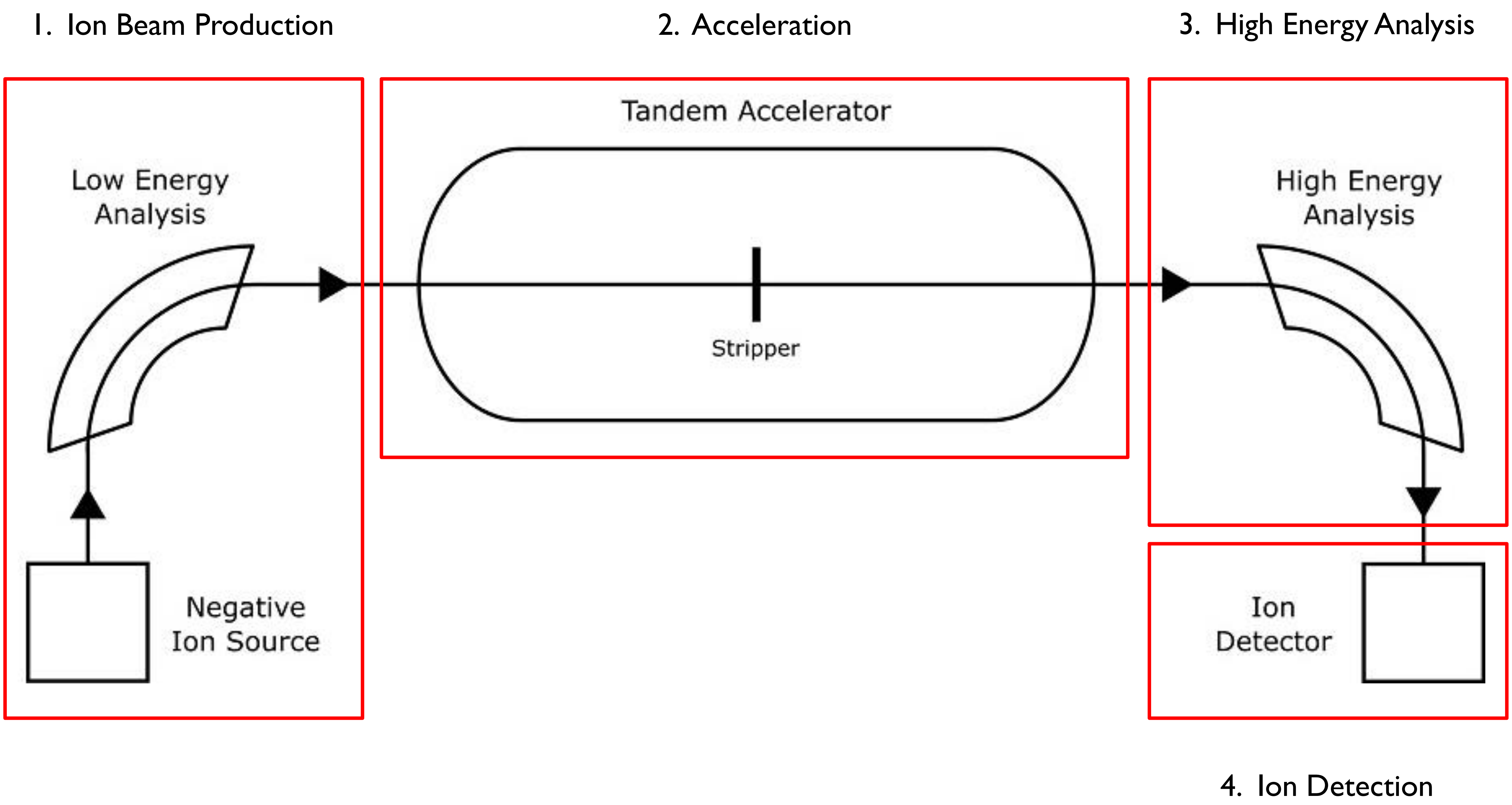


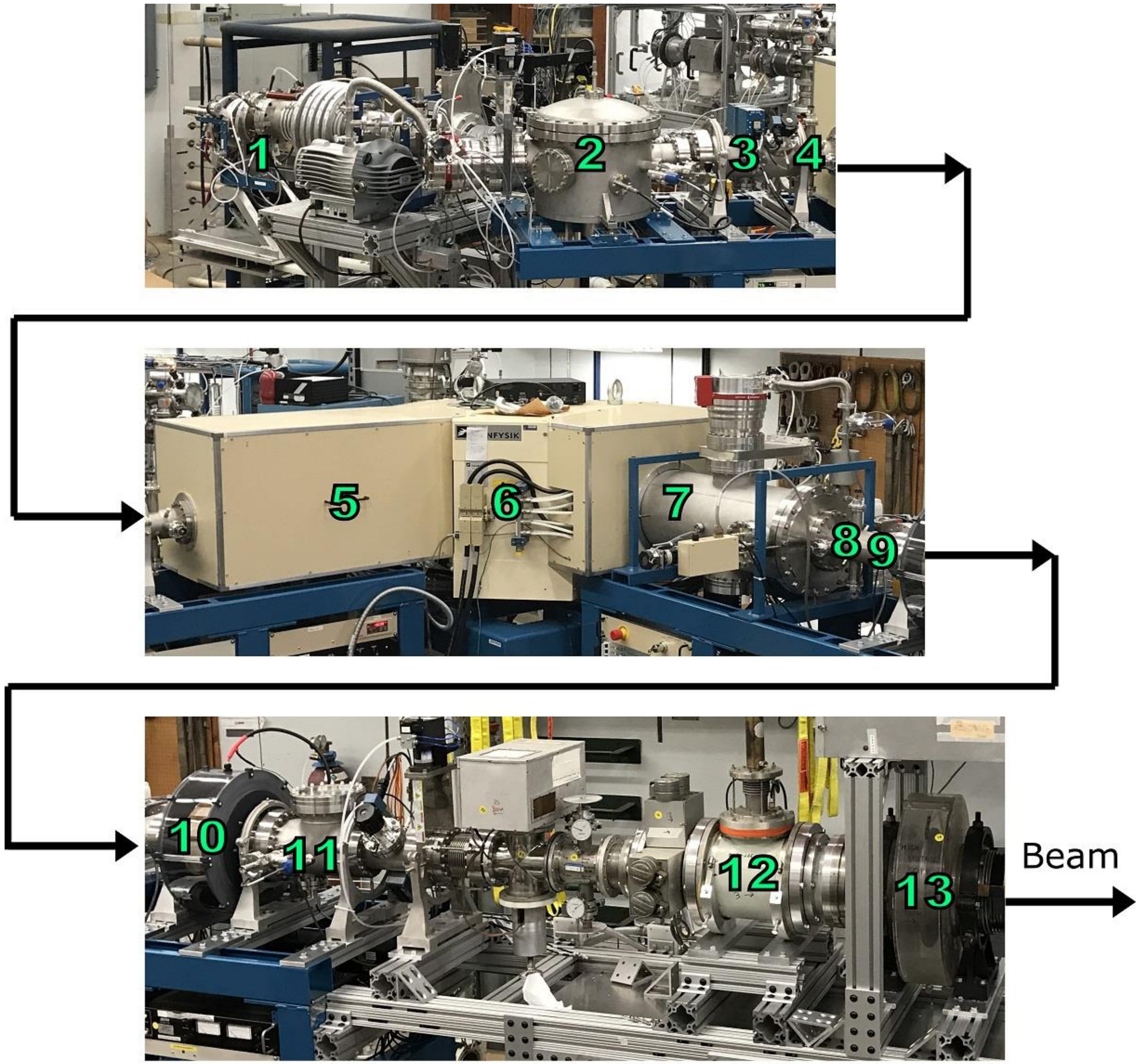



Fission Yields for Common Nuclear Fuel
% Yield of Mass Number per Fission
1
10^{-1}
10^{-2}
60
80
100
120
140
160
180
Mass Number of Fission Product (A)
235U Thermal Fission
238U Fast Fission
239Pu Thermal Fission

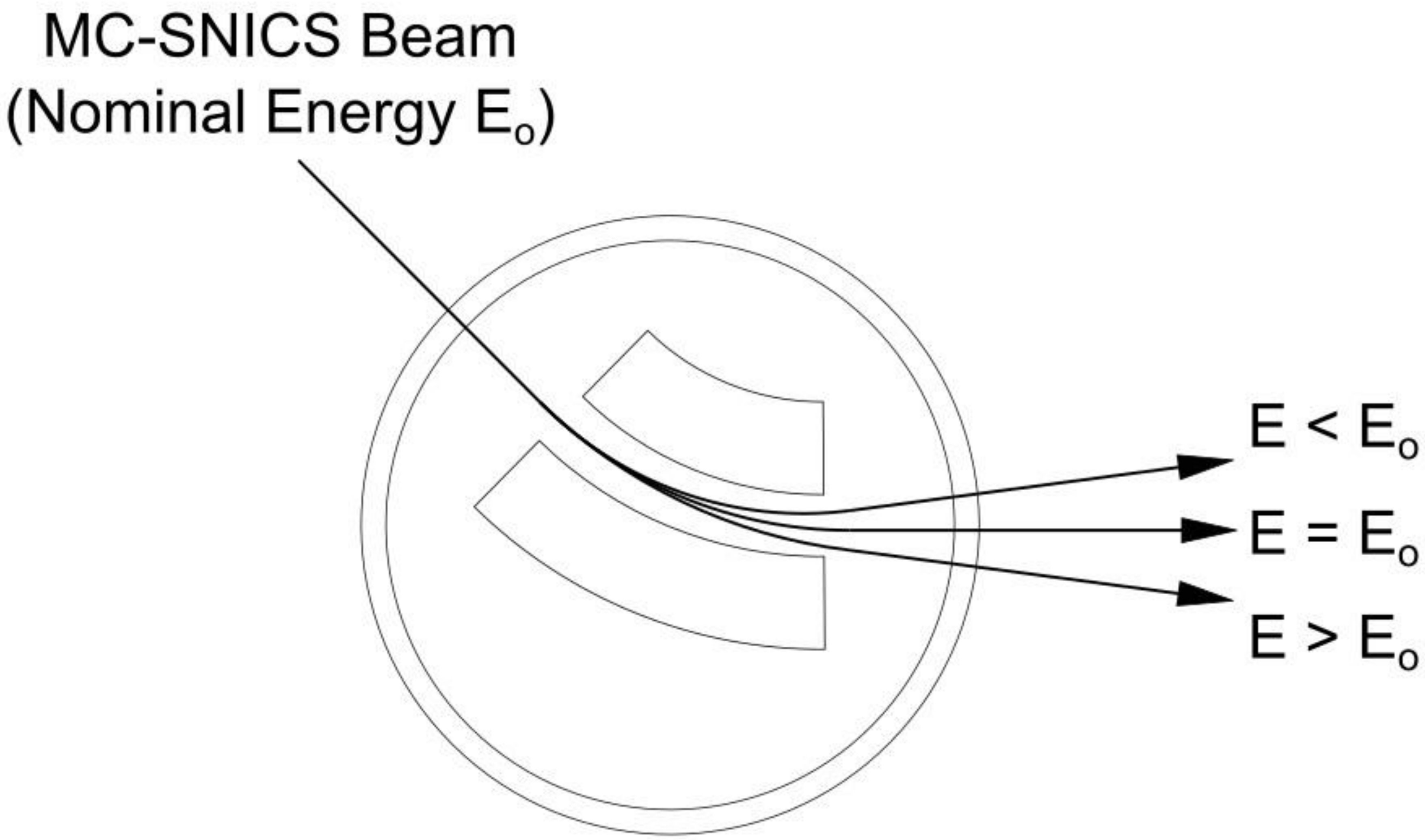
PUREX PROCESS

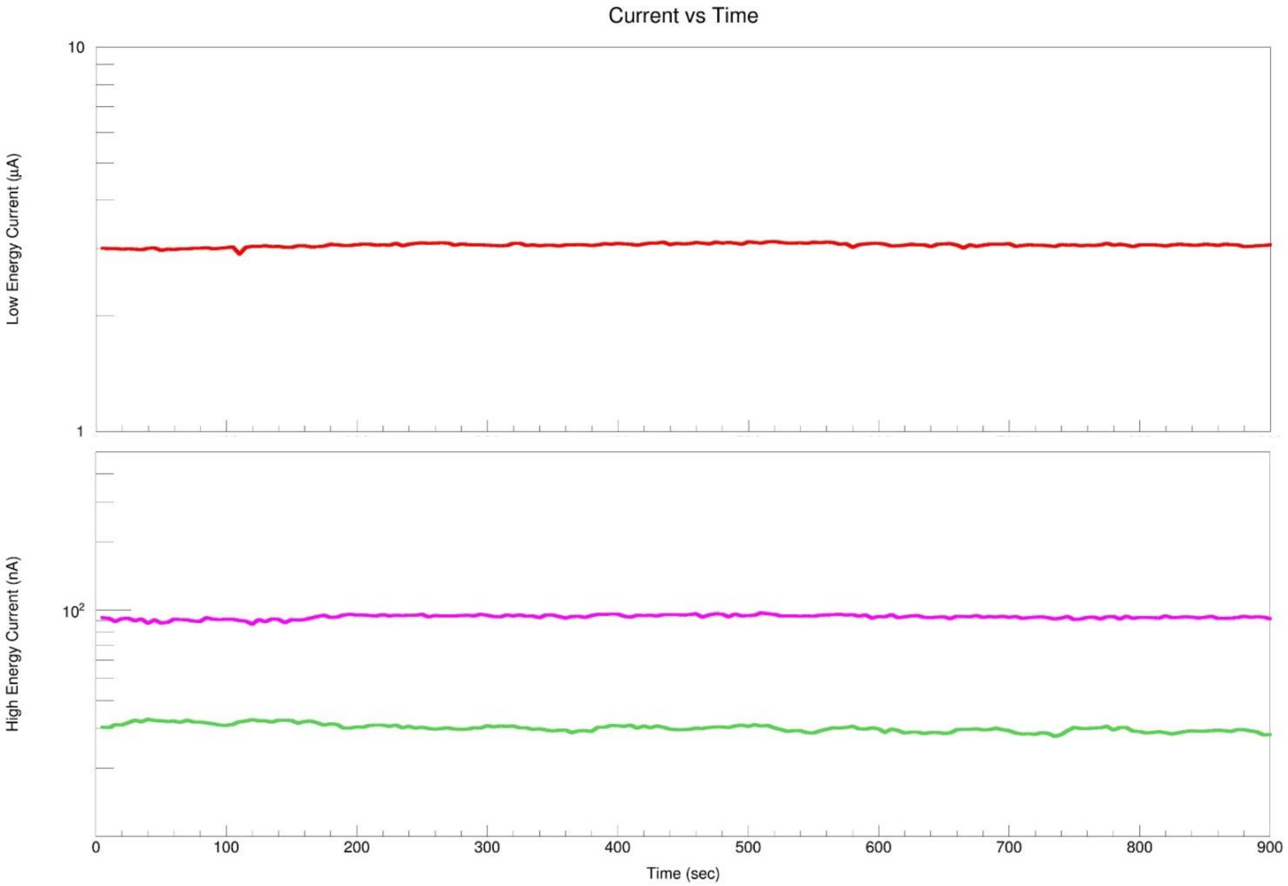



1. Ion Beam Production
2. Acceleration
3. High Energy Analysis
Tandem Accelerator
Low Energy Analysis
Stripper
High Energy Analysis
Negative Ion Source
Ion Detector
4. Ion Detection

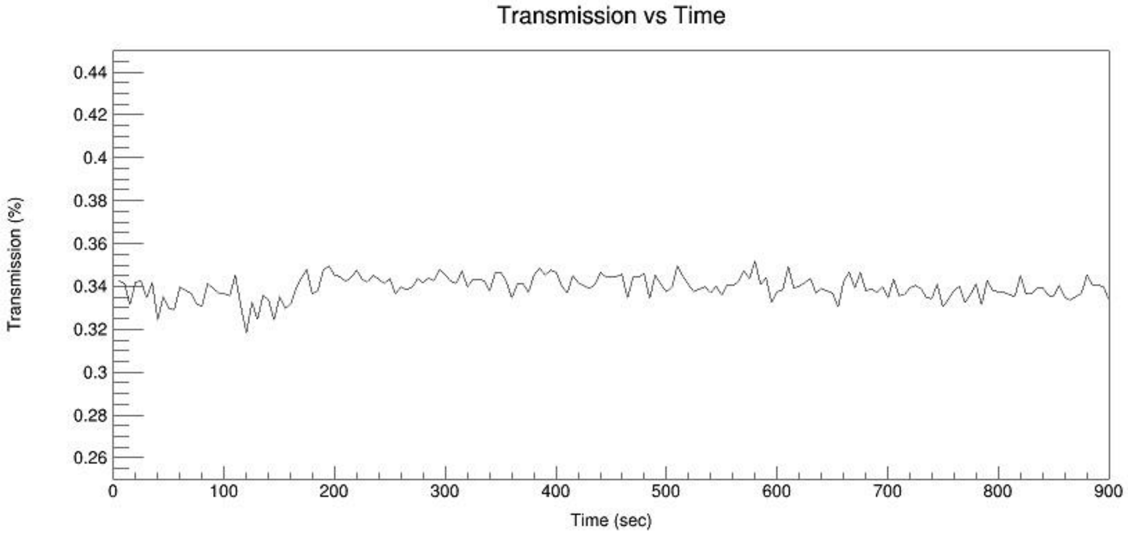


1. MC-SNICS
2. ESA
3. Vertical Steerer
4. Object Slits
5. MBS Cage
6. Injection Magnet
7. Offset Faraday Cups
8. Image Slits
9. Faraday Cup
10. Einzel Lens 1
11. Horizontal/Vertical Steerer
12. LE Faraday Cup
13. Einzel Lens 2

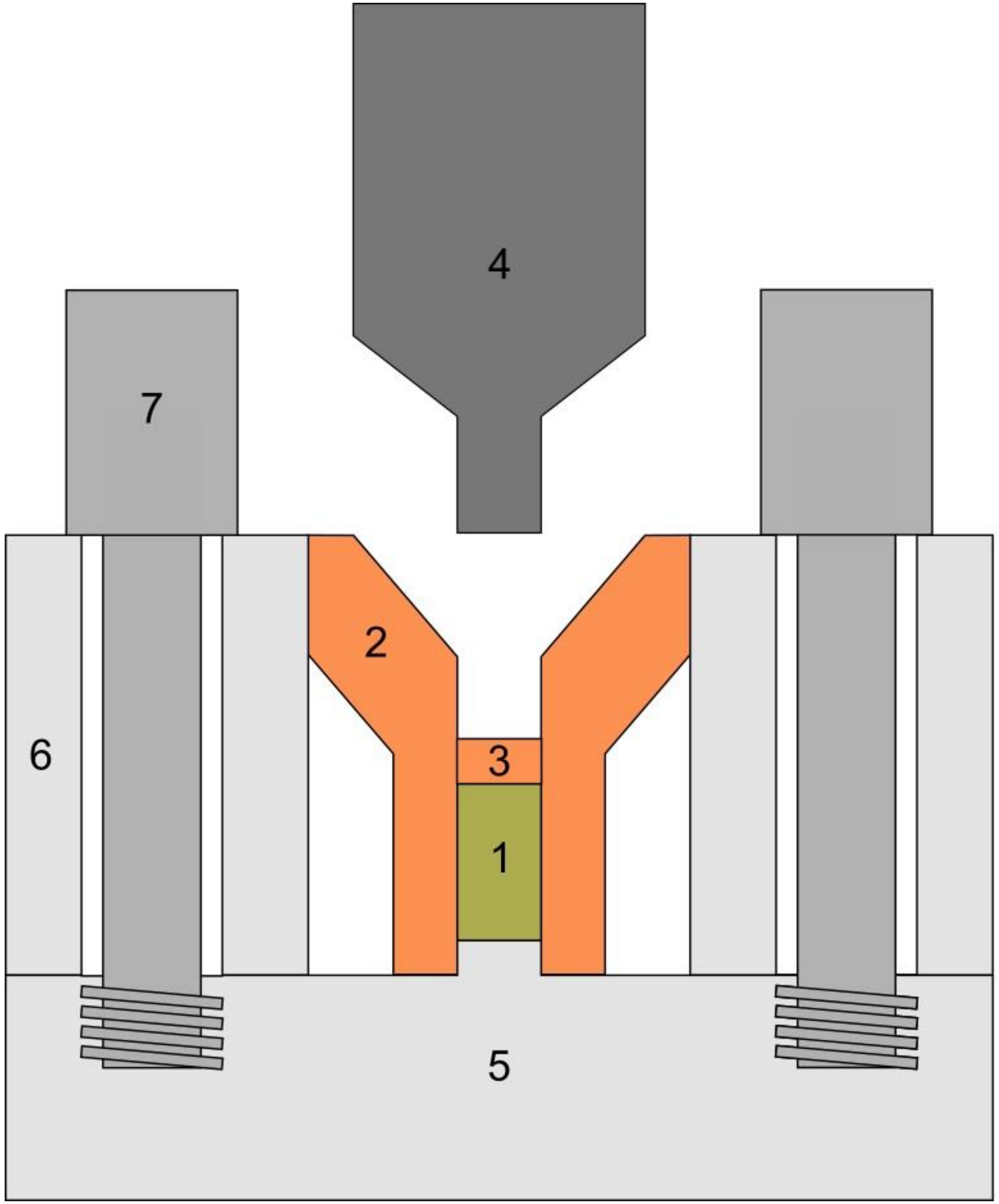

MC-SNICS Beam
(Nominal Energy E_o)
$E < E_o$
$E = E_o$
$E > E_o$



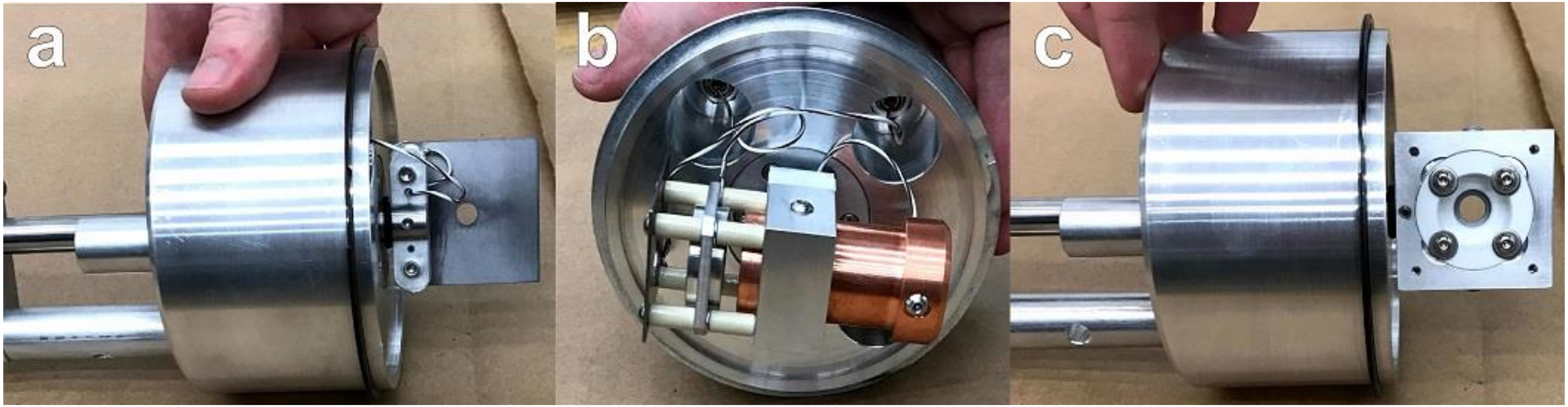
Current vs Time
10
1
Low Energy Current (μA)
10²
High Energy Current (nA)
0
100
200
300
400
500
600
700
800
900
Time (sec)



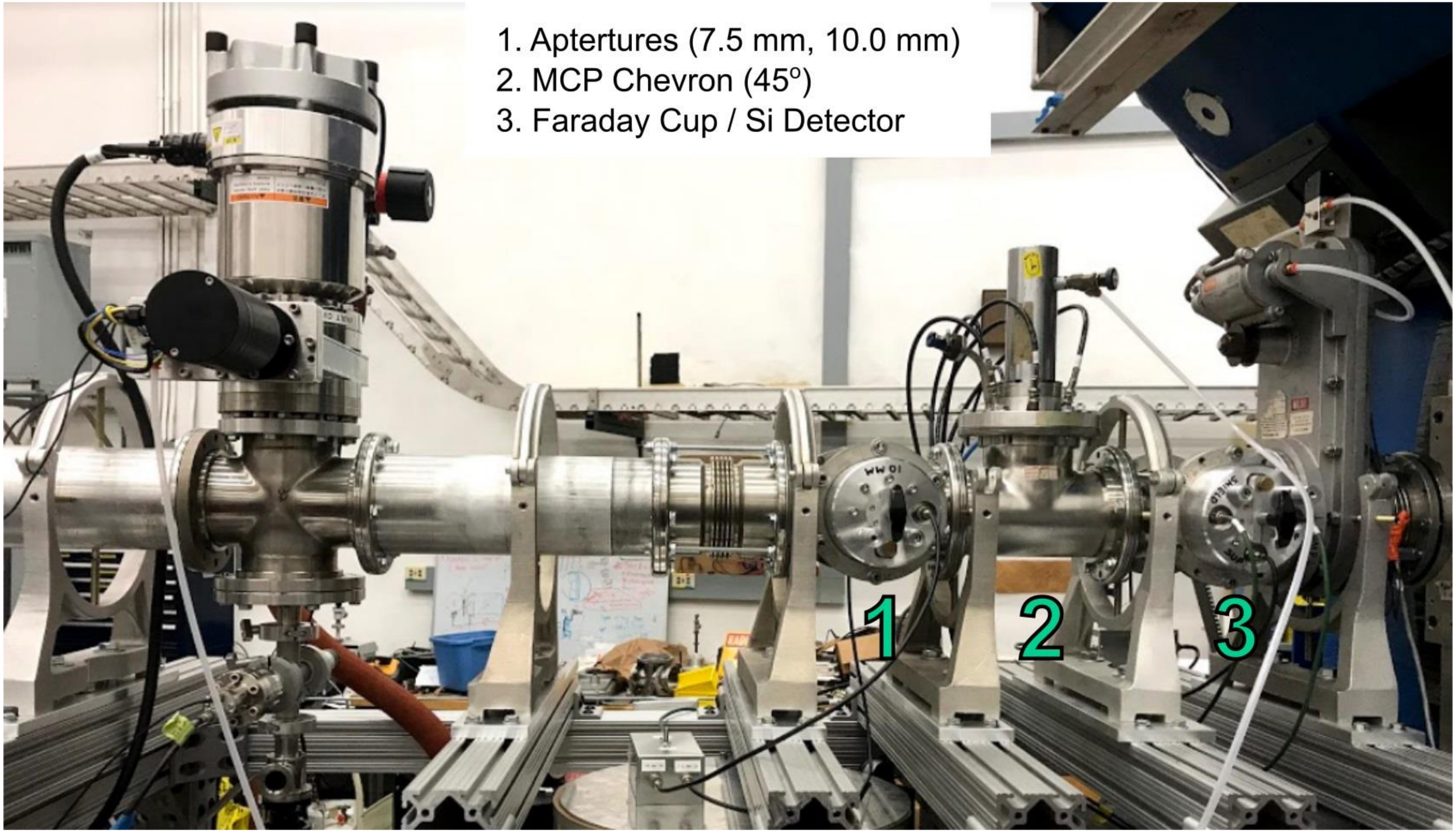
Transmission vs Time
0.44
0.42
0.4
0.38
0.36
0.34
0.32
0.3
0.28
0.26
Transmission (%)
0
100
200
300
400
500
600
700
800
900
Time (sec)

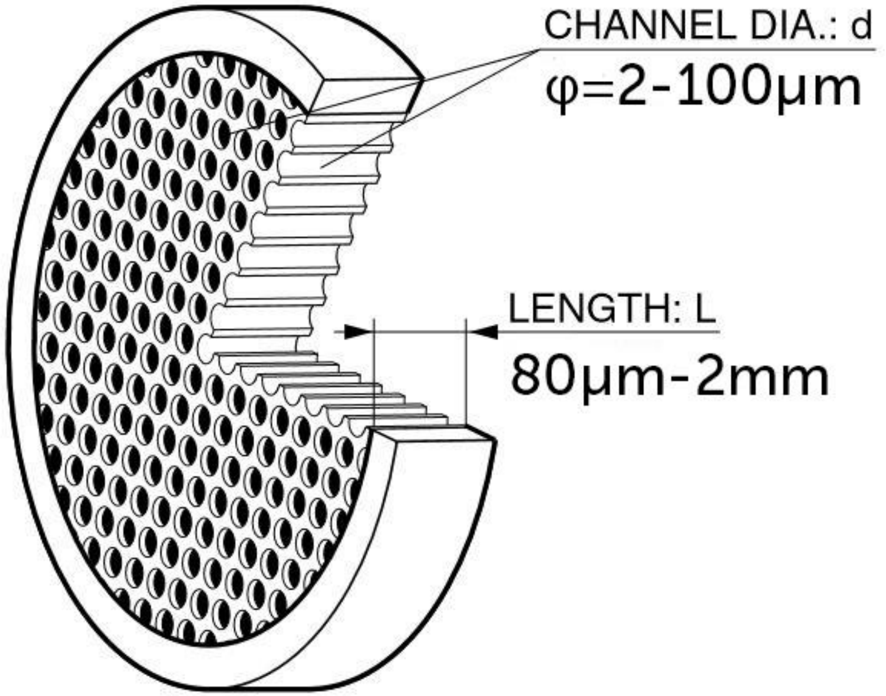


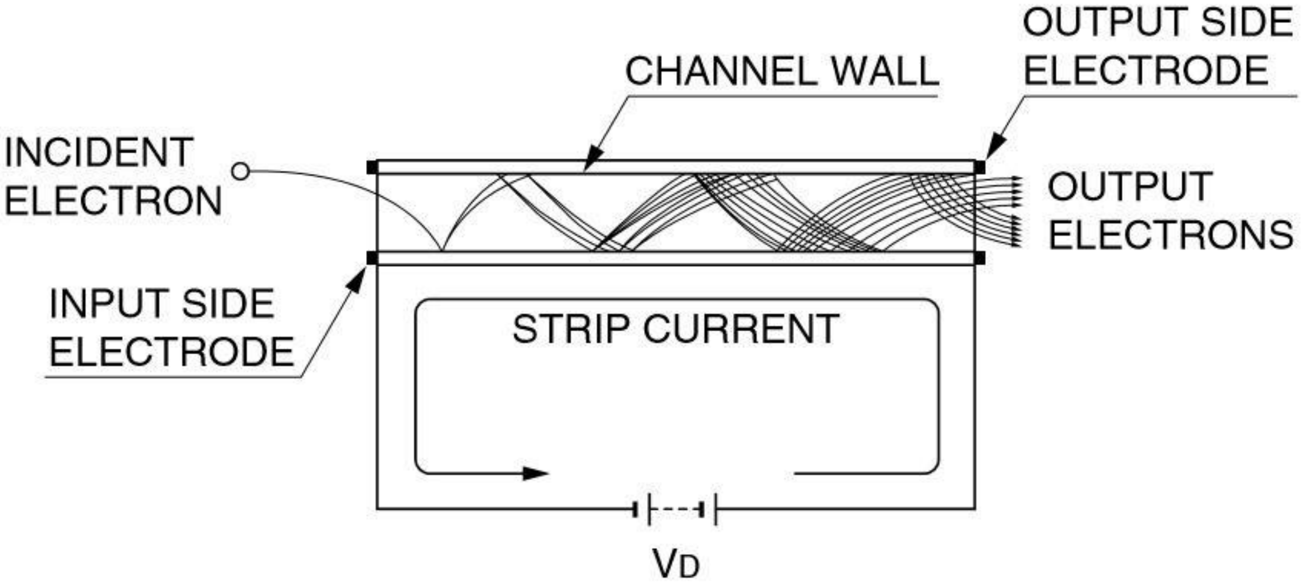
1. AgI + Nb
2. Cu Cathode
3. Cu Cathode Plug
4. Cathode Packing Rod
5. Cathode Packing Base
6. Cathode Retaining Disc
7. Thumb Screws

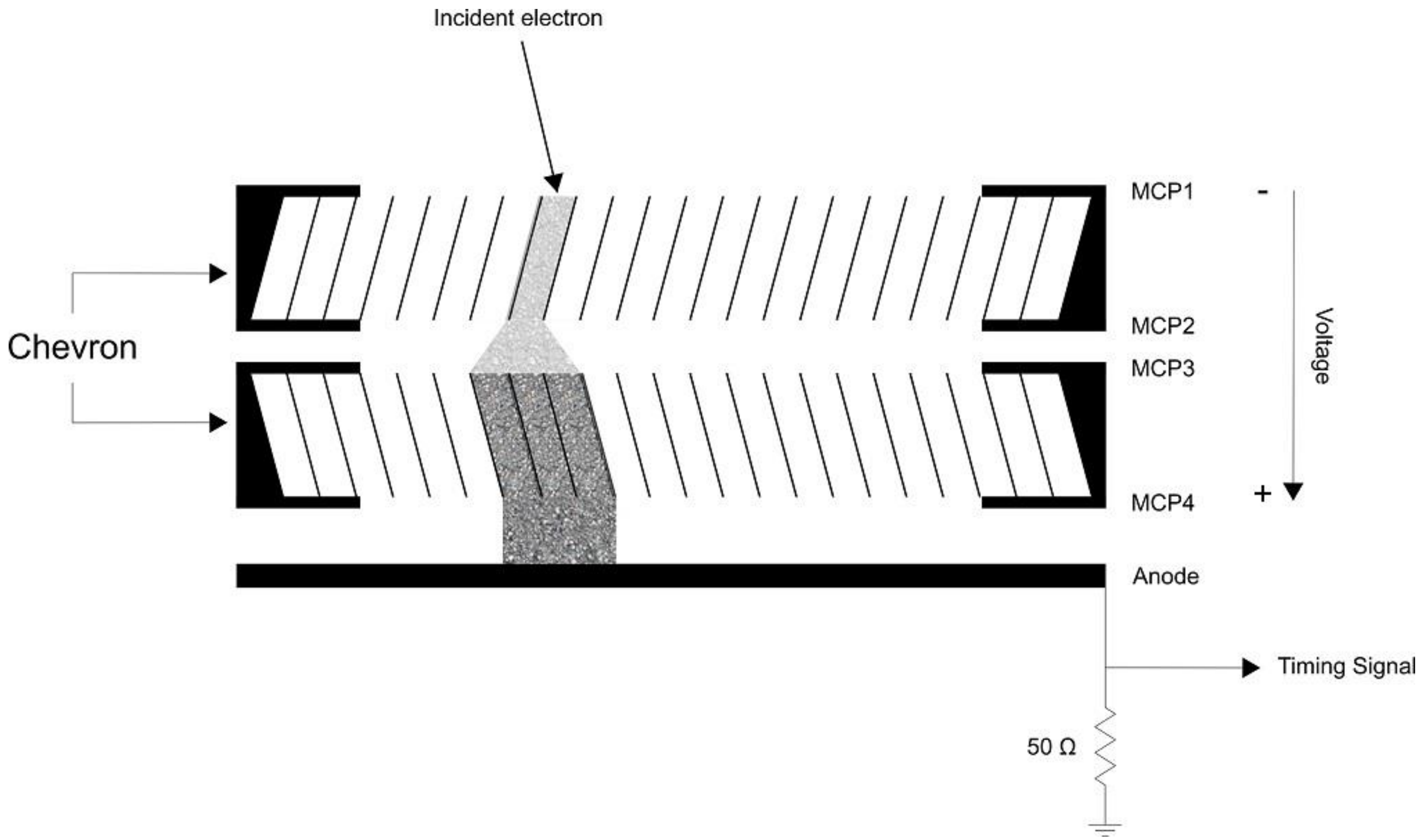

a
b
c

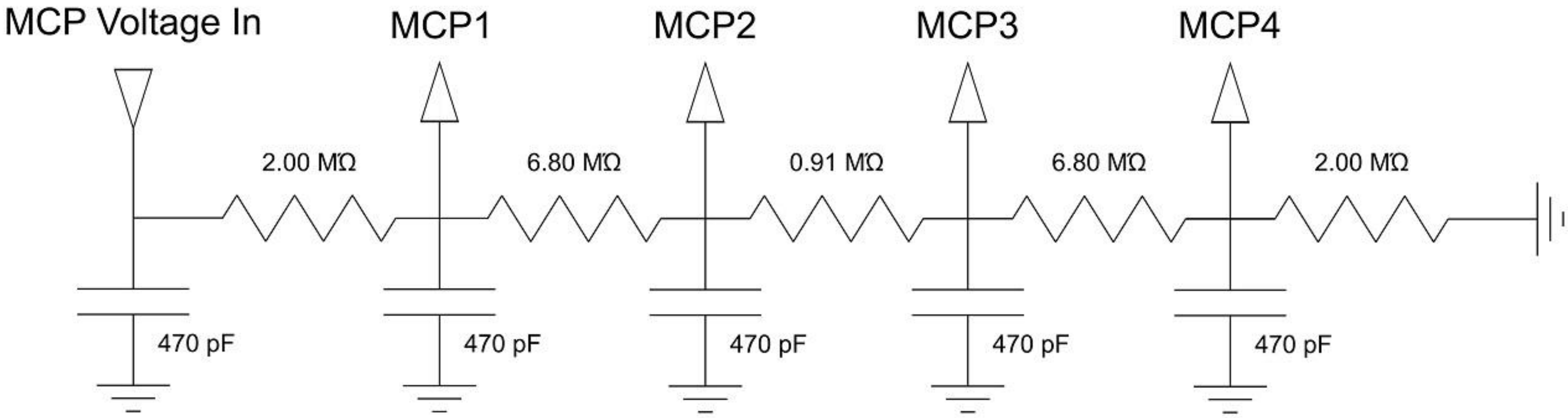
1. Aptertures (7.5 mm, 10.0 mm)
2. MCP Chevron (45°)
3. Faraday Cup / Si Detector



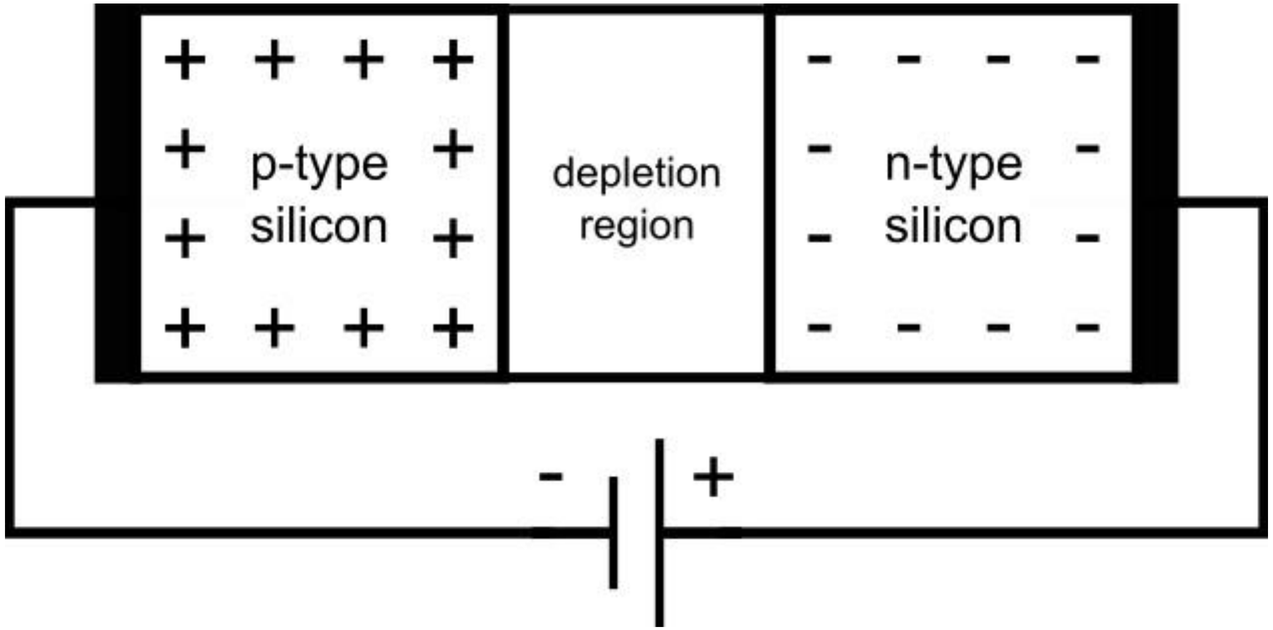

CHANNEL DIA.: d
φ=2-100μm
LENGTH: L
80μm-2mm

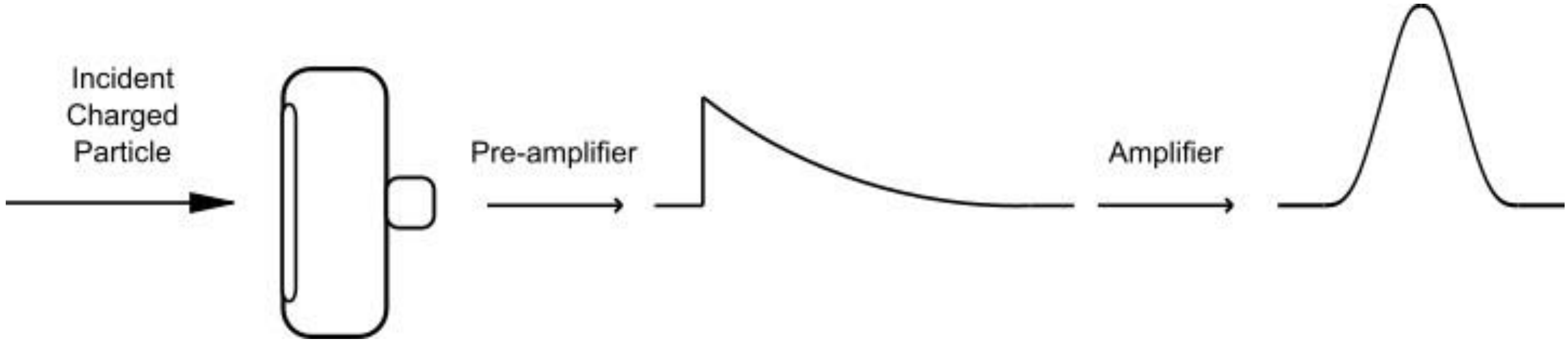

CHANNEL WALL
OUTPUT SIDE ELECTRODE
INCIDENT ELECTRON
OUTPUT ELECTRONS
INPUT SIDE ELECTRODE
STRIP CURRENT
VD

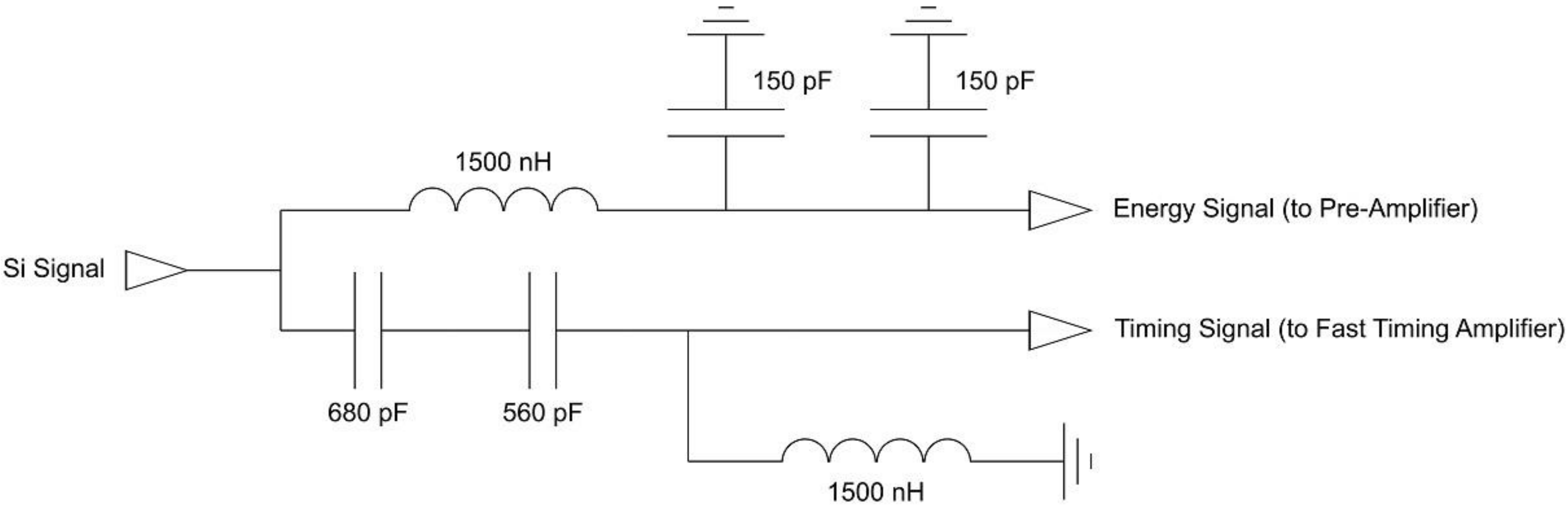

Incident electron
Chevron
MCP1
MCP2
MCP3
MCP4
-
+
Voltage
Anode
Timing Signal
50 Ω


MCP Voltage In
MCP1
MCP2
MCP3
MCP4
2.00 MΩ
6.80 MΩ
0.91 MΩ
6.80 MΩ
2.00 MΩ
470 pF
470 pF
470 pF
470 pF
470 pF

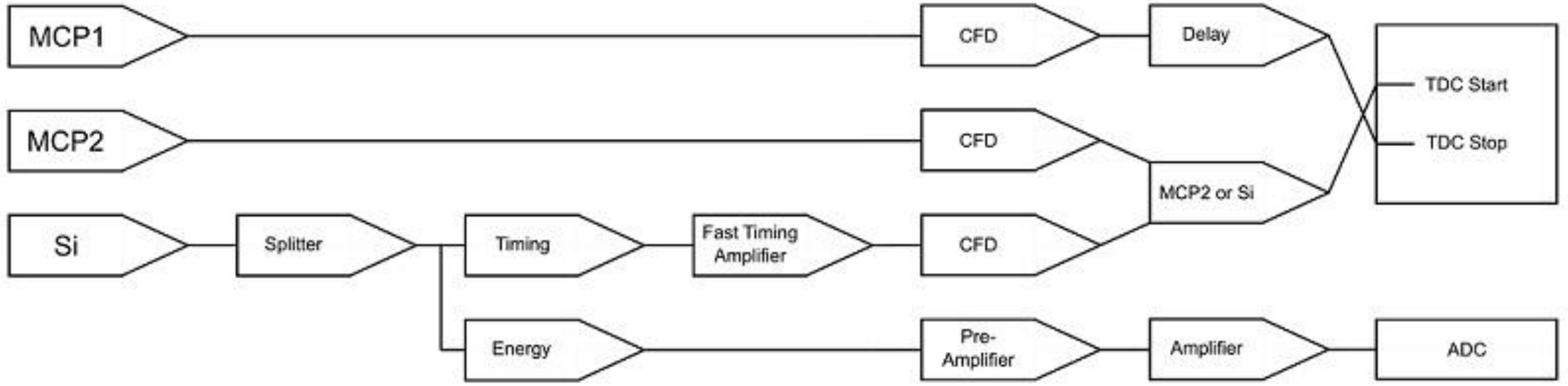

Tek PreVu
Noise Filter Off
1
1 100mV
2.00ns -2.68000ns
1 -296mV
< 10 Hz
13:40:55


p-type
silicon
depletion
region
n-type
silicon
-
+

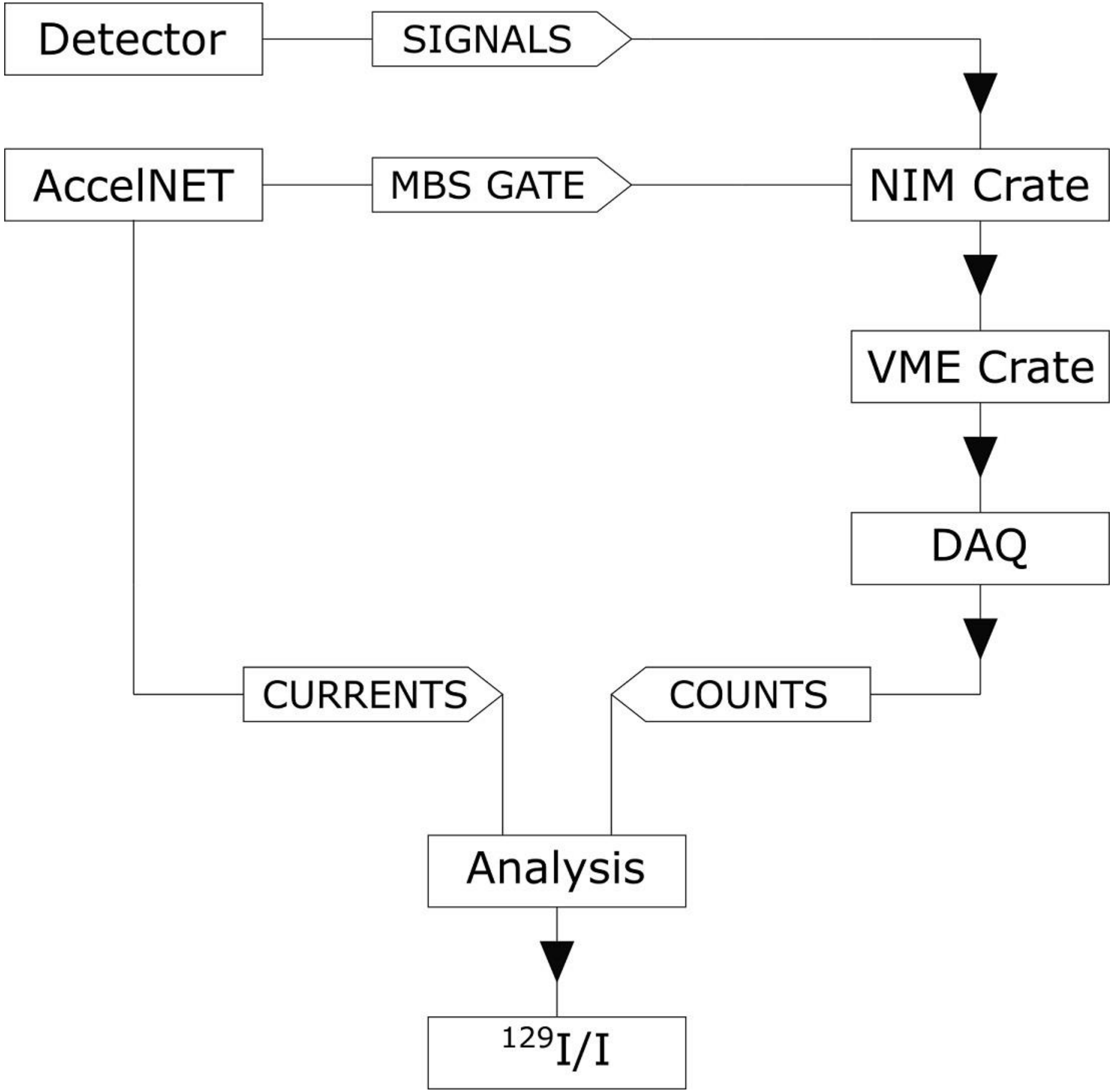

Incident Charged Particle
Pre-amplifier
Amplifier

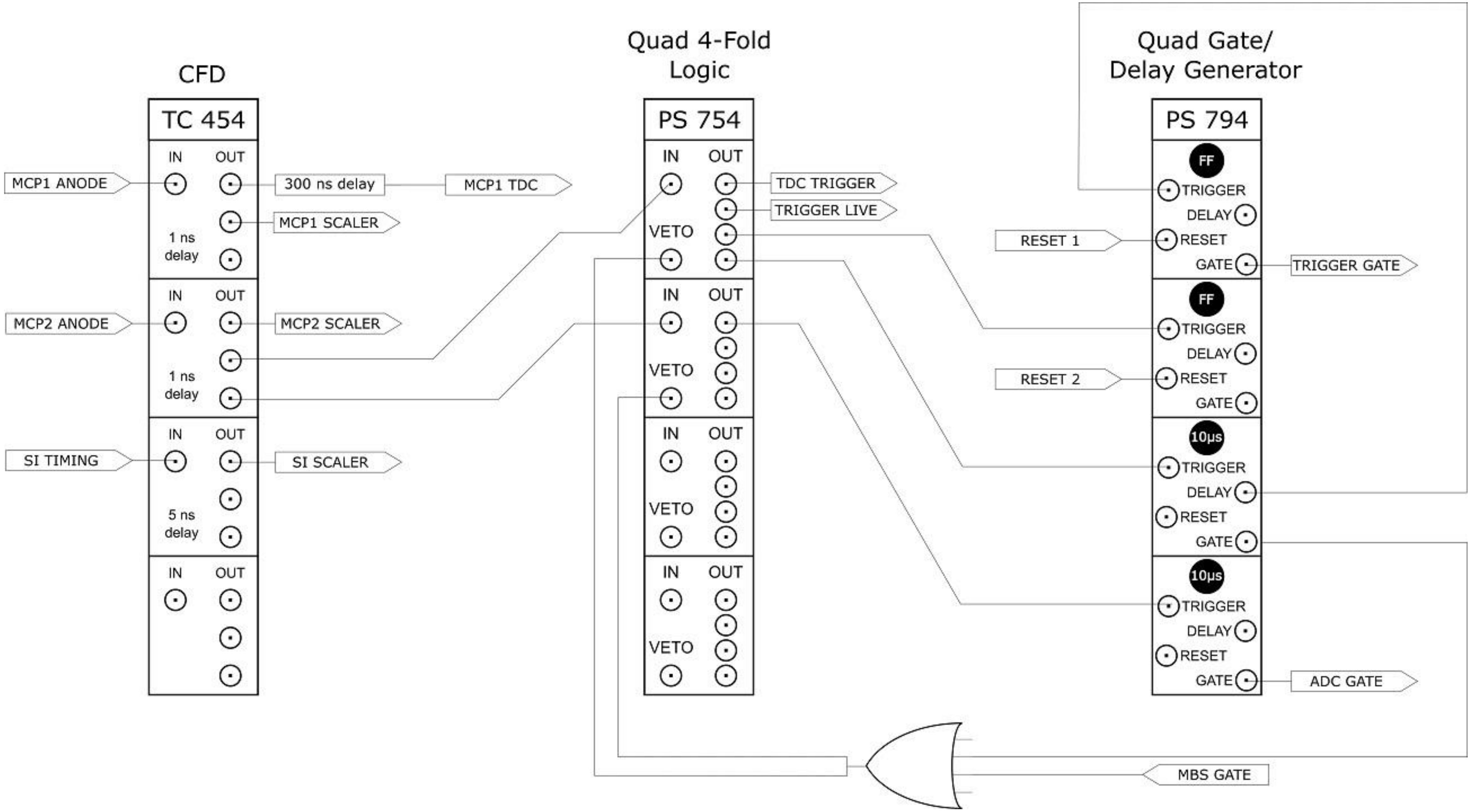

150 pF
150 pF
1500 nH
Energy Signal (to Pre-Amplifier)
Si Signal
Timing Signal (to Fast Timing Amplifier)
680 pF
560 pF
1500 nH

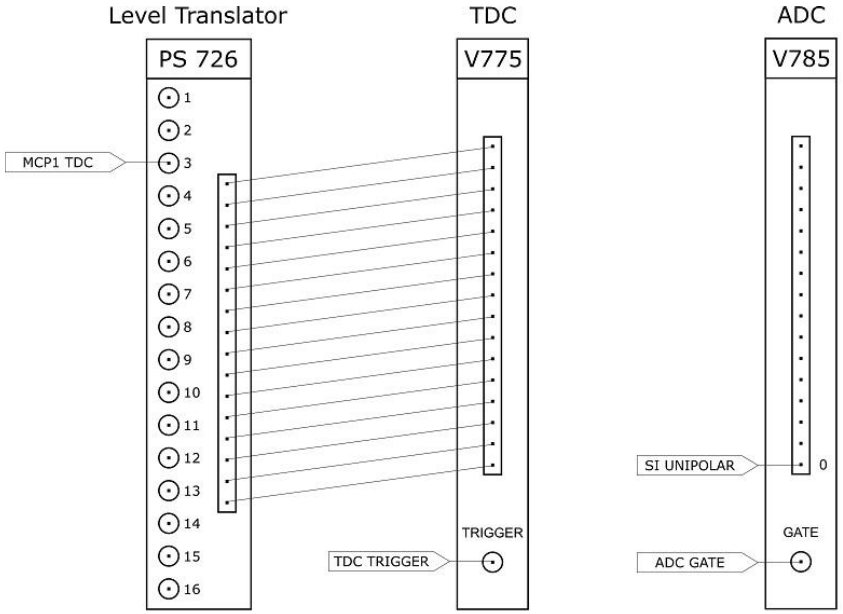
TOF DATA ACQUISITION SCHEME

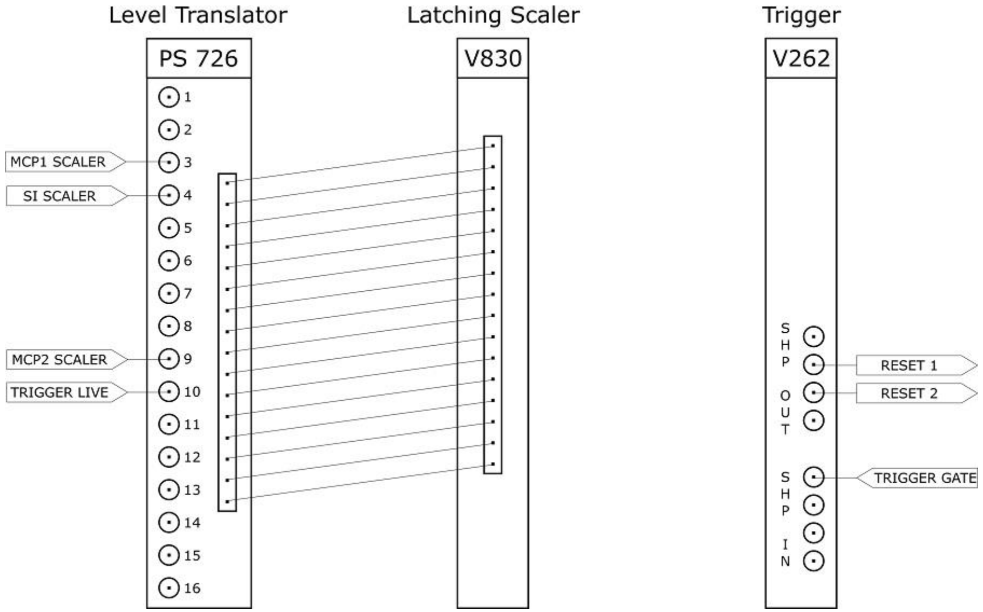


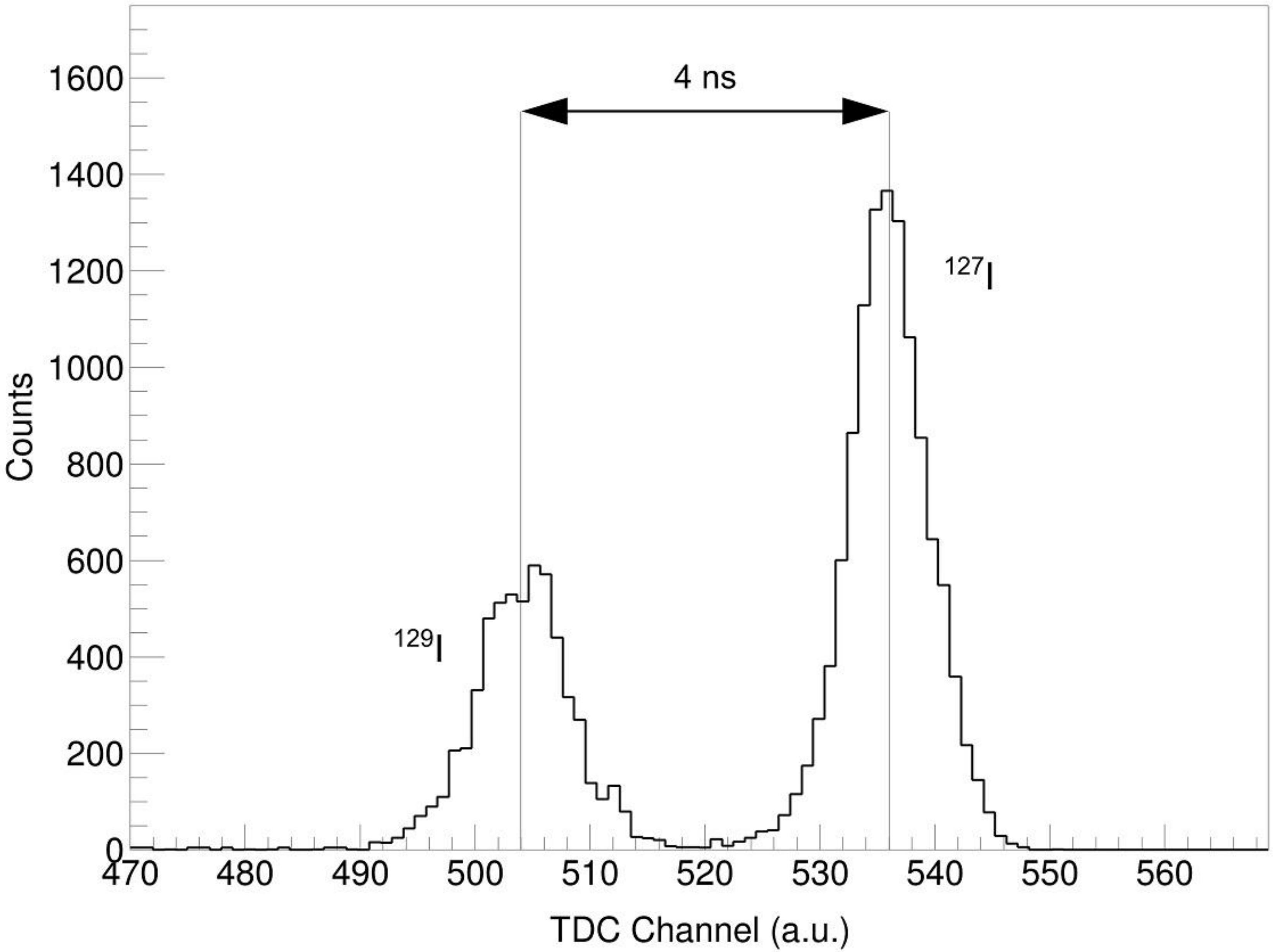
- Makes sure that only ions reaching the end of the beamline triggers the DAQ
- TOF signals are really Delay-TOF (larger values mean faster ions)


Detector
SIGNALS
AccelNET
MBS GATE
NIM Crate
VME Crate
DAQ
CURRENTS
COUNTS
Analysis
$^{129}I/I$


CFD
TC 454
IN
OUT
MCP1 ANODE
300 ns delay
MCP1 TDC
MCP1 SCALER
1 ns delay
MCP2 ANODE
MCP2 SCALER
1 ns delay
SI TIMING
SI SCALER
5 ns delay
Quad 4-Fold Logic
PS 754
VETO
TDC TRIGGER
TRIGGER LIVE
Quad Gate/ Delay Generator
PS 794
FF
TRIGGER
DELAY
RESET
GATE
RESET 1
TRIGGER GATE
RESET 2
10μs
ADC GATE
MBS GATE

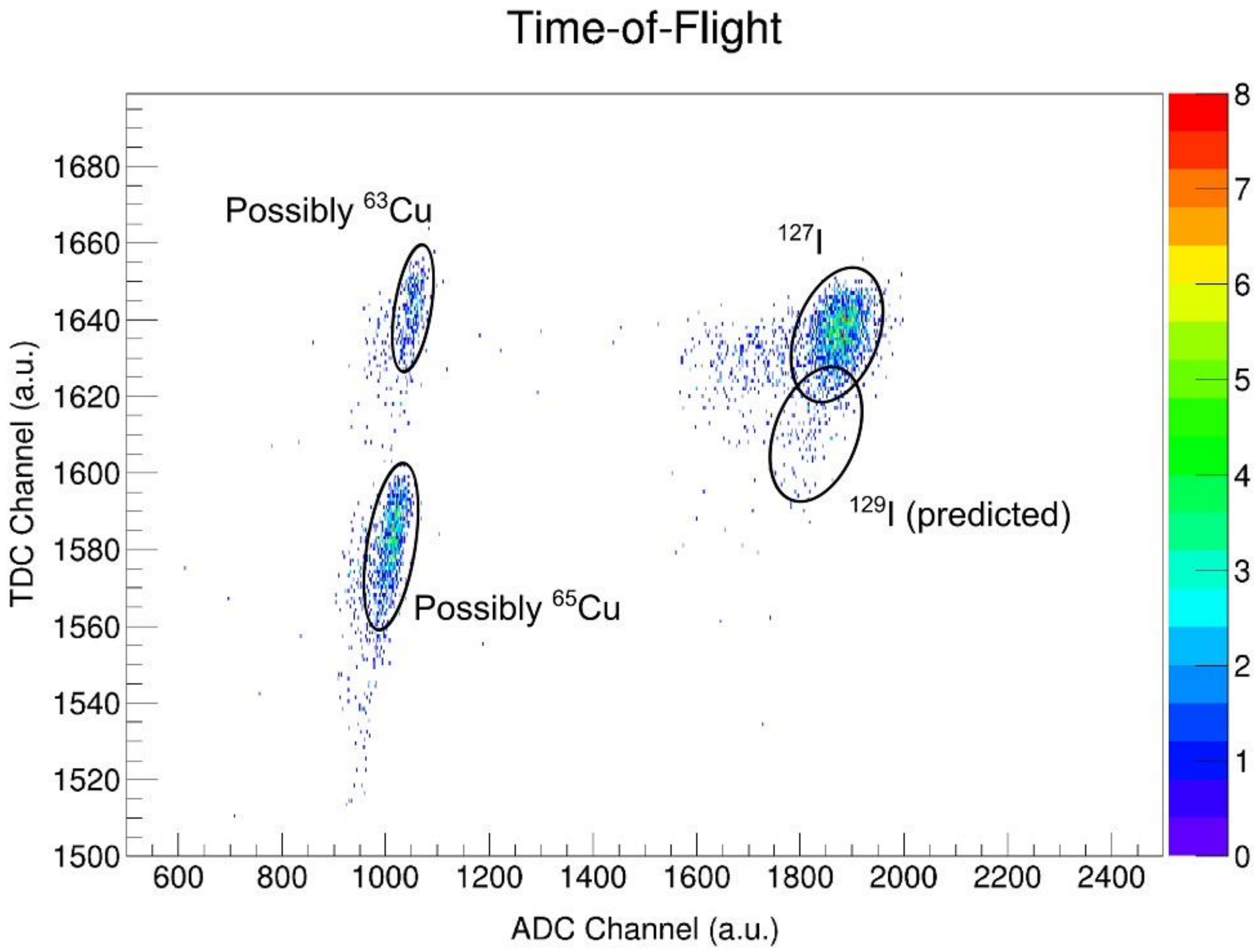

Level Translator
PS 726
TDC
V775
ADC
V785
MCP1 TDC
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
TRIGGER
TDC TRIGGER
SI UNIPOLAR
0
GATE
ADC GATE


Level Translator
PS 726
Latching Scaler
V830
Trigger
V262
MCP1 SCALER
SI SCALER
MCP2 SCALER
TRIGGER LIVE
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
SHP OUT
RESET 1
RESET 2
SHP IN
TRIGGER GATE


4 ns
^{129}I
^{127}I
Counts
1600
1400
1200
1000
800
600
400
200
0
470
480
490
500
510
520
530
540
550
560
TDC Channel (a.u.)

INTERFERENCES TO ^{129}I MEASUREMENT

- Charge state contaminants
- Sample contaminants
- Interferences during fast switching

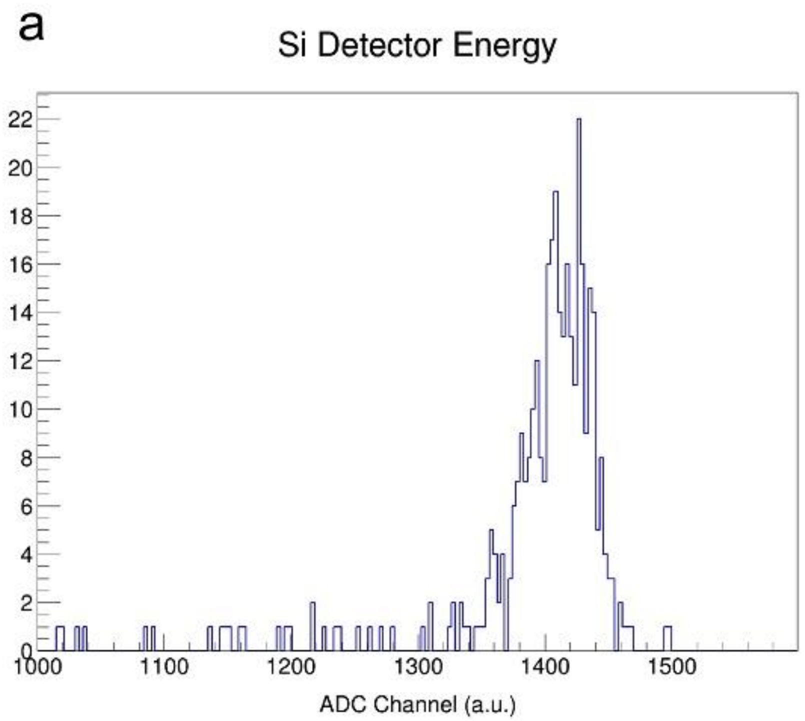


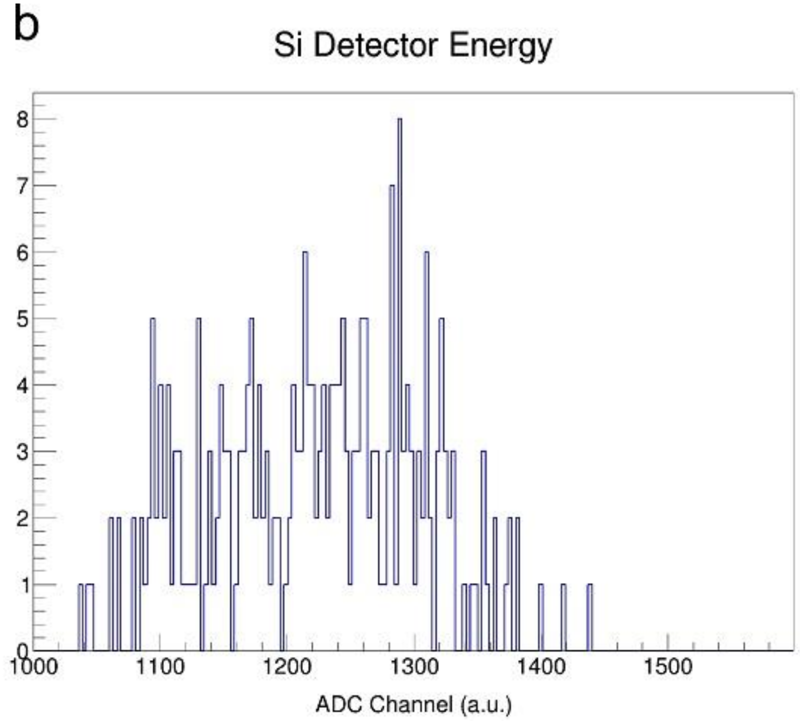
Time-of-Flight
TDC Channel (a.u.)
ADC Channel (a.u.)
Possibly 63Cu
Possibly 65Cu
127I
129I (predicted)
1500
1520
1540
1560
1580
1600
1620
1640
1660
1680
600
800
1000
1200
1400
1600
1800
2000
2200
2400
0
1
2
3
4
5
6
7
8

^{127}I

^{127}I not present during injection of ^{129}I, but during injection to the HE offset cup ^{127}I counts pass through the detector system, but are vetoed from the spectra

- Analyzing magnet, Wien filter slits kept narrow to reduce ^{127}I count rates

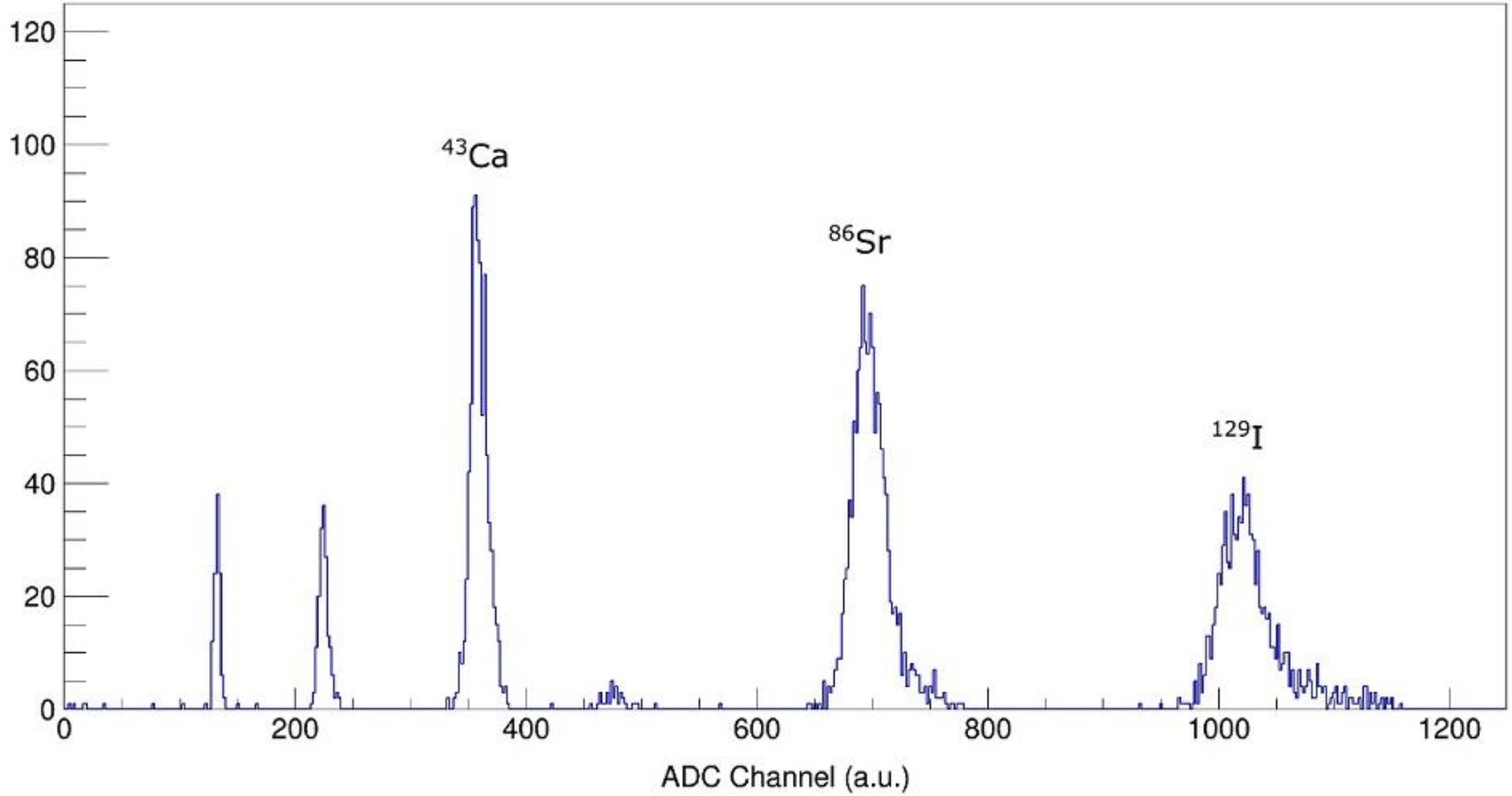




^{43}Ca, ^{86}Sr

Similarly to the copper dimers, ^{43}Ca and ^{86}Sr will make it through the system when using a ^{129}I charge state that is a multiple of 3, but is injected at mass 129

- Present in water samples, but the count rates are comparable to the ^{129}I count rates



200 mg I_2 crystals are added to an acid-leached HDPE bottle

↓

50 mL DI H_2O is added to the HDPE bottle, dissolving the I_2 crystals and making a light brown solution

↓

KOH + $Na_2S_2O_3$ reducing solution is added in 0.5 mL increments until the solution turns clear (all iodine is reduced)

↓

Solution is diluted to a total mass of 100 g with DI H_2O, for a final concentration of 2 mg I^- / g solution

1 g of carrier solution (~2 mg iodine) is added to an acid-leached 45 mL centrifuge tube

↓

40 mL water sample is added to the centrifuge tube

↓

10 drops of 16 M HNO_3 are added to convert iodine to I^- and prevent the reduction of silver during precipitation

↓

1 F $AgNO_3$ is added dropwise, in excess, to ensure all iodine is precipitated as AgI, vortexed to increase the rate of precipitation, and centrifuged for 15 minutes

↓

Solution is decanted, precipitate is washed with DI H_2O to 20 mL, vortexed, and centrifuged

↓

Solution is decanted, and 20 drops of 30% NH_4OH is added to dissolve silver halides other than AgI (primarily AgCl), diluted to 20 mL, vortexed, and centrifuged

↓

Solution is decanted, precipitate is washed with DI H_2O to 20 mL, 10 drops of 16 M HNO_3 is added to return the solution to low pH, vortexed, and centrifuged

↓

Solution is decanted, precipitate is washed with DI H_2O to 20 mL, vortexed, and centrifuged

↓

Solution is decanted, and the precipitate is dried in an oven at 65 °C overnight

Standard solution is added to an acid-leached 45 mL centrifuge tube and diluted to 20 mL with DI H_2O

↓

10 drops of 16 M HNO_3 are added to convert iodine to I^- and prevent the reduction of silver during precipitation

↓

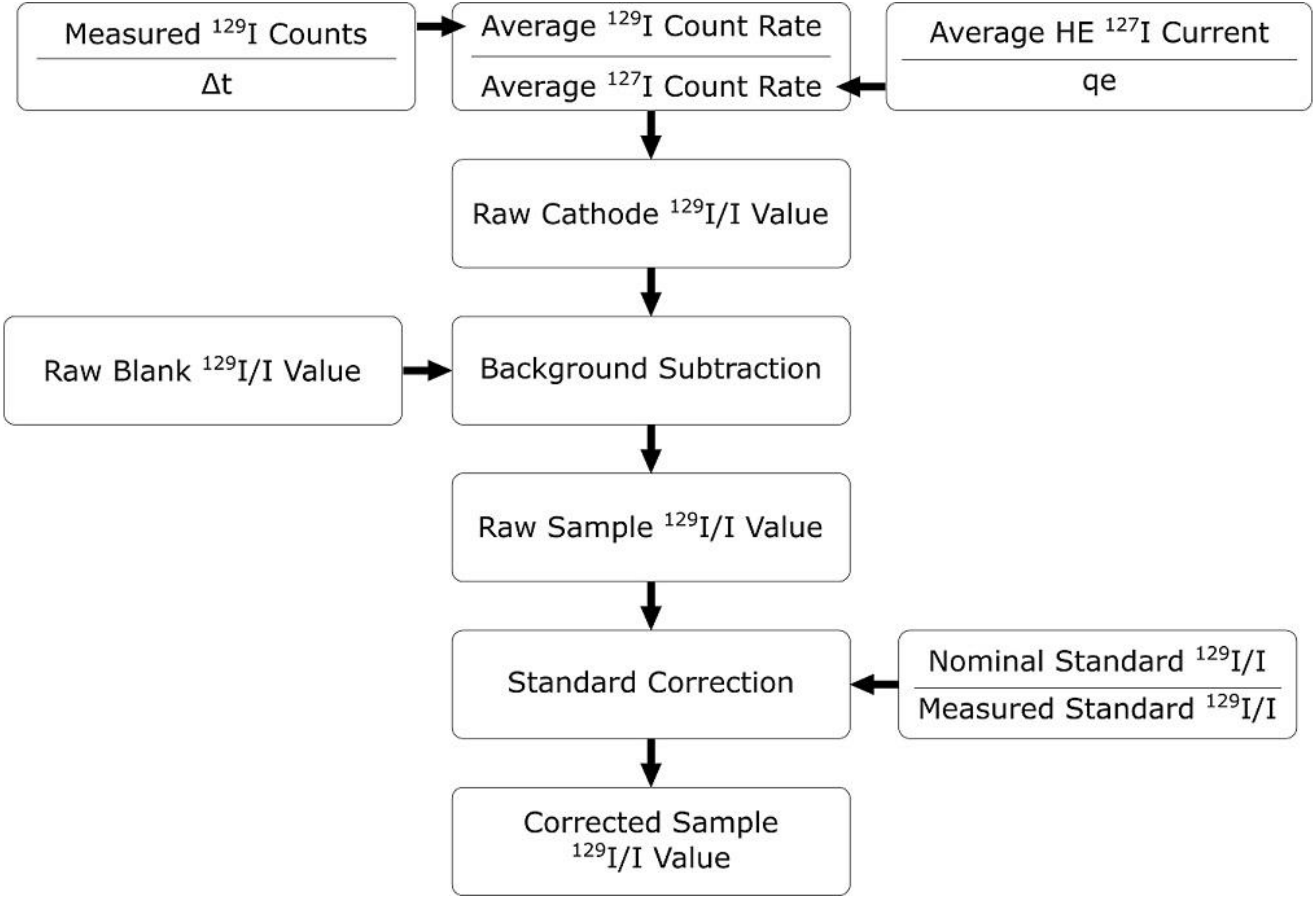
1 F $AgNO_3$ is added dropwise, in excess, to ensure all iodine is precipitated as AgI, vortexed to increase the rate of precipitation, and centrifuged for 15 minutes

↓

Solution is decanted, precipitate is washed with DI H_2O to 20 mL, vortexed, and centrifuged

↓

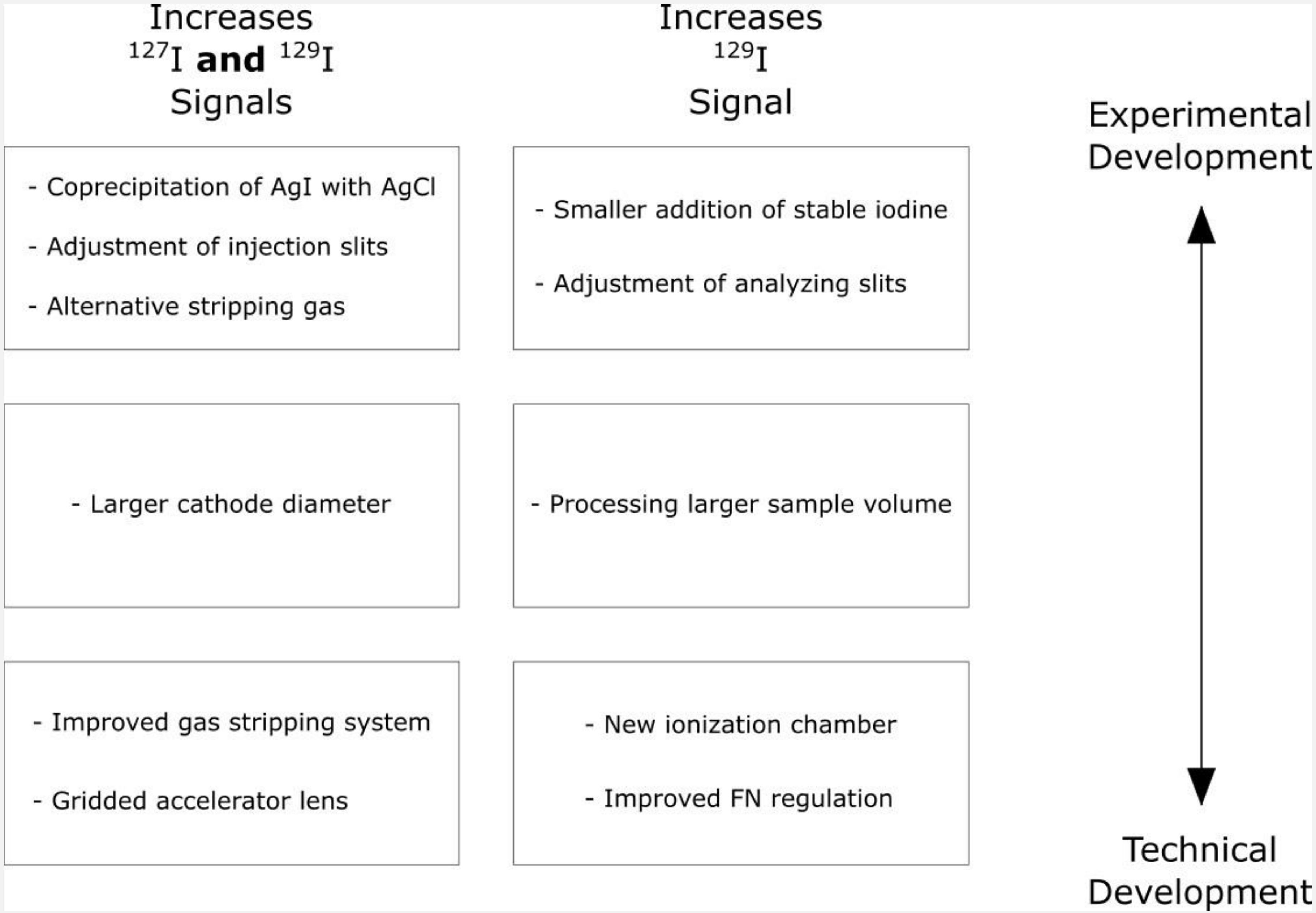
Solution is decanted, and the precipitate is dried in an oven at 65 ^{o}C overnight

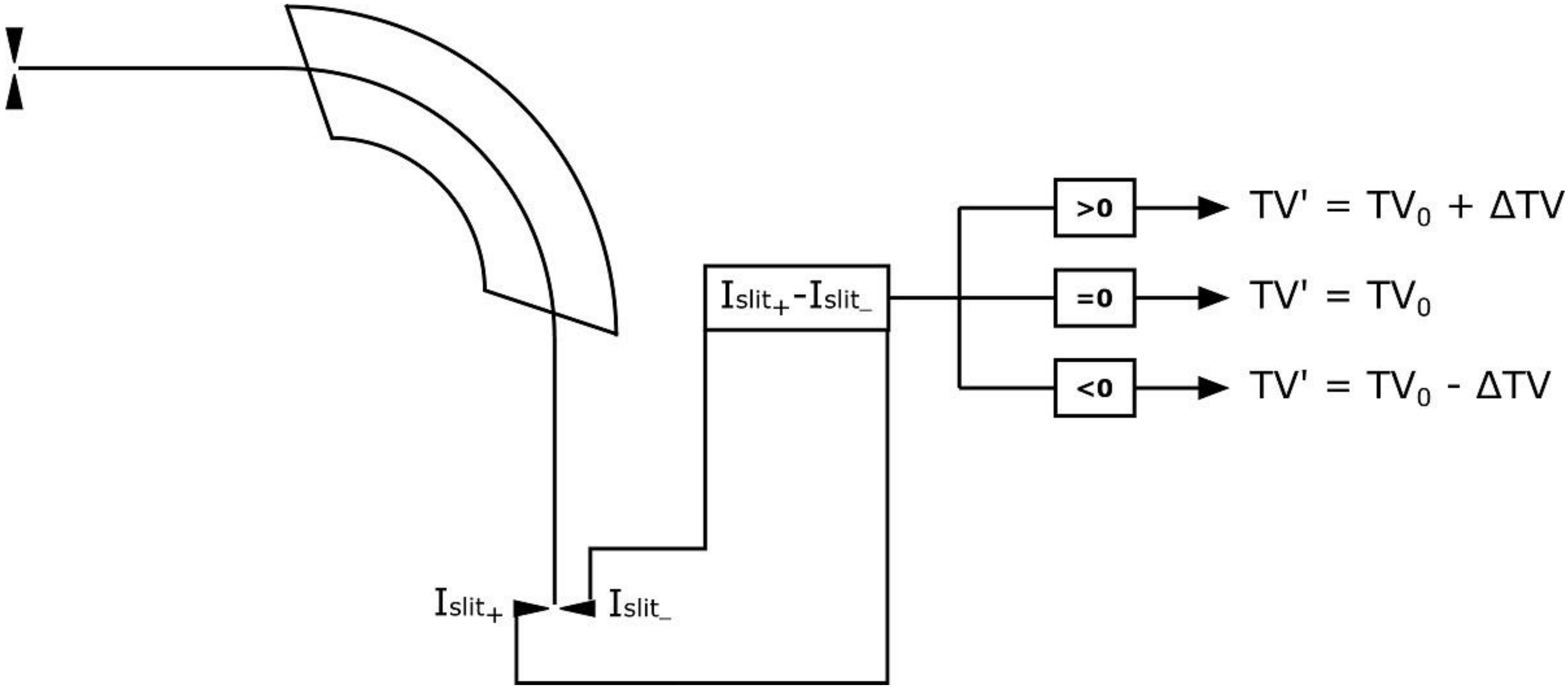


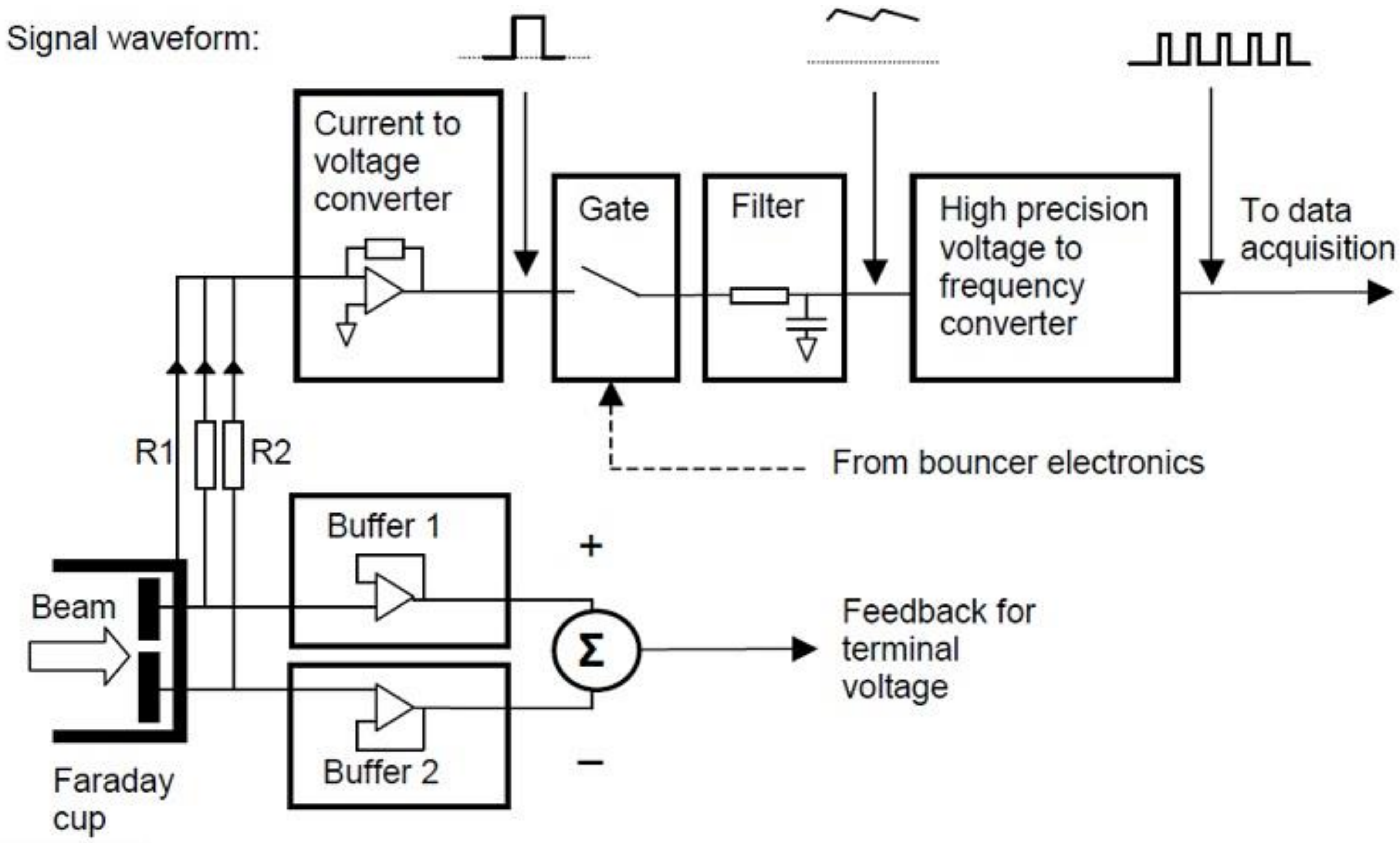
Measured 129I Counts
Δt
Average 129I Count Rate
Average 127I Count Rate
Average HE 127I Current
qe
Raw Cathode 129I/I Value
Raw Blank 129I/I Value
Background Subtraction
Raw Sample 129I/I Value
Standard Correction
Nominal Standard 129I/I
Measured Standard 129I/I
Corrected Sample 129I/I Value

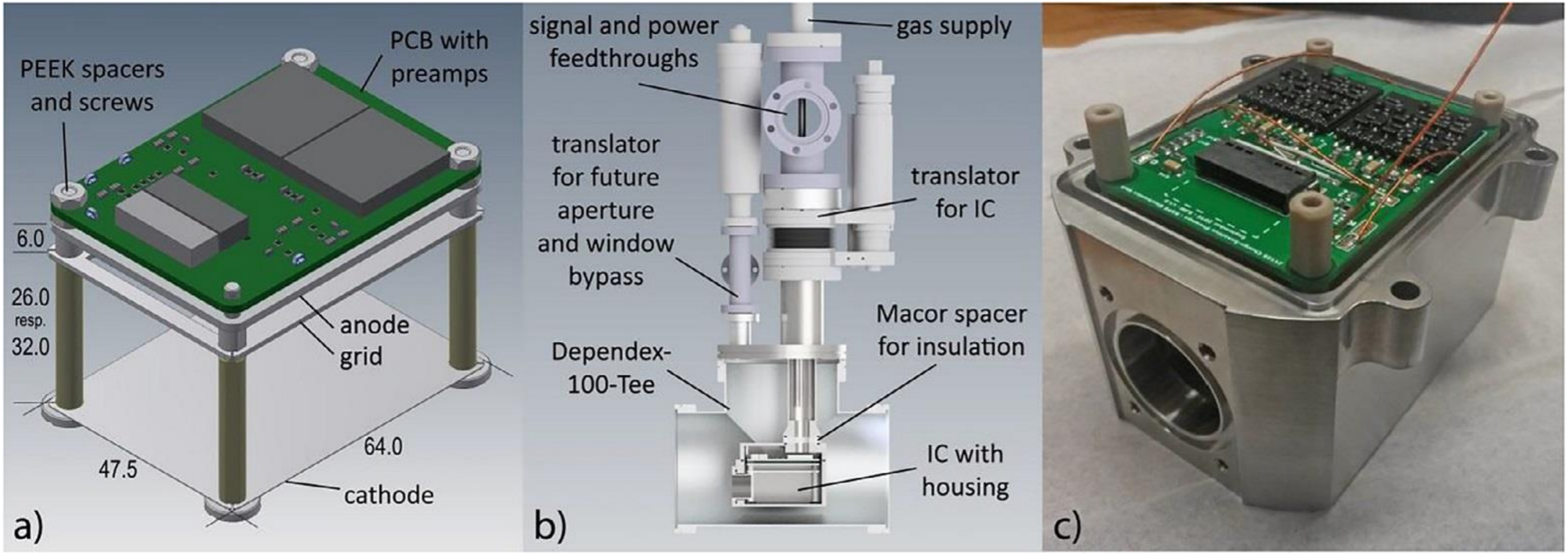
EQUIPMENT FAILURES

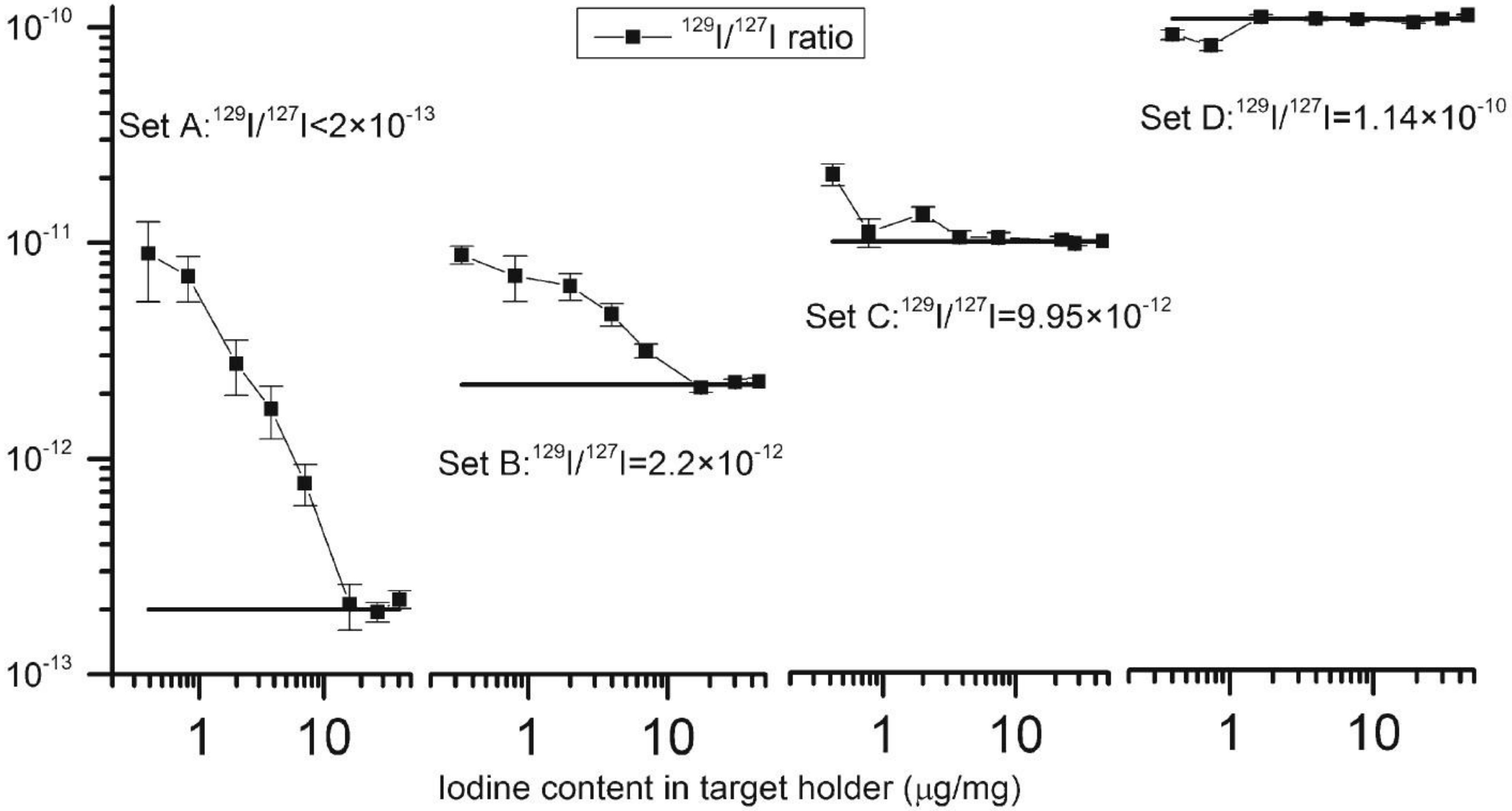
- Original 2 weeks
  - Accelerator chain failure – 1 week
  - Breakdown of Einzel lens – 4 days
  - Experiment shut down
- Restart 2 weeks
  - Analyzing magnet power supply drift – 5 days
  - Used up AMS standards being replaced – 2 days
  - Source being clogged with cesium and cleaned – 1 day
  - Source being out of cesium, replaced – 1 day
- Last ~5 days
  - Only 8 samples could be measured (low sample currents, low $^{129}I/I$ concentration)

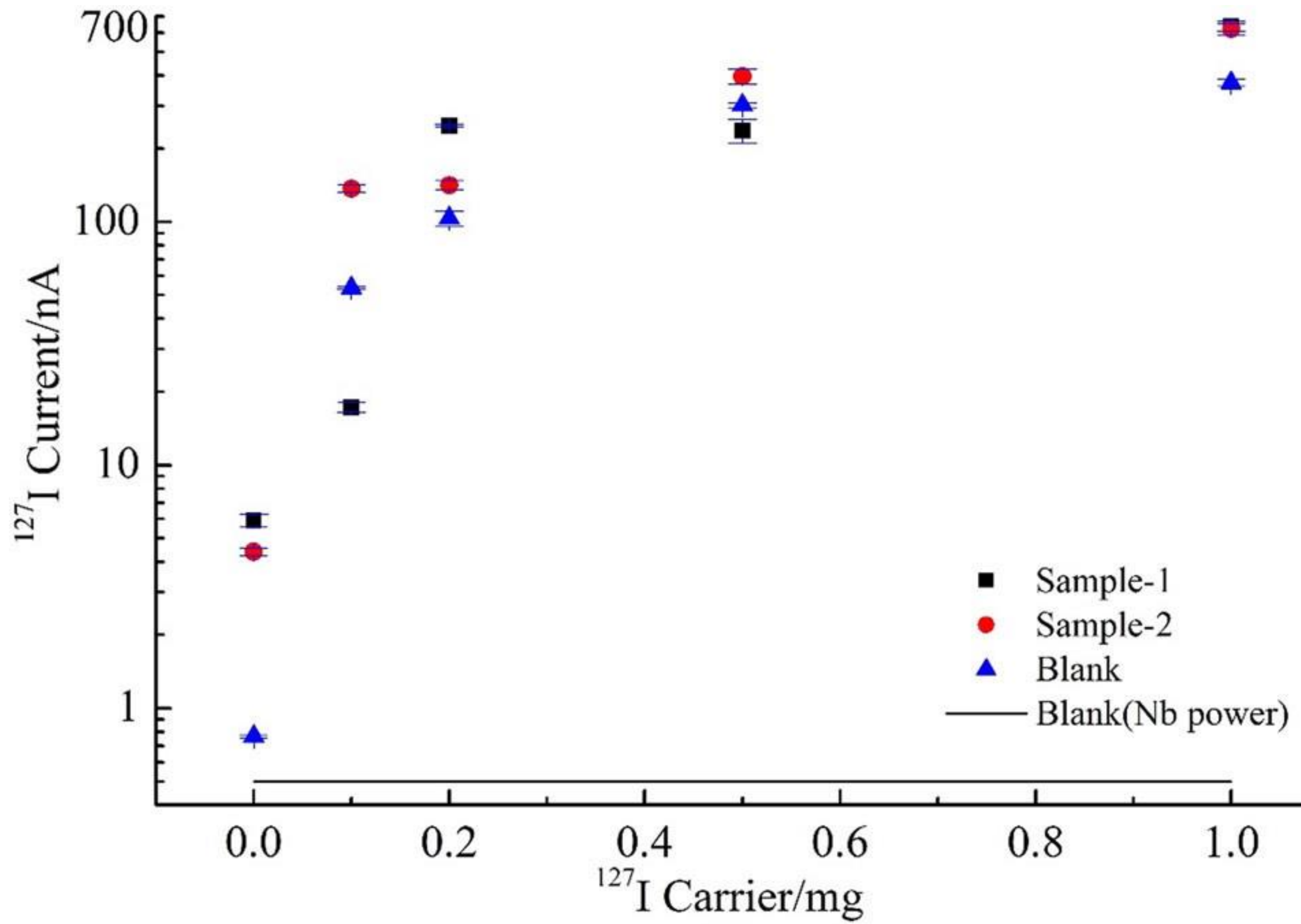

Increases
^{127}I and ^{129}I
Signals
Increases
^{129}I
Signal
Experimental
Development
- Coprecipitation of AgI with AgCl
- Adjustment of injection slits
- Alternative stripping gas
- Smaller addition of stable iodine
- Adjustment of analyzing slits
- Larger cathode diameter
- Processing larger sample volume
- Improved gas stripping system
- Gridded accelerator lens
- New ionization chamber
- Improved FN regulation
Technical
Development


Islit+-Islit_
>0
=0
<0
TV' = TV0 + ΔTV
TV' = TV0
TV' = TV0 - ΔTV
Islit+
Islit_

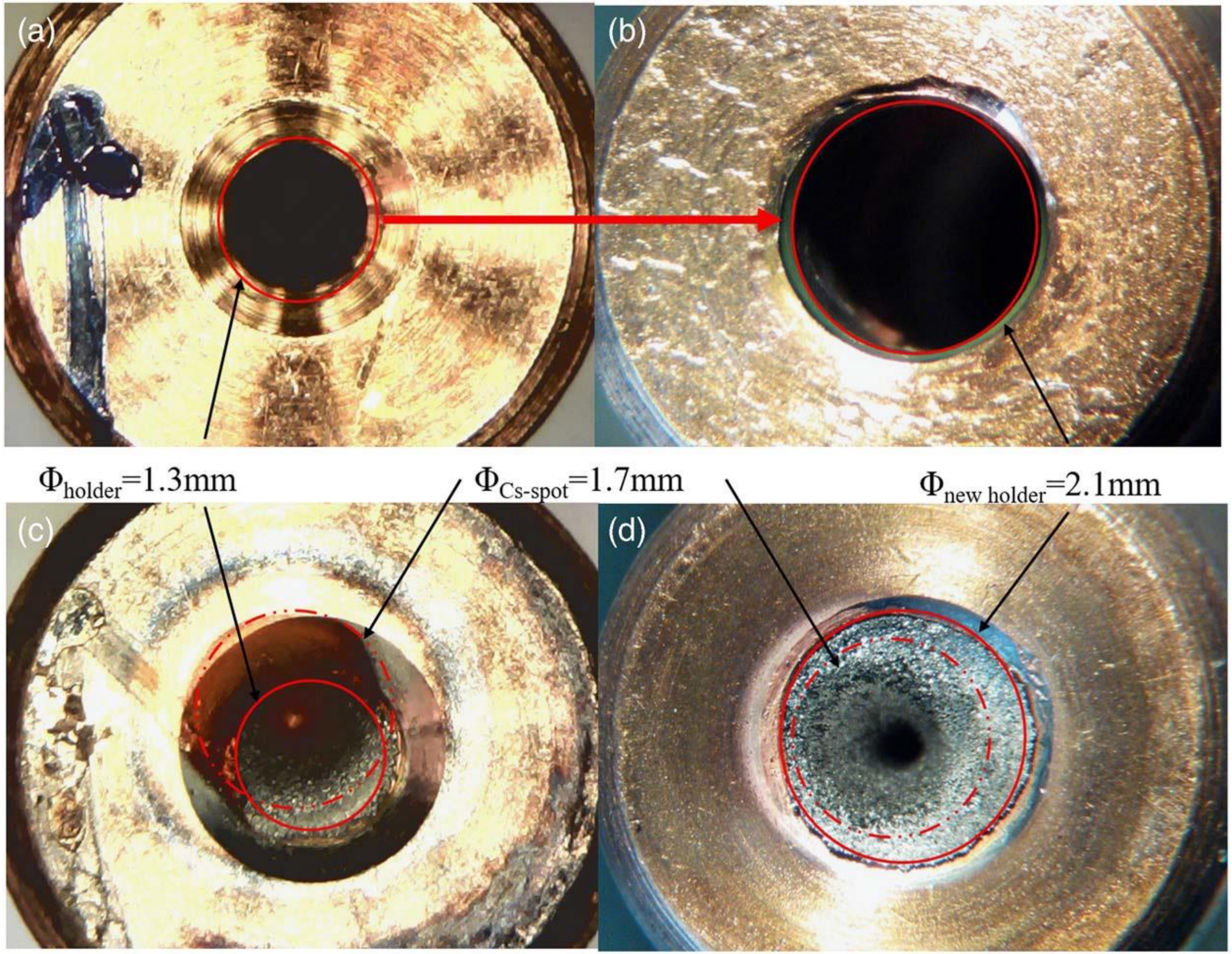

Signal waveform:
Current to voltage converter
Gate
Filter
High precision voltage to frequency converter
To data acquisition
R1
R2
From bouncer electronics
Buffer 1
Buffer 2
+
Σ
−
Feedback for terminal voltage
Beam
Faraday cup


PEEK spacers
and screws
PCB with
preamps
6.0
26.0
resp.
32.0
anode
grid
47.5
64.0
cathode
a)
signal and power
feedthroughs
gas supply
translator
for future
aperture
and window
bypass
translator
for IC
Macor spacer
for insulation
Dependex-
100-Tee
IC with
housing
b)
c)


129I/127I ratio
Set A: 129I/127I<2×10-13
Set B: 129I/127I=2.2×10-12
Set C: 129I/127I=9.95×10-12
Set D: 129I/127I=1.14×10-10
10-10
10-11
10-12
10-13
1
10
Iodine content in target holder (μg/mg)



700
100
10
1
0.0
0.2
0.4
0.6
0.8
1.0
127I Current/nA
127I Carrier/mg
Sample-1
Sample-2
Blank
Blank(Nb power)


(a)
(b)
(c)
(d)
Φholder=1.3mm
ΦCs-spot=1.7mm
Φnew holder=2.1mm