

Progress of an accelerator mass spectrometry system on the Tsukuba 12UD Pelletron tandem accelerator

*-11th International Conference on Heavy Ion Accelerator Technology - Venezia (Italy) –
8-12, June, 2009*

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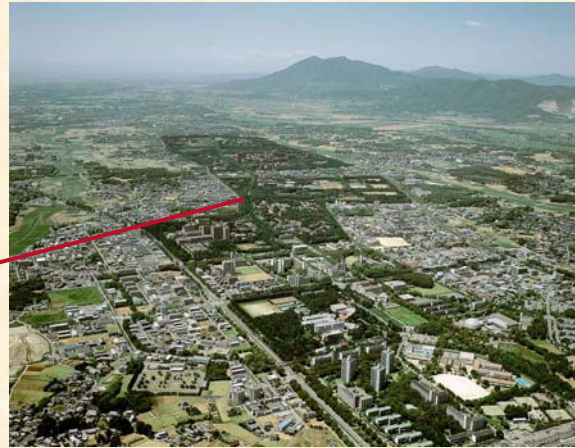


Outline of presentation

- **Introduction**
 - 12UD Pelletron tandem at the University of Tsukuba
 - AMS and facilities
- **AMS system on the 12UD Pelletron tandem**
 - Description of the Tsukuba AMS system
 - Recent progress
 - Performance of ^{26}Al , ^{36}Cl and ^{129}I AMS
- **Summary and future plans**

- **Introduction**

- **12UD Pelletron tandem at the University of Tsukuba**
- **AMS and facilities**



University of Tsukuba,
Tsukuba science city

60 km from Tokyo in the northeast

2 accelerator facilities

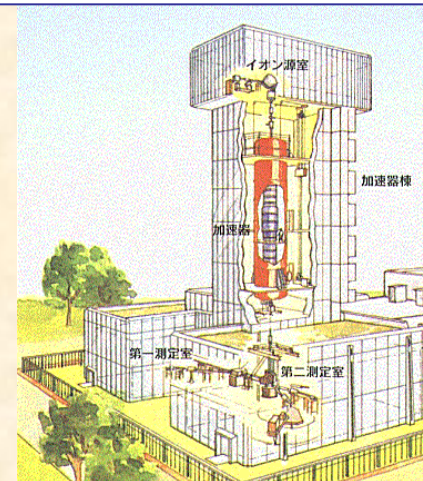
Proton Medical Research Center: PMRC

Tandem Accelerator Complex: UTTAC



• 250 MeV Proton Synchrotron (2001)

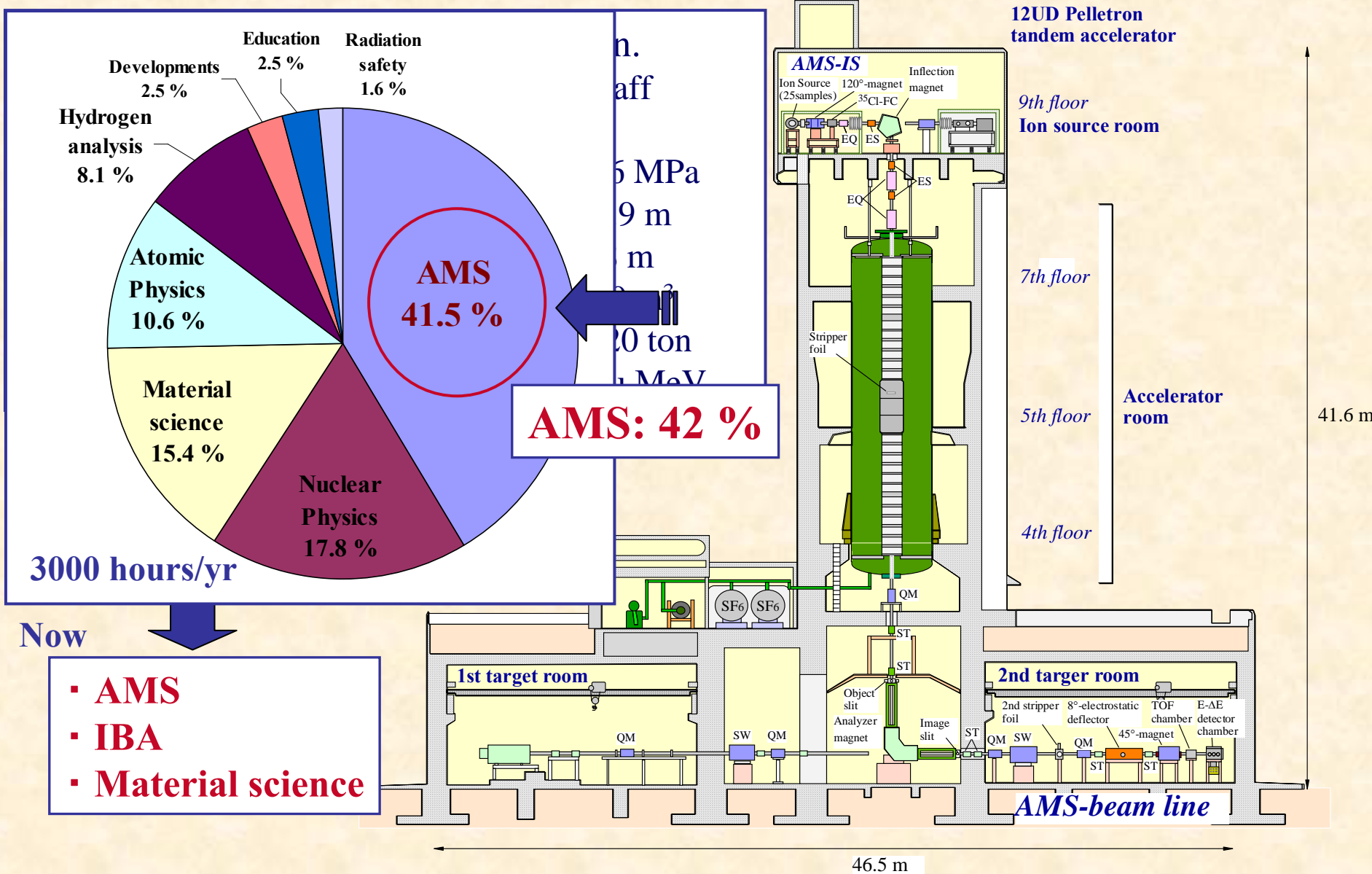
➔ Proton Beam Radiotherapy



• 12UD Pelletron Tandem Accelerator (1975)
• 1MV Tandetrion Accelerator (1987)



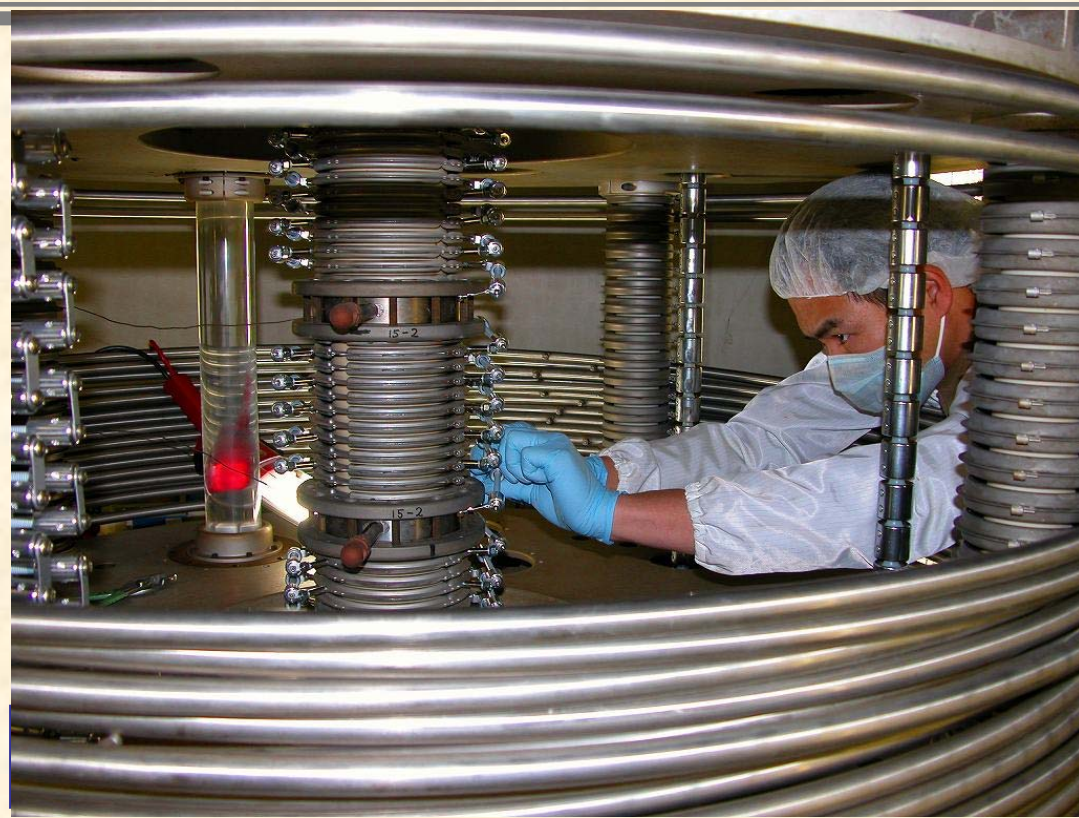
12UD Pelletron tandem accelerator (1975)



Upgrade of the 12UD Pelletron tandem



12UD at Tsukuba University, 1975



2009 Divided resistor system

We replaced the old corona needles with the divided resistor system.

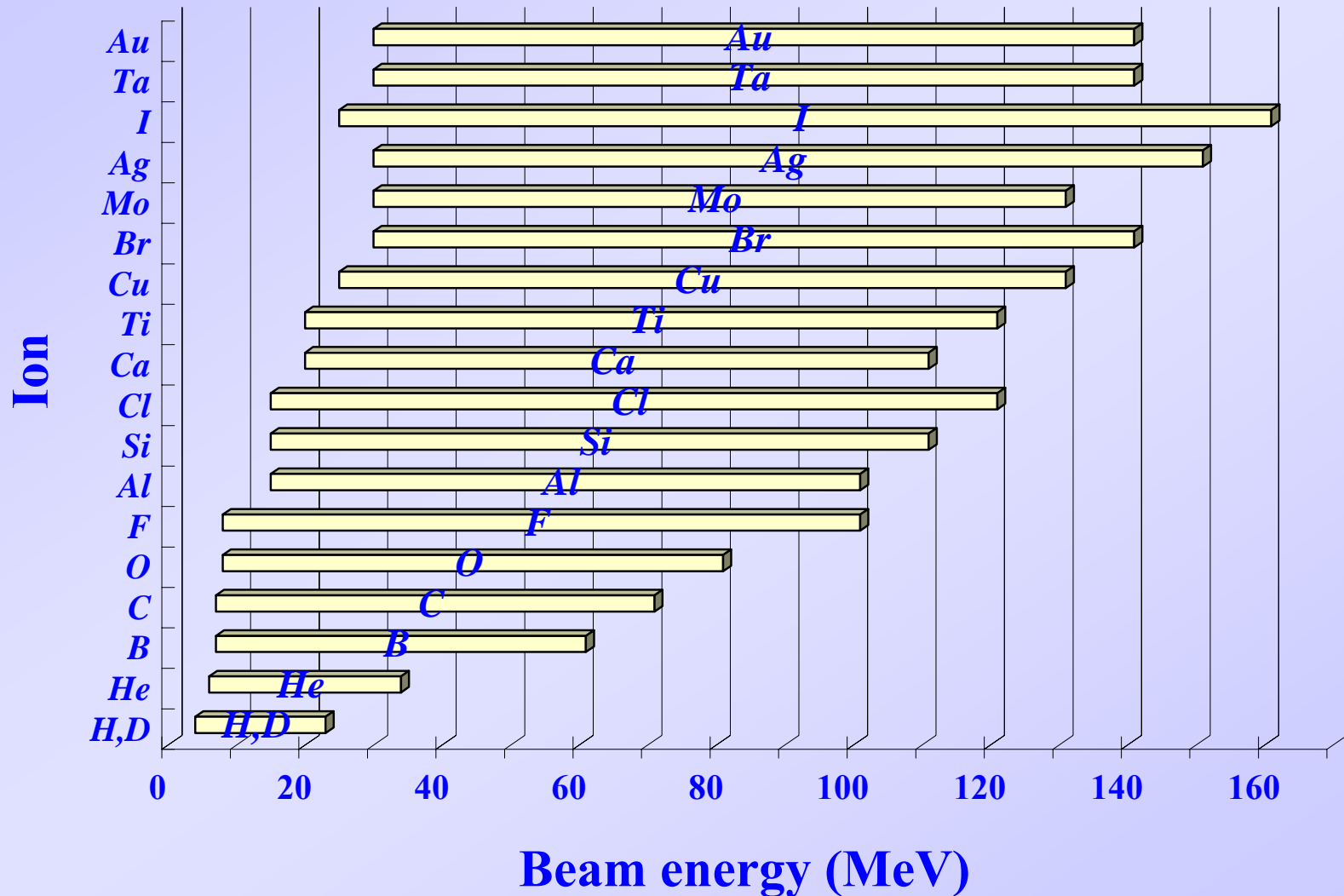
Variable terminal voltage
(No shorting column)



$$V_t = 1 \sim 12 \text{ MV}$$

Beam energy for the 12UD Pelletron

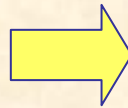
Terminal voltage: 1 - 12 MV



Accelerator Mass Spectrometry

Targets of AMS

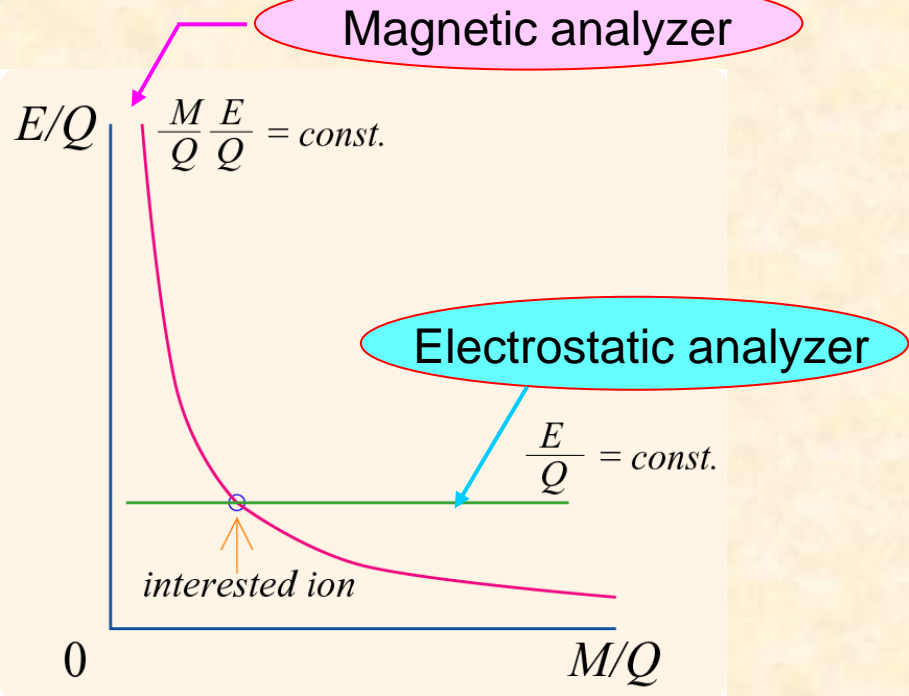
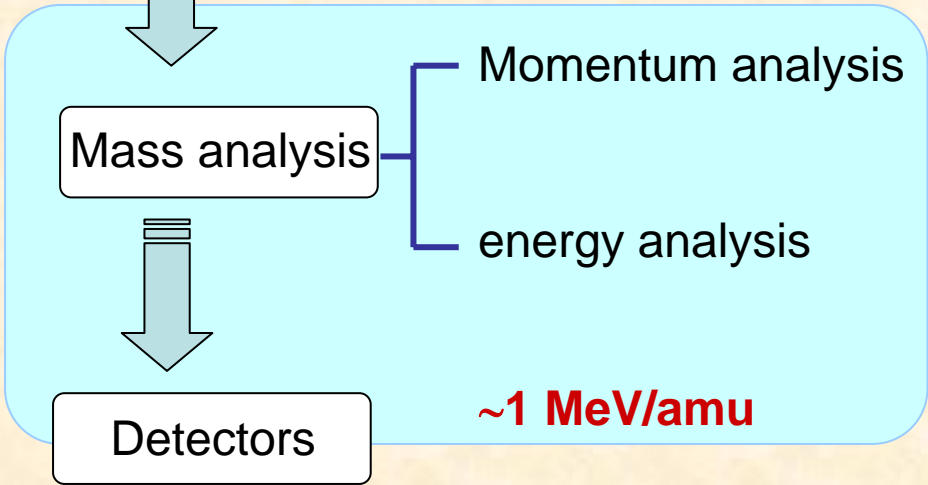
- ^{10}Be ($T_{1/2}=1.36\times 10^6$ yr)
- ^{14}C (5730 yr)
- ^{26}Al (7.1×10^5 yr)
- ^{36}Cl (3.0×10^5 yr)
- ^{129}I (1.57×10^7 yr)
- ...



Isobar, Isobaric Molecular suppression

Accelerator

High energy part



AMS facilities

^{14}C -AMS

LLNL, KIGAM...

3MV

→ 1MV

→ 500 kV

Oxford, Groningen, Kiel,
Arizona, NOSAMS,
Nagoya, SNU, ...

ETH, Georgia, UC-Irvine,
Posnan, Peking, Paleo-labo,
...

Multi Nuclides AMS

^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{129}I , ...

5MV and higher (← originally for nuclear physics)

LLNL(10MV), PRIME-Lab(8MV), ANSTO(9MV), ANU(14MV),
ETH(6MV), Tokyo(5MV), Tsukuba(12MV), SWERC(5MV)


3MV

VERA, LUND, JAEA-Mutsu, ...

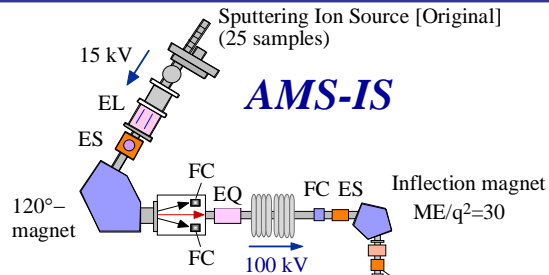
- **AMS on the 12UD Pelletron tandem**
 - **Description of the Tsukuba AMS system**
 - **Recent progress**
 - **Performance of ^{26}Al , ^{36}Cl and ^{129}I AMS**

Tsukuba AMS system

Progress of the Tsukuba AMS system

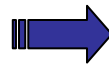
- | | |
|-----------|--|
| 1993-1996 | Trial AMS measurement for ^{14}C . |
| 1996-1998 | Development of AMS system
^{14}C -AMS
AMS ion source (original)
Mass separator beam line |
| 1999- | Development of ^{26}Al , ^{36}Cl -AMS
Pilot beam methods (Instead of GVM control) |
| 2002- | Development of ^{129}I -AMS |
| 2007- | ^{36}Cl AMS 9 MV \rightarrow 10 MV (Improved beam line)
Background: $^{36}\text{Cl}/\text{Cl} < 1 \times 10^{-15}$
Repetition accuracy: $\pm 3\%$ |
| 2009- | Upgrade of the 12UD Pelletron (Resister system)
 GVM terminal control system |

Tsukuba AMS system



AMS-IS

A pilot beam method is used to stabilize the terminal voltage.



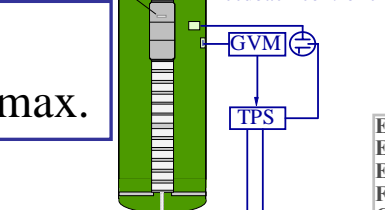
The terminal voltage is kept stable within 0.1 %.

12UD Pelletron tandem accelerator

^{26}Al , ^{36}Cl , ^{129}I - AMS

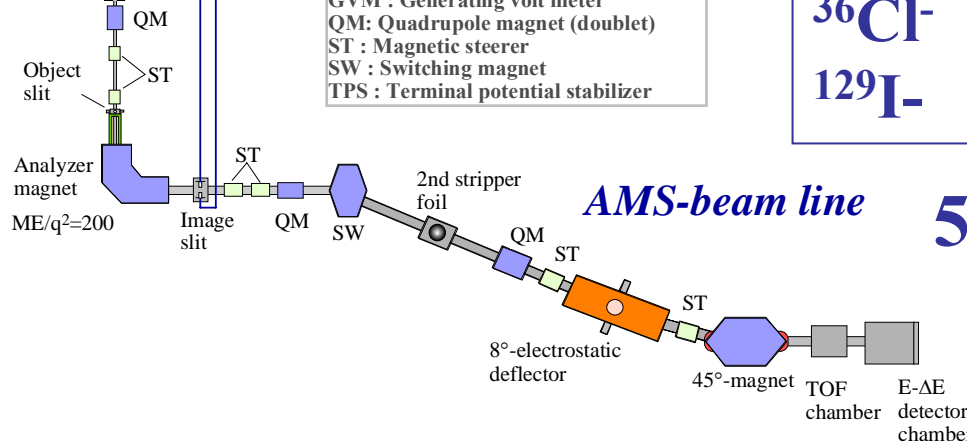
$V_T = 12 \text{ MV max.}$

1st Stripper foil
Slit current feedback controller



EL : Einzel lens
ES : Electrostatic steerer
EQ : Electrostatic quadrupole (triplet)
FC : Faraday cup
GVM : Generating volt meter
QM : Quadrupole magnet (doublet)
ST : Magnetic steerer
SW : Switching magnet
TPS : Terminal potential stabilizer

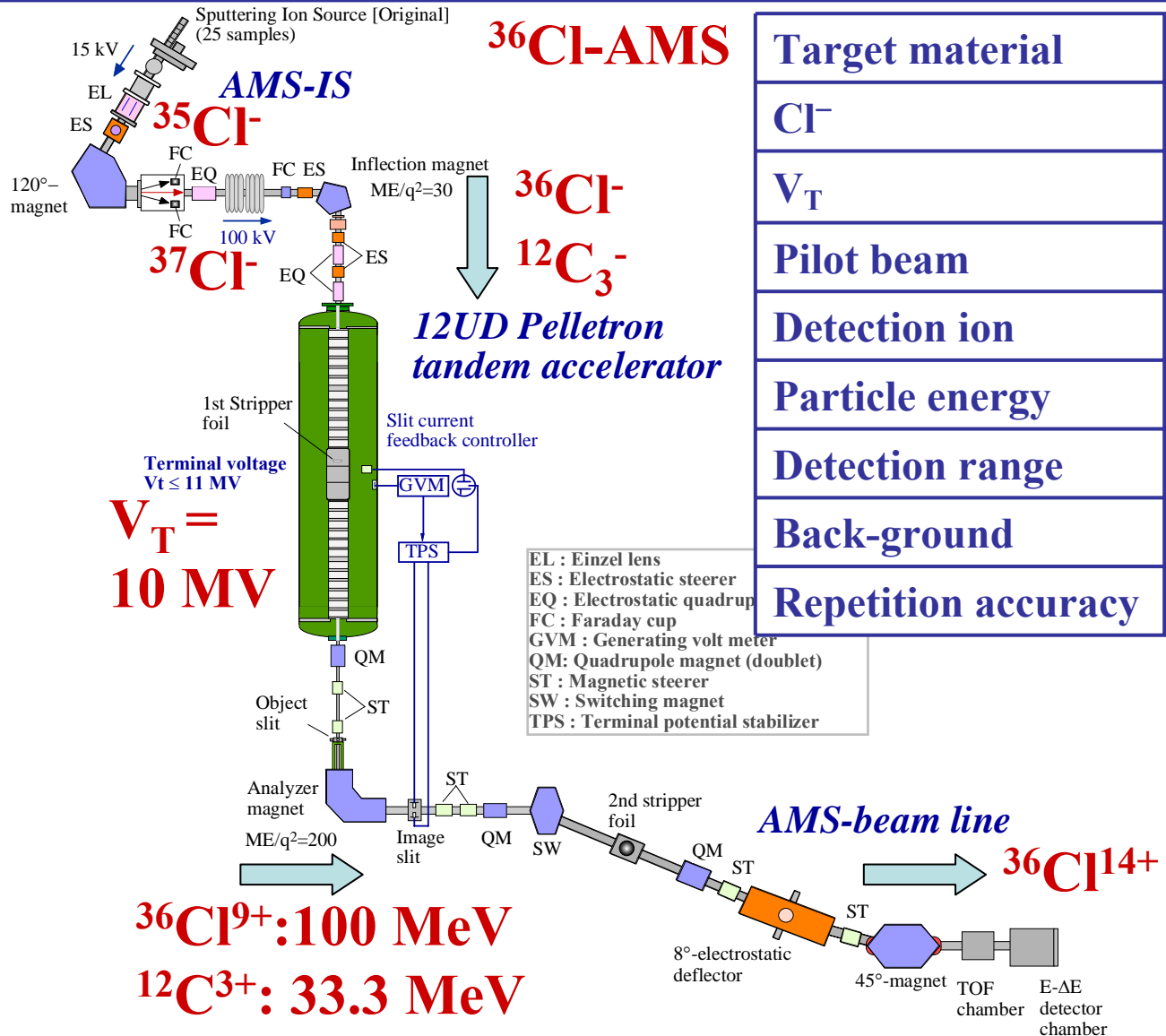
Pilot beams (isobar)



AMS-beam line

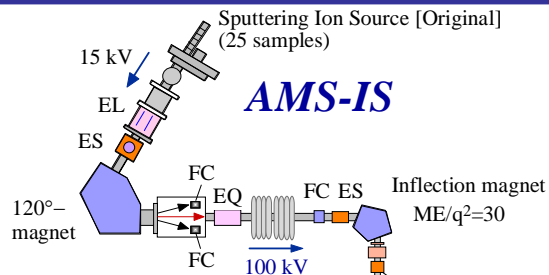
500 samples/year.

^{36}Cl -AMS by the Tsukuba AMS system



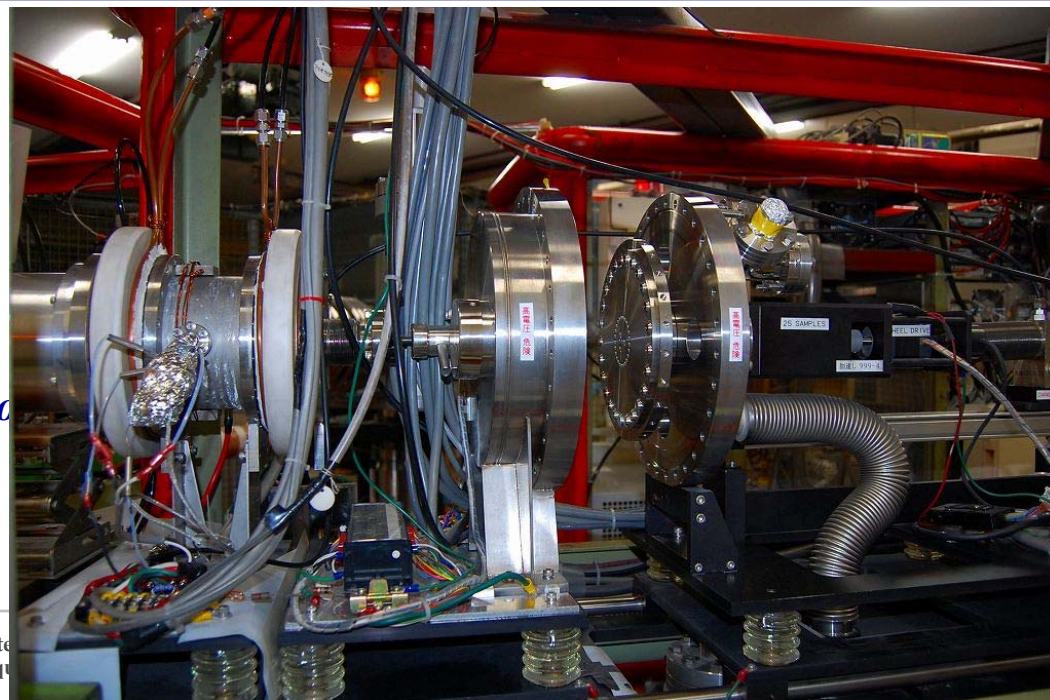
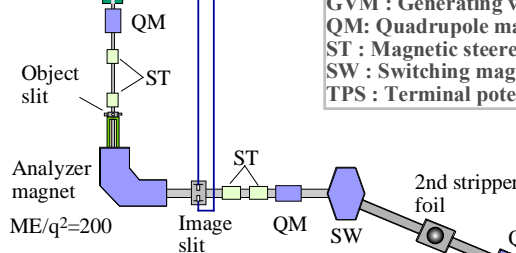
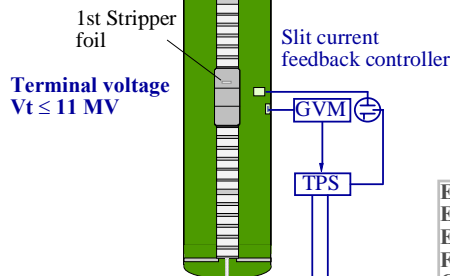
Target material	AgCl + C ₆₀
Cl ⁻	~ 20 μA
V _T	10 MV
Pilot beam	$^{12}\text{C}_3^-$
Detection ion	$^{36}\text{Cl}^{14+}$
Particle energy	100 MeV
Detection range	$^{36}\text{Cl}/\text{Cl} = 10^{-10} \sim 10^{-14}$
Back-ground	$^{36}\text{Cl}/\text{Cl} < 10^{-15}$
Repetition accuracy	± 3 % ($^{36}\text{Cl}/\text{Cl} \sim 10^{-12}$)

AMS Cs sputtering ion source



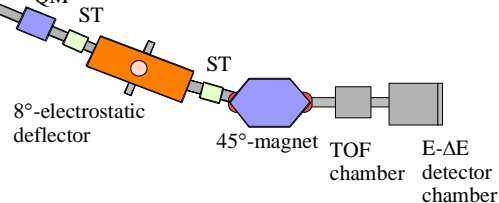
AMS-IS

**12UD Pelletron
tandem accelerator**



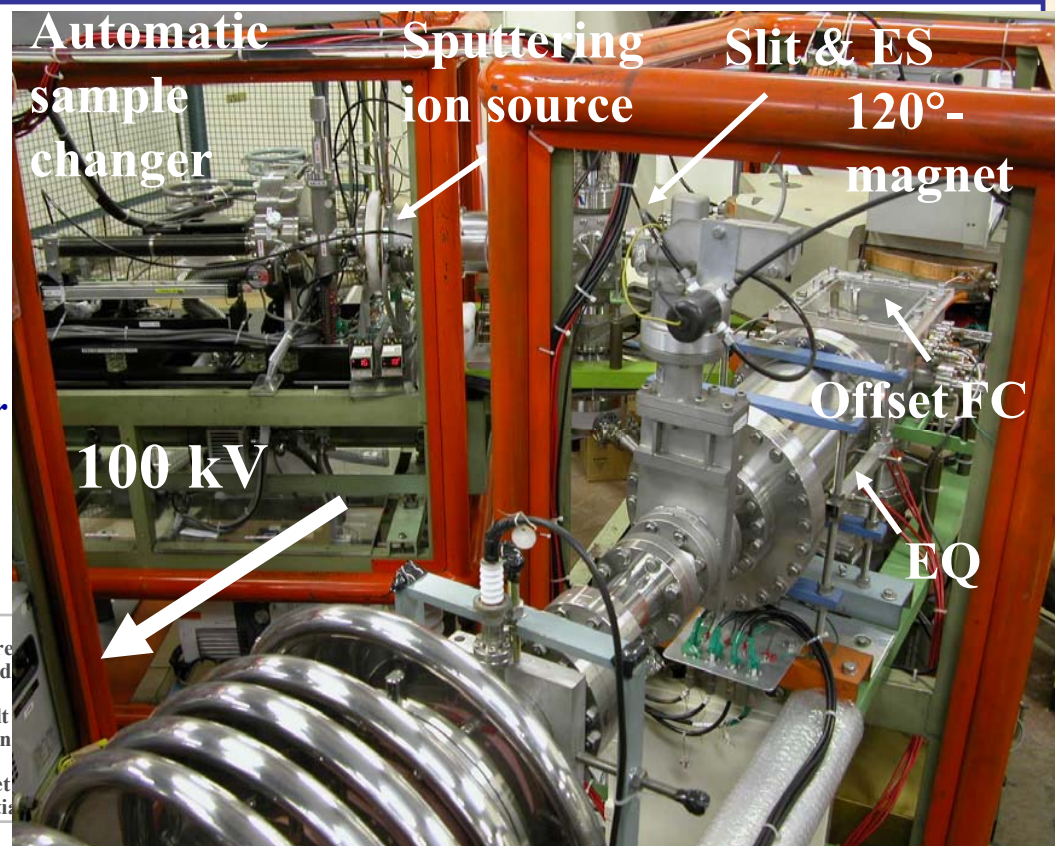
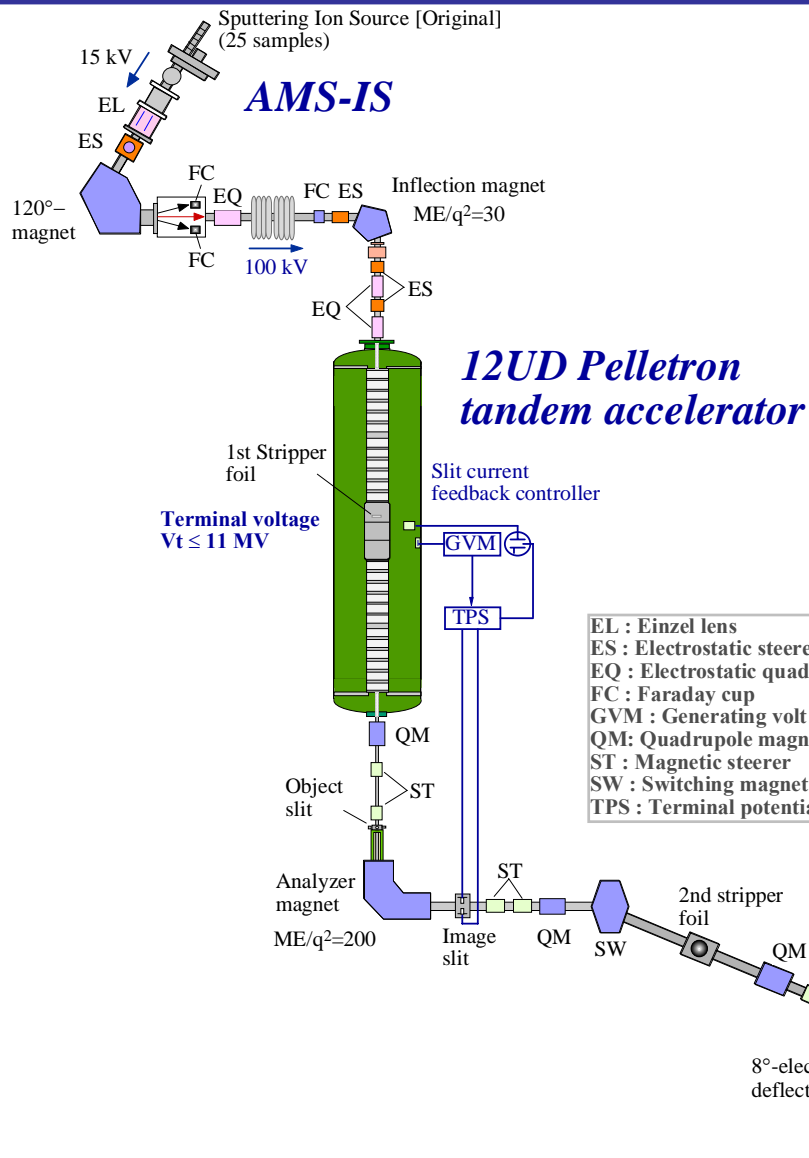
Original 25-sample changer

AMS-beam line

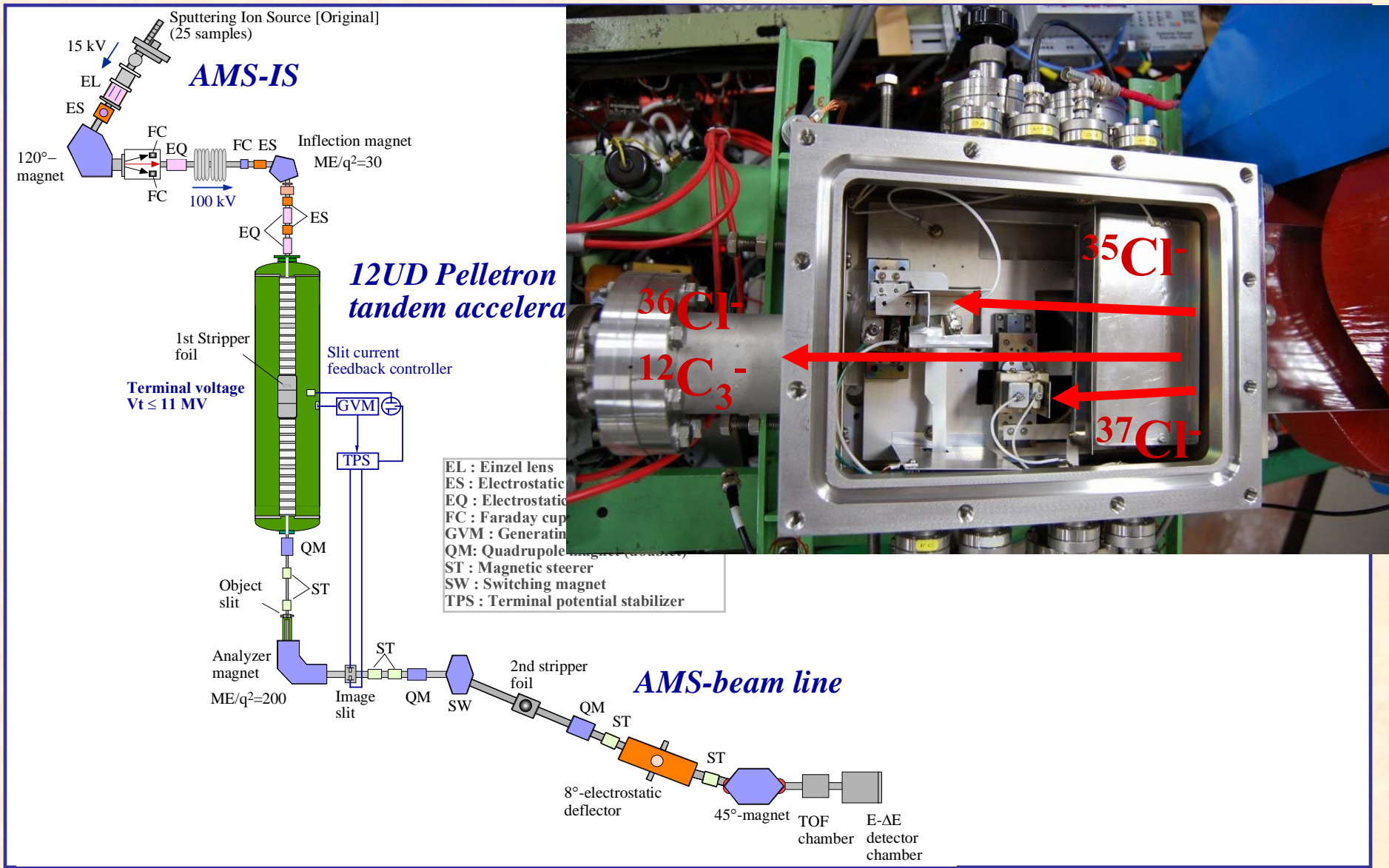


EL : Einzel lens
 ES : Electrostatic steering
 EQ : Electrostatic quadrupole
 FC : Faraday cup
 GVM : Generating volt meter
 QM : Quadrupole magnet
 ST : Magnetic steering
 SW : Switching magnet
 TPS : Terminal potential

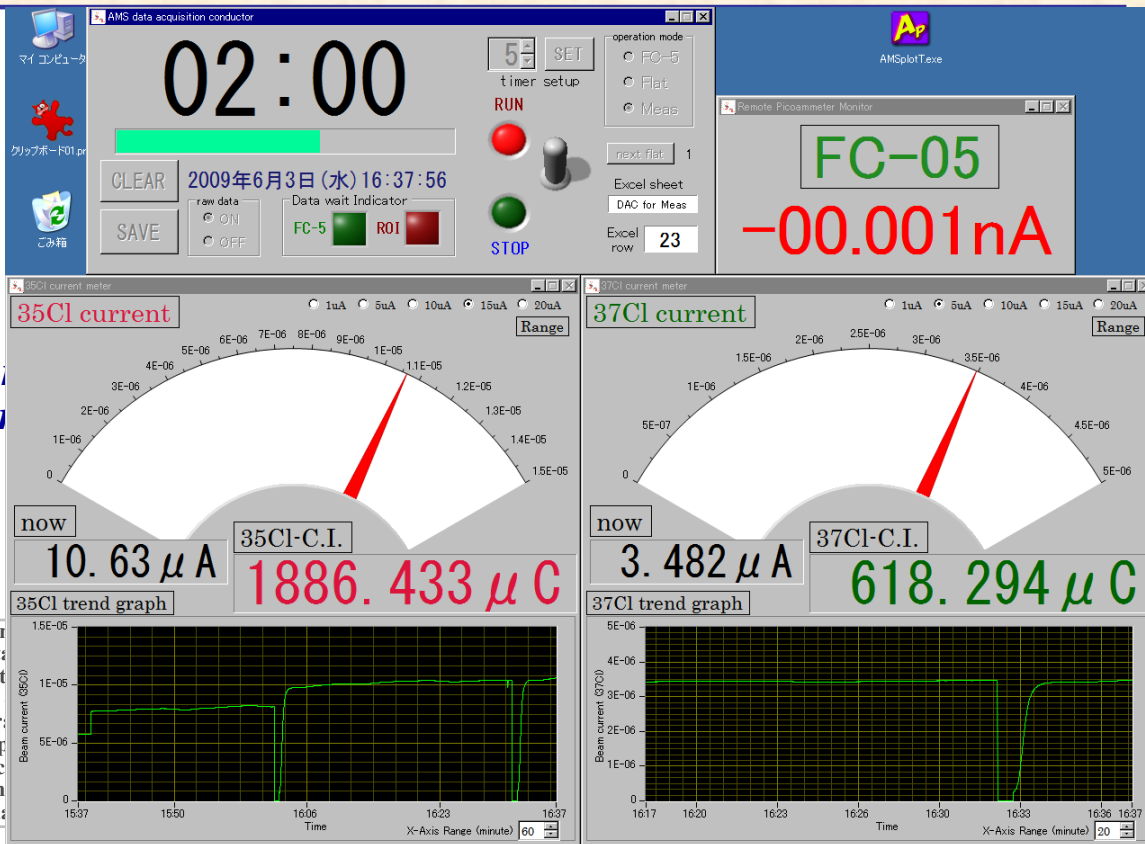
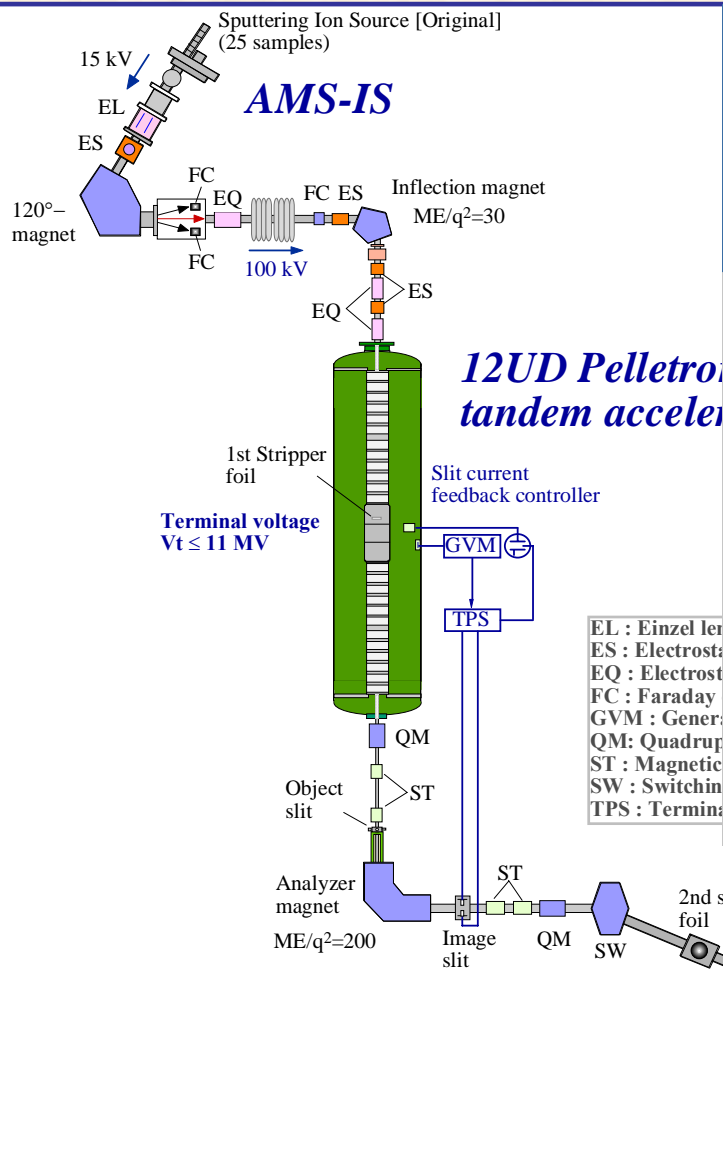
AMS Cs sputtering ion source



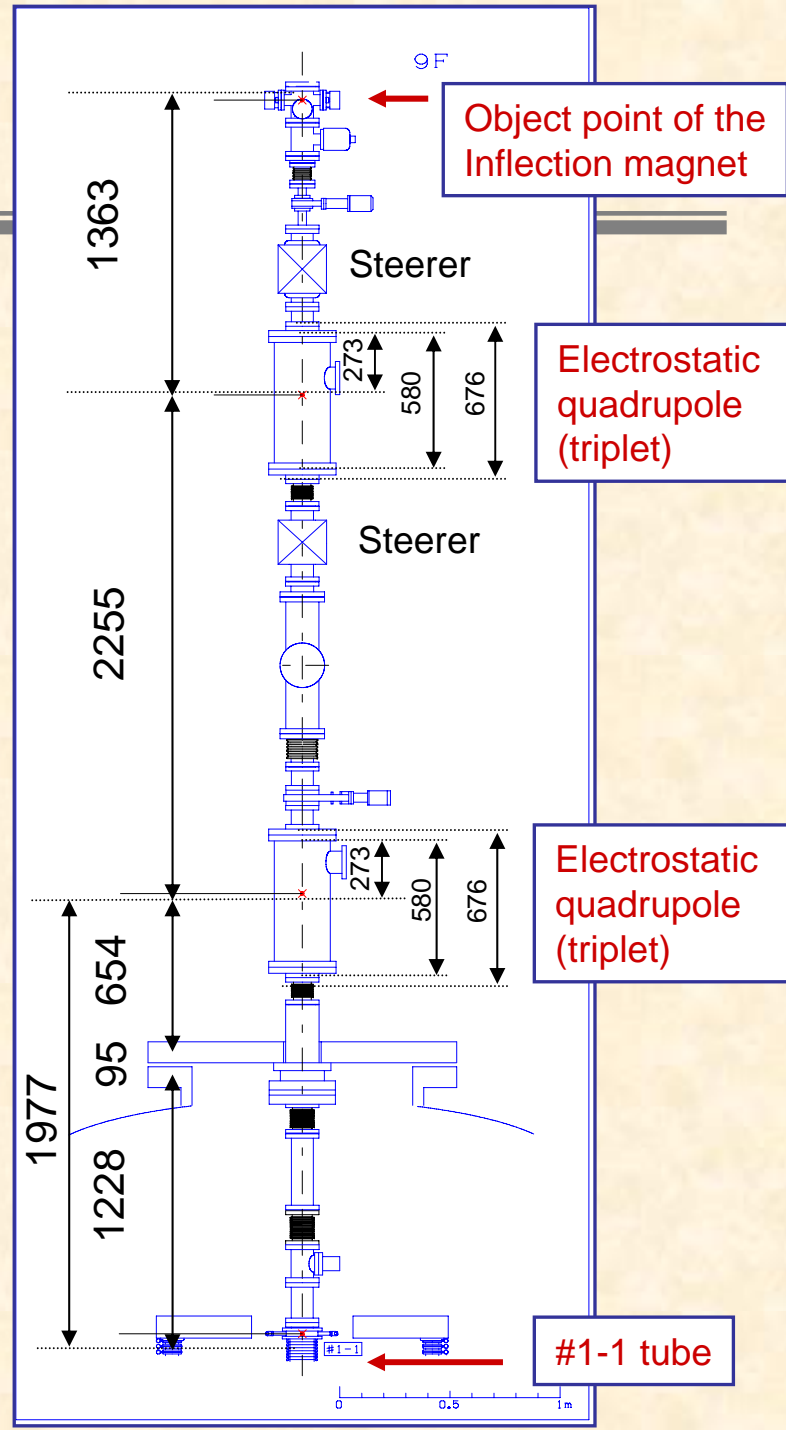
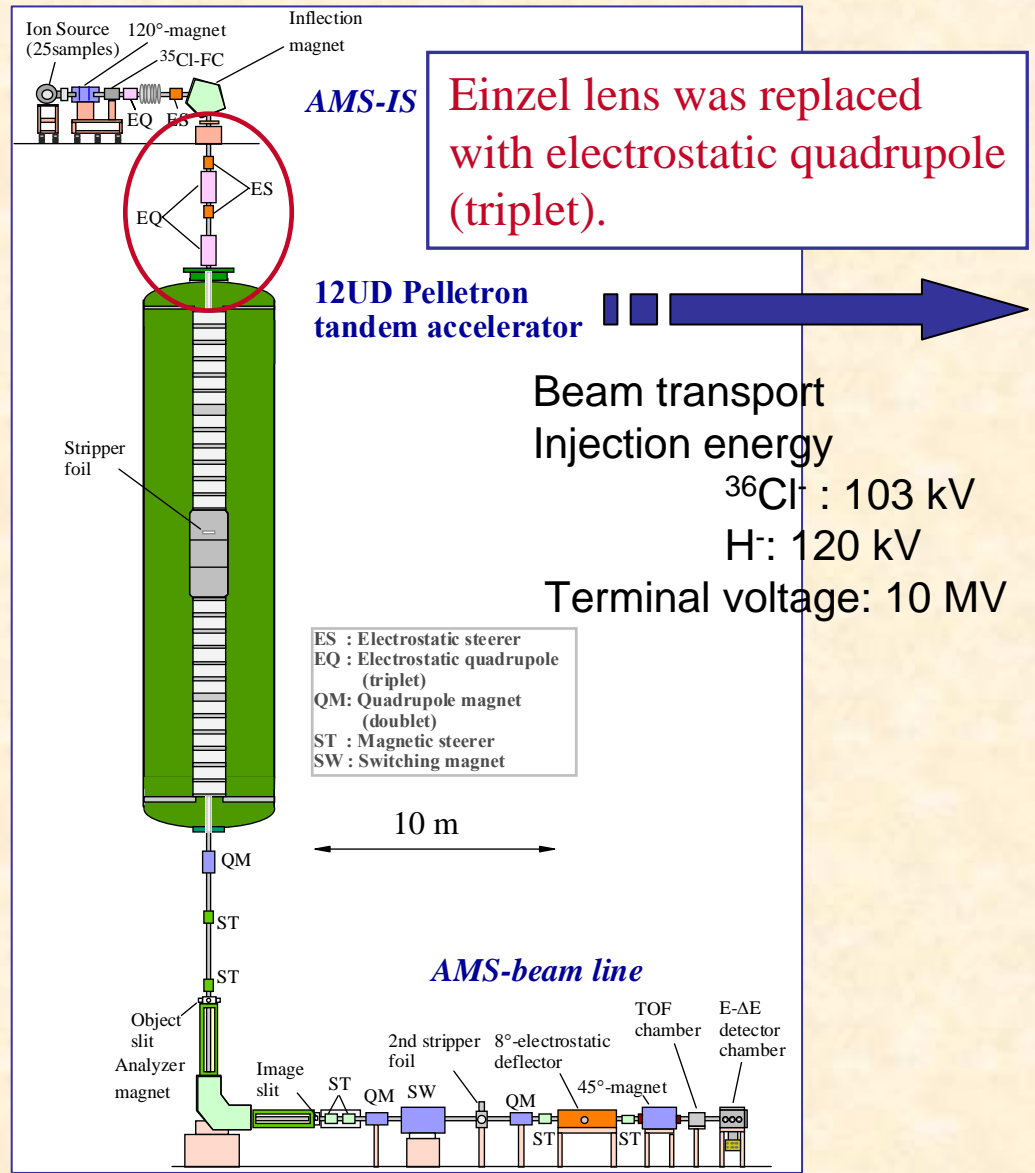
AMS Cs sputtering ion source



AMS Cs sputtering ion source



Upgrade of LEBT for 12UD Pelletron tandem accelerator

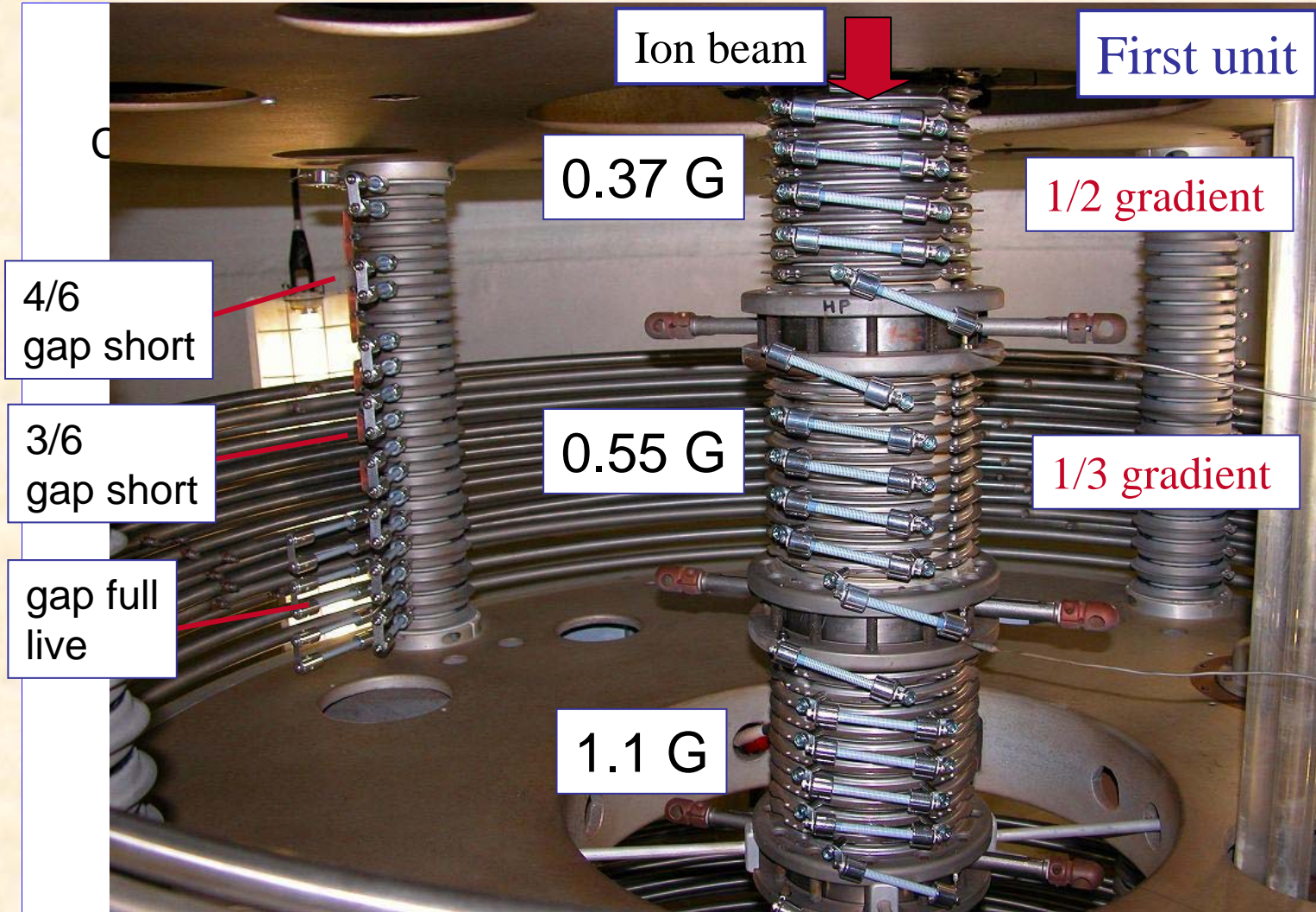


Tsukuba 12UD first unit (2009)

Lower resistance
at the entrance.



Adjustment for the focusing effect

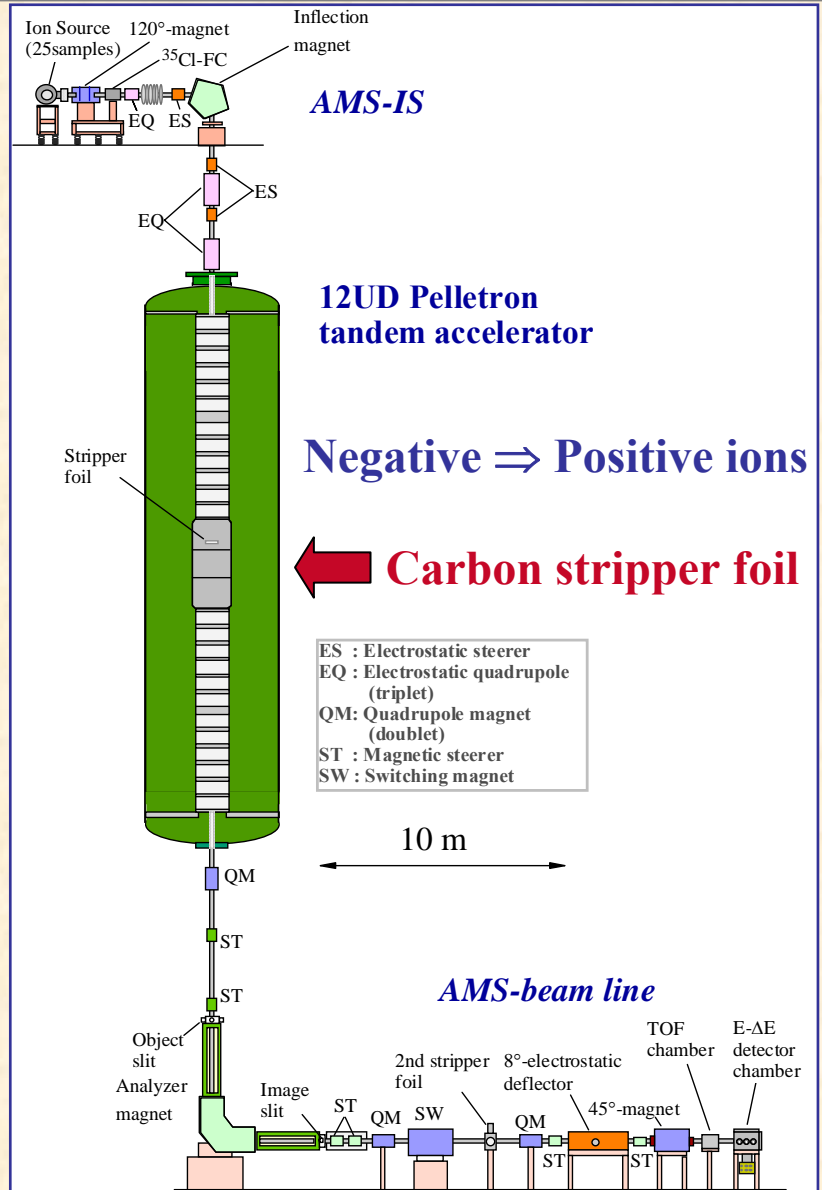


Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal ($\phi 20$) in 2004.



2 foil units

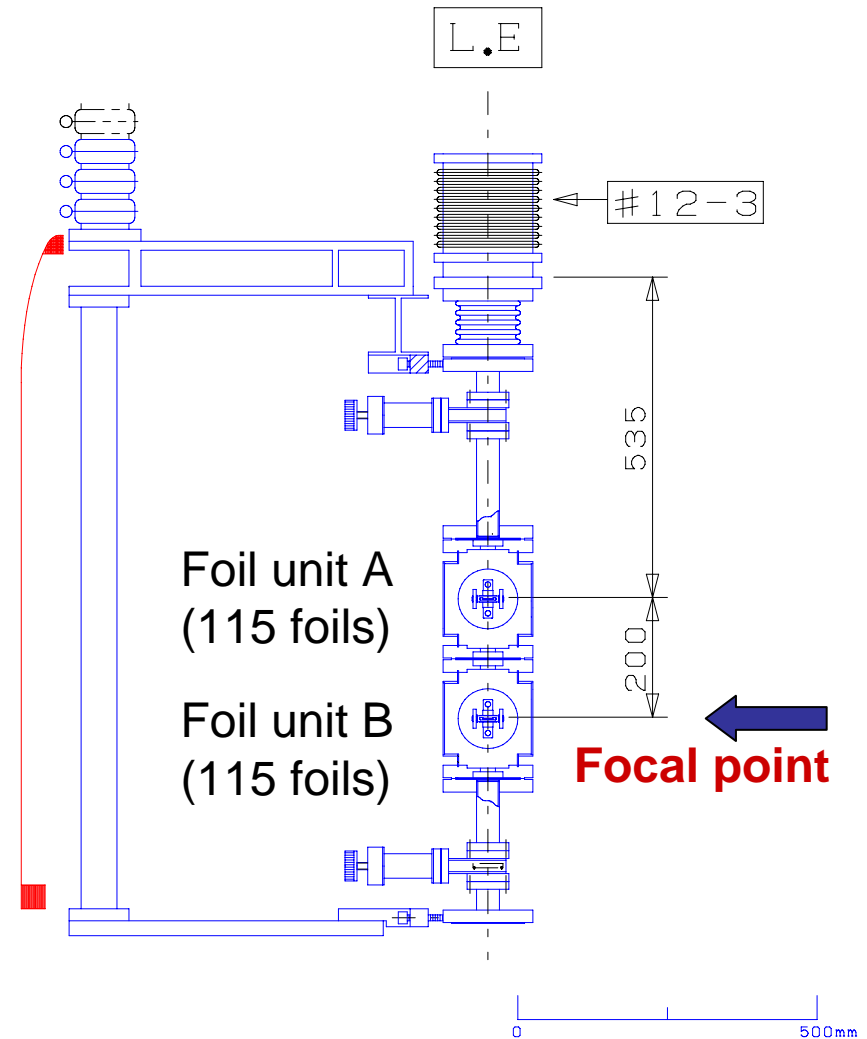


Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal ($\phi 20$) in 2004.

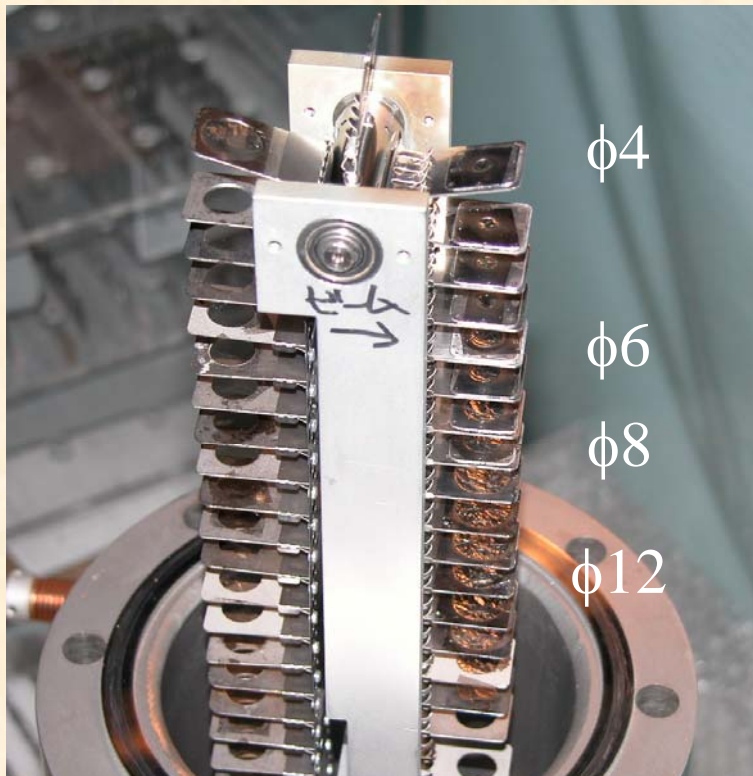


2 foil units

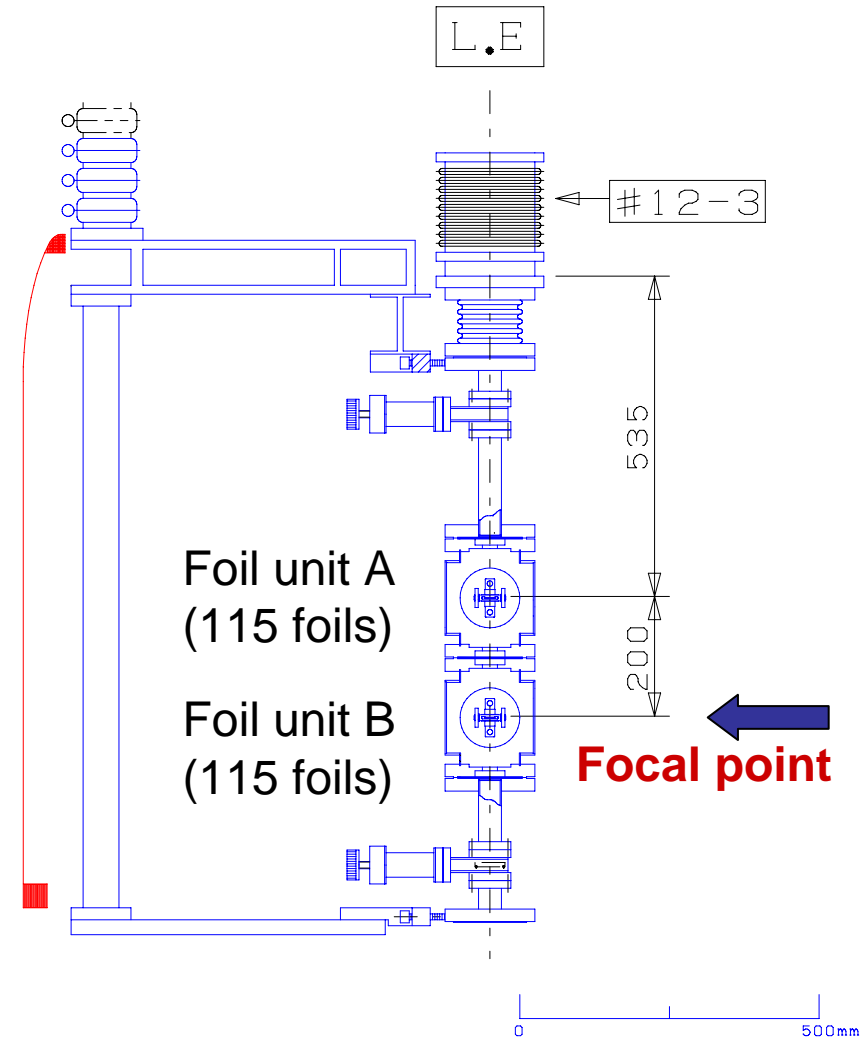


Terminal section (Charge exchange)

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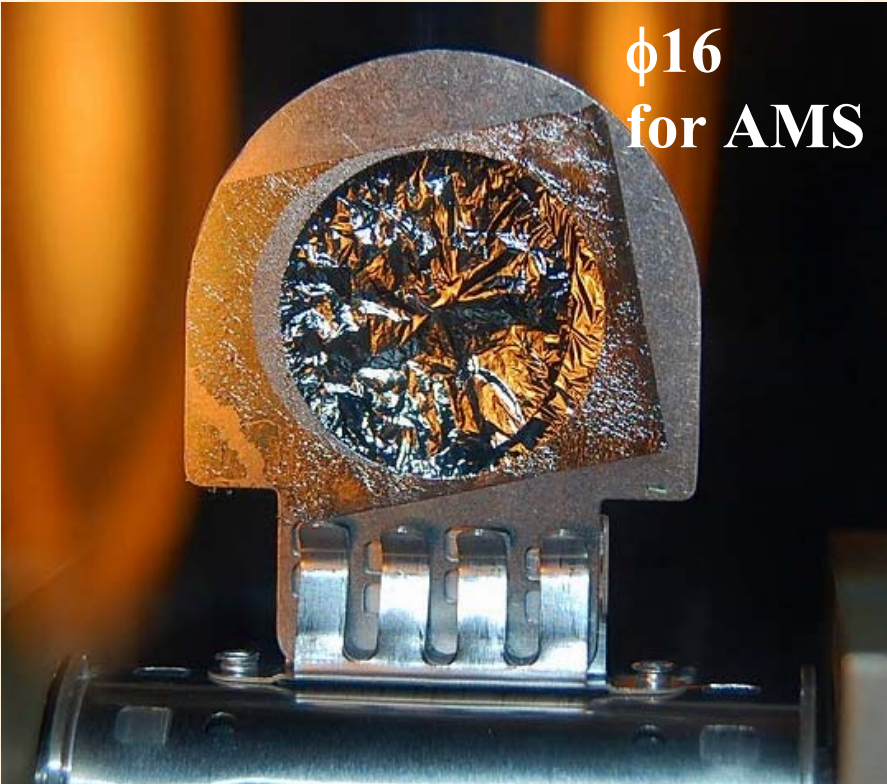


Carbon foil for AMS: $5 \mu\text{g}/\text{cm}^2$



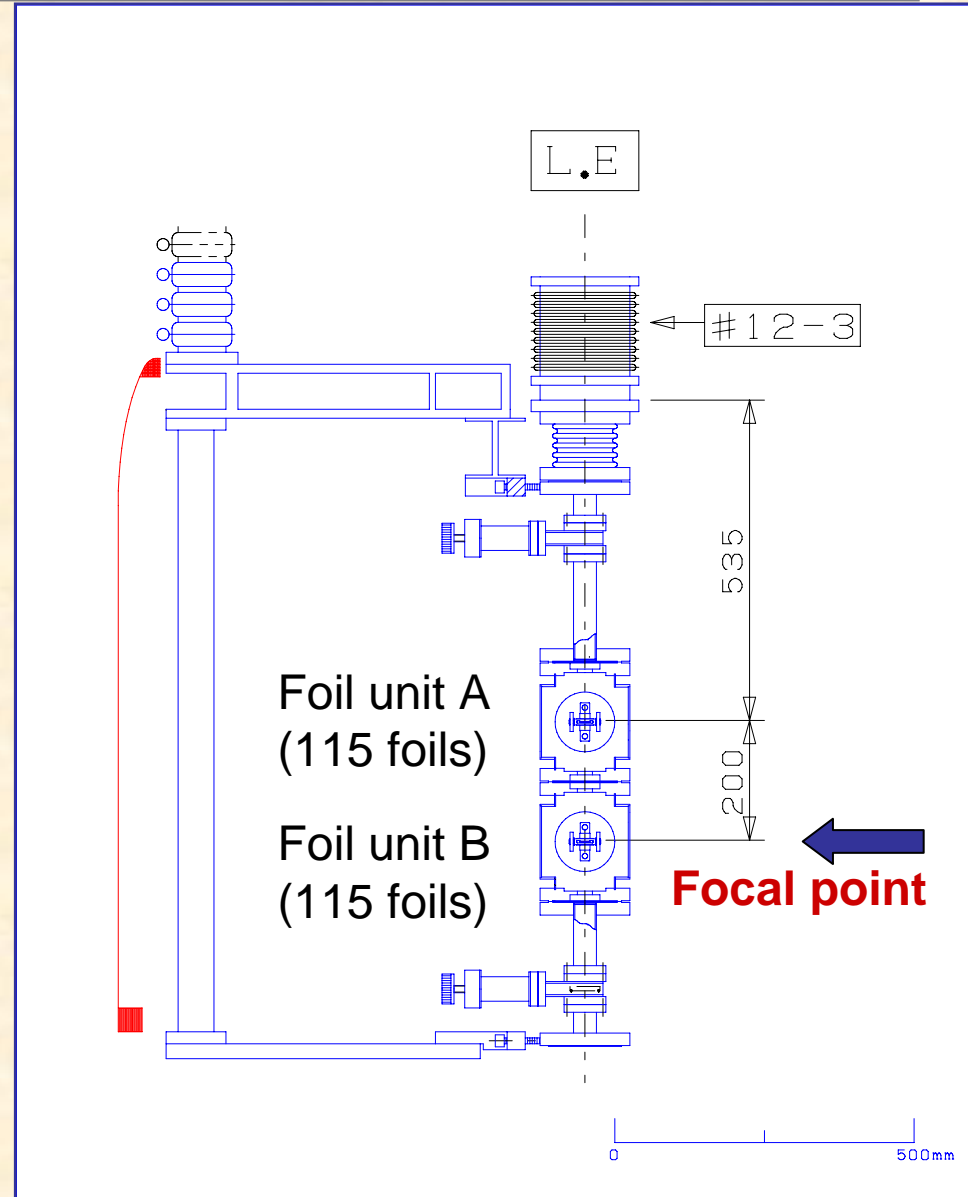
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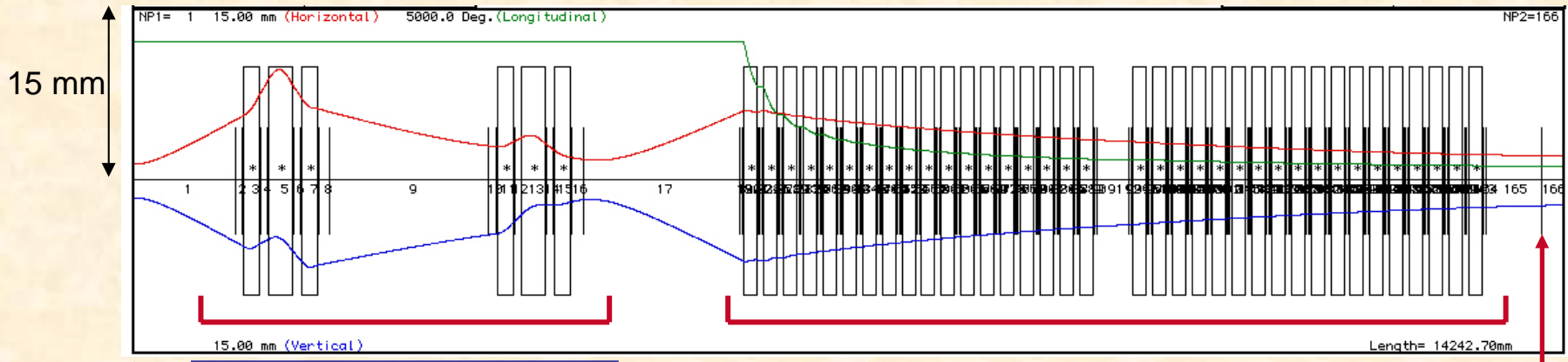
$\phi 16$
for AMS

Carbon foil for AMS: $5 \mu\text{g}/\text{cm}^2$



Beam transport of the 12UD Pelletron

2009



Electrostatic Q-triplet

12UD accelerator tubes

Terminal
Carbon stripper foil

Magnification: 1.5 times
on the Terminal stripper.

Object point

Acceptance
~ 5 mm mrad

LEBT

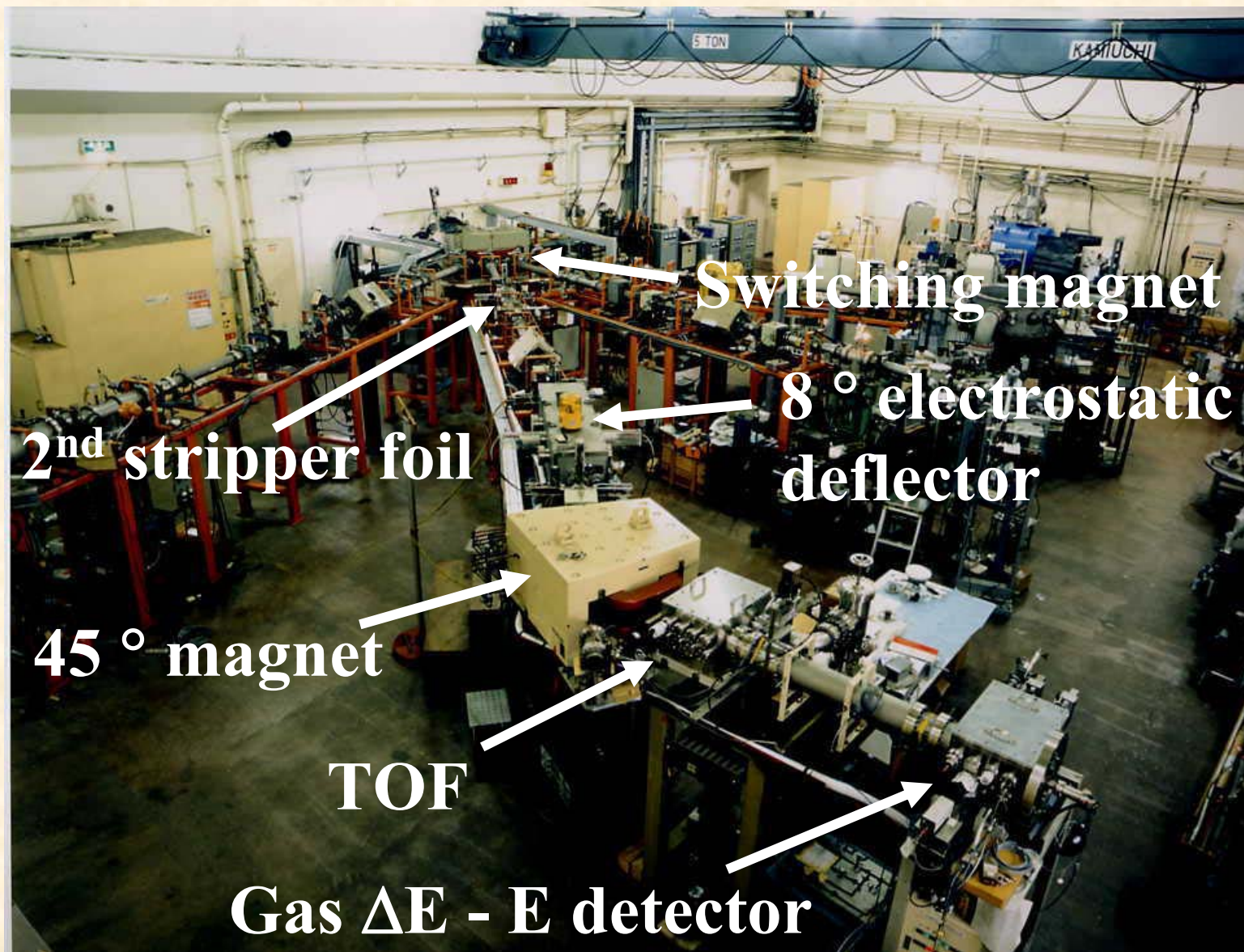
$^{36}\text{Cl}^-$

Injection energy: 103 kV

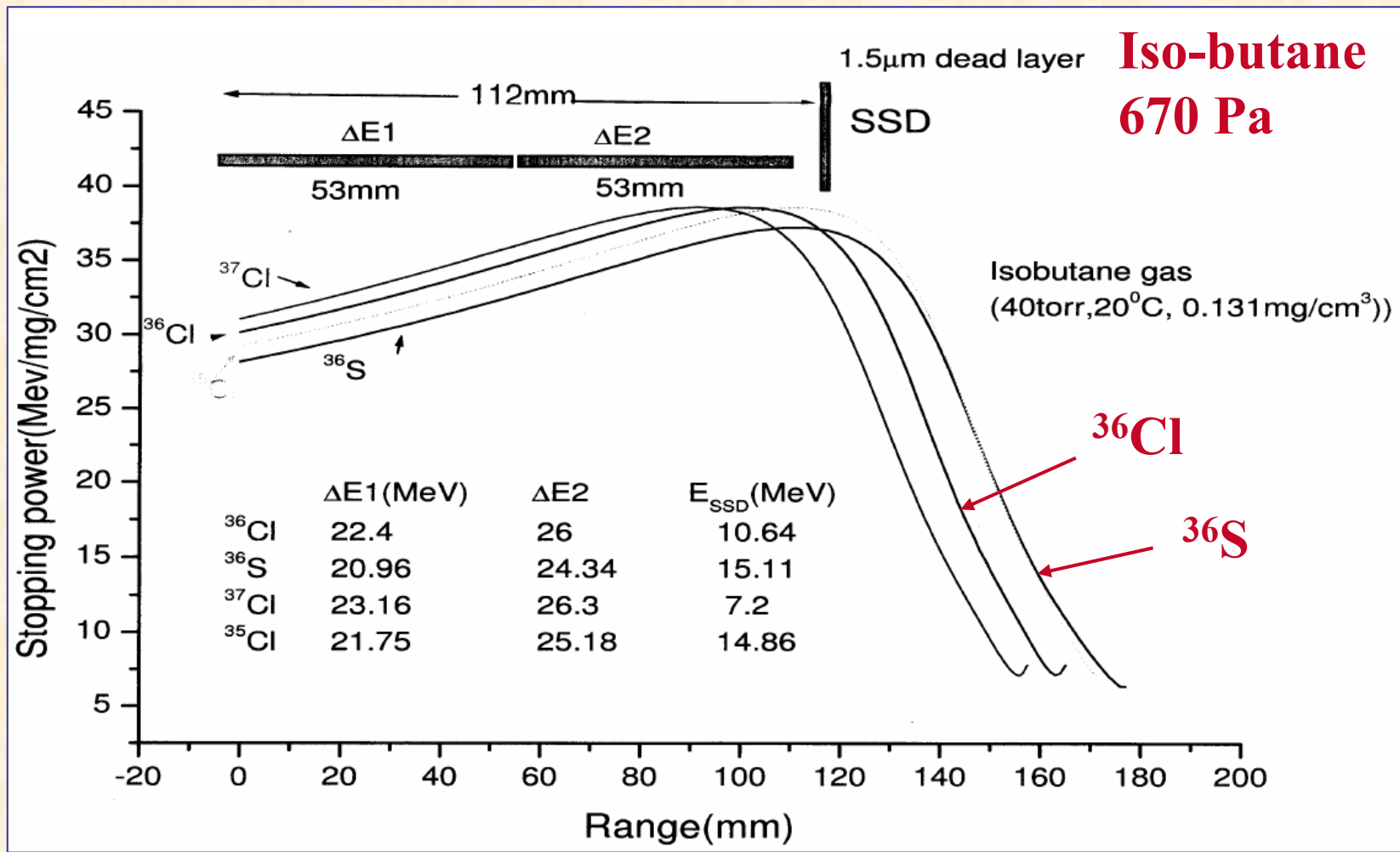
V_T : 10 MV

Beam spot on the stripper foil: ϕ 6 mm

Mass separator beam line

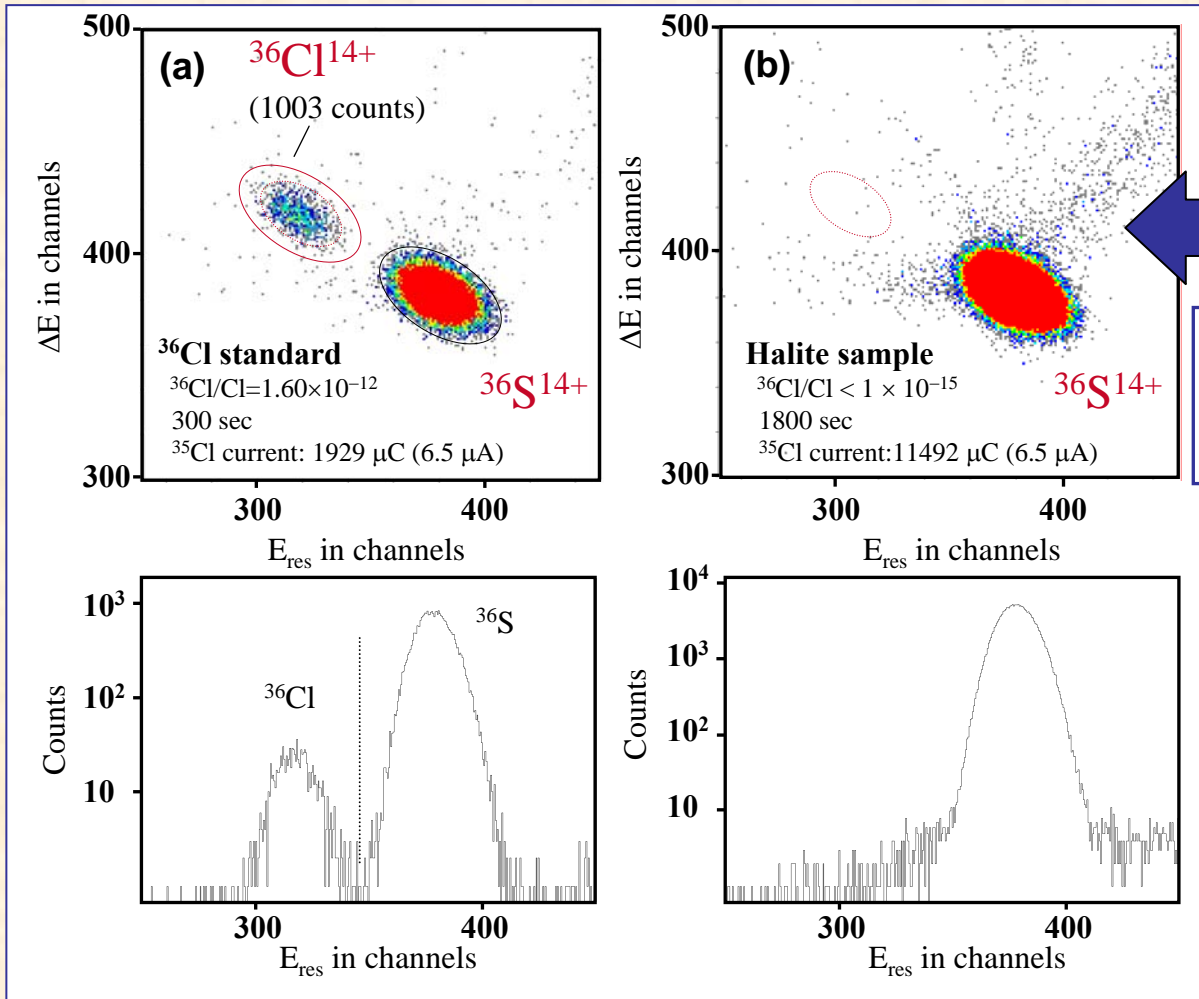


Gas E- ΔE detector



100 MeV ^{36}Cl in the gas detector.

^{36}Cl -AMS (2-dimensional spectrum)



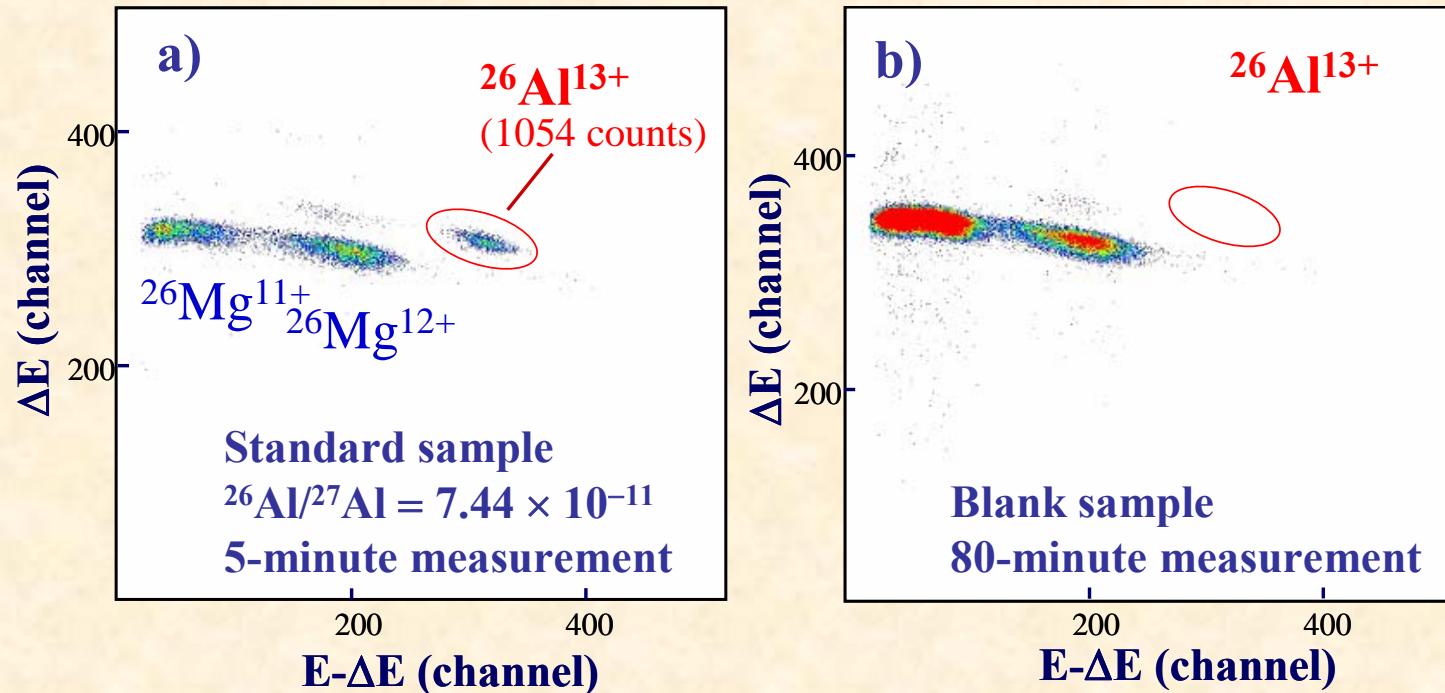
$^{36}\text{Cl}/\text{Cl}$
Background: $\sim 1 \times 10^{-15}$

Standard sample

$^{36}\text{Cl}/\text{Cl} = 1.60 \times 10^{-12}$

Blank sample

^{26}Al -AMS (2-dimensional spectrum)



- Full stripping technique

- Pilot beam: $^{26}\text{MgO}^-$

- Beam current of AlO^- from Al_2O_3 sample : $\sim 1.5 \mu\text{A}$

- ^{26}Al is very clearly separated from ^{26}Mg .

- Background of the ^{26}Al -AMS: $< 1 \times 10^{-15}$.

Performance of the Tsukuba AMS system

A pilot beam method is used to stabilize the terminal voltage.

²⁶Al-AMS

Target material	V_T	Injection ion	Pilot beam	Detection ion	Particle energy	Back-ground
$\text{Al}_2\text{O}_3 + ^{26}\text{MgO}_2 + \text{Ag}$	10.2 MV	$^{26}\text{AlO}^-$	$^{26}\text{MgO}^-$	$^{26}\text{Al}^{13+}$	78 MeV	$< 1 \times 10^{-15}$

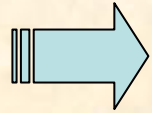
³⁶Cl-AMS

Target material	V_T	Injection ion	Pilot beam	Detection ion	Particle energy	Back-ground
$\text{AgCl} + \text{C}_{60}$	10 MV	$^{36}\text{Cl}^-$	$^{12}\text{C}_3^-$	$^{36}\text{Cl}^{14+}$	100 MeV	$< 1 \times 10^{-15}$

¹²⁹I-AMS

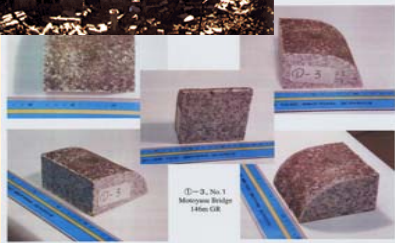
Target material	V_T	Injection ion	Pilot beam	Detection ion	Particle energy	Back-ground
$\text{AgI} + \text{MoO}_2 + \text{Nb}$	9.7 MV	$^{129}\text{I}^-$	$^{97}\text{MoO}_2^-$	$^{129}\text{I}^{26+}$	126 MeV	$< 1 \times 10^{-13}$

Applications by the Tsukuba AMS system



Mainly for earth and environmental sciences.

**Nuclear safety research
Atomic bomb, neutron fluence**



Hiroshima A-bomb sample

**Soil
sediment**



Soil

**Rock
meteorite**



Limestone



Meteorite

**Groundwater, rain,
ice**



Rain water



Ice core

Biological sample



Human hair

Summary and future plans

- **12UD Pelletron tandem at the University of Tsukuba**

We have upgraded the 12UD Pelletron tandem.

⇒ LEBT, divided resister system, terminal stripper.

The beam time for AMS research has increased to about 42% of the total operation time.

- **Tsukuba AMS system**

We are able to measure long-lived radioisotopes of ^{26}Al , ^{36}Cl and ^{129}I by employing a molecular pilot beam method that stabilize the terminal voltage with 0.1% accuracy.

⇒ Main research fields are earth and environmental sciences.

Future plans

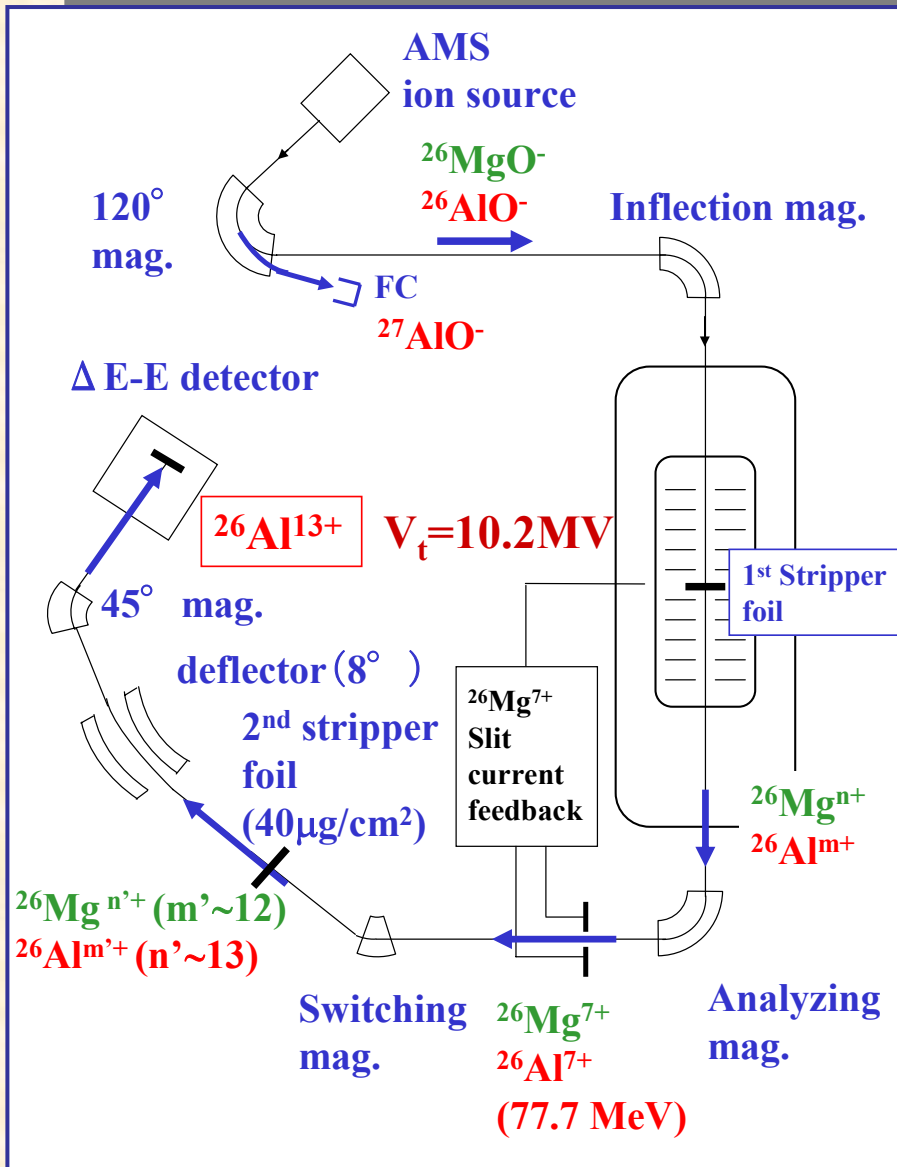
- **GVM control system**
- **New injection beam line (MC-SNICS)**

Thank you for your kind attention.



Appendix

^{26}Al -AMS



- Full stripping technique:
 $^{26}\text{Al}^{13+}/^{27}\text{AlO}^-$ [counts/ μC]
- Pilot beam: $^{26}\text{MgO}^-$
- Beam current of AlO^-
from Al_2O_3 sample : $\sim 1.5\mu\text{A}$

The beam transmission of full stripped Al^{13+} : 10%.

AMS facilities in JAPAN

10 AMS facilities (12 accelerators)

Paleo Labo Co., Ltd.
Tandem (500 kV): ^{14}C

JAEA Tono
Tandem (5MV): ^{14}C

Kyoto University
Tandem (8MV): ^{14}C

Kyushu University
Tandem (10MV): ^{14}C

Nagoya University
Tandetron (3MV): ^{14}C
Tandetron (2.5MV): ^{14}C

JAEA Mutsu
Tandetron (3MV): ^{14}C , ^{129}I

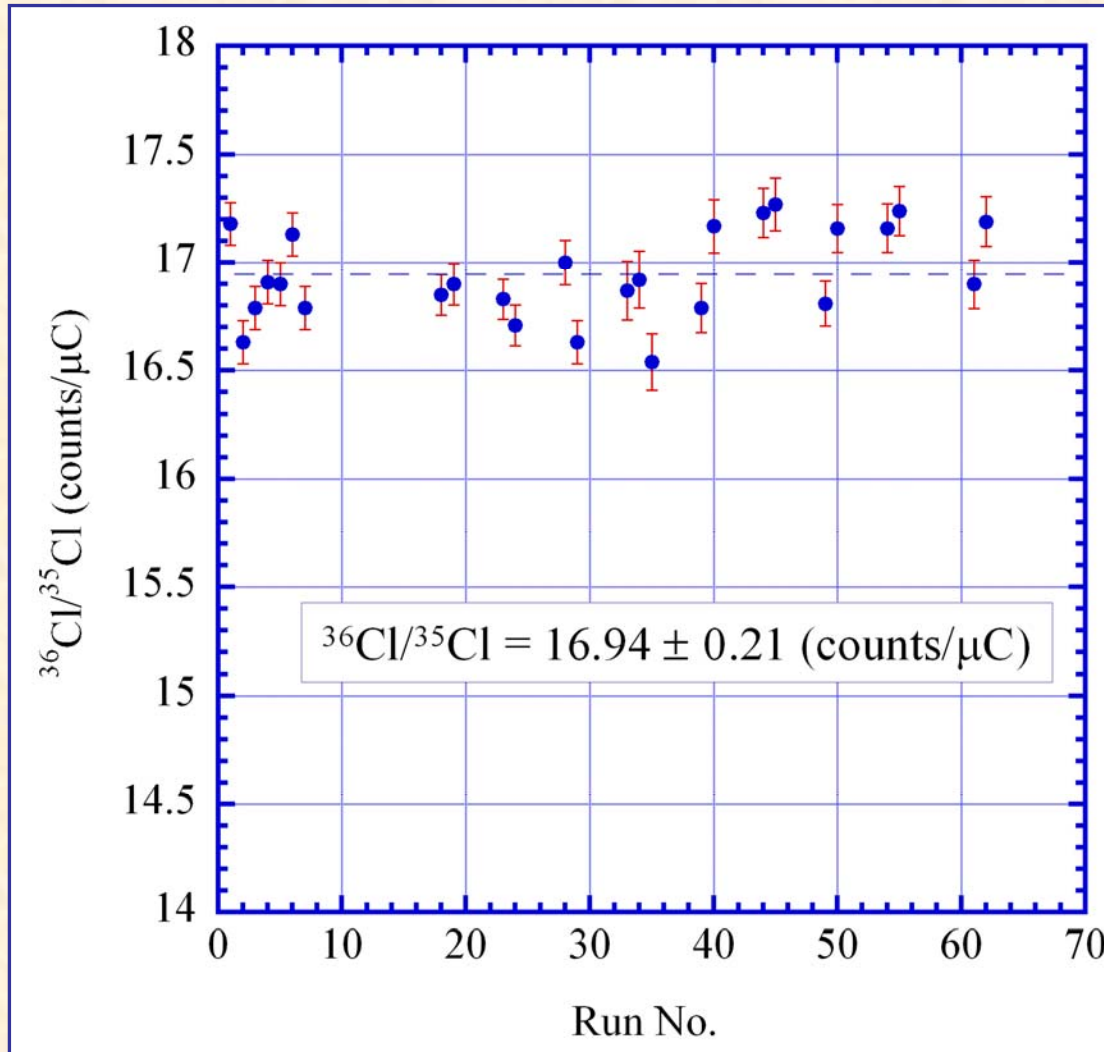
Institute of Acceleratory
Analysis Ltd.
Tandem (3MV): ^{14}C
Tandem (500kV): ^{14}C

NIES-TERRA
Tandem (5MV): ^{14}C

University of Tsukuba
Tandem (12MV): ^{26}Al , ^{36}Cl , ^{129}I

MALT, The University of Tokyo
Tandem (5MV): ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{129}I

Replicated ^{36}Cl standard measurements *UTTAC*



Standard sample:

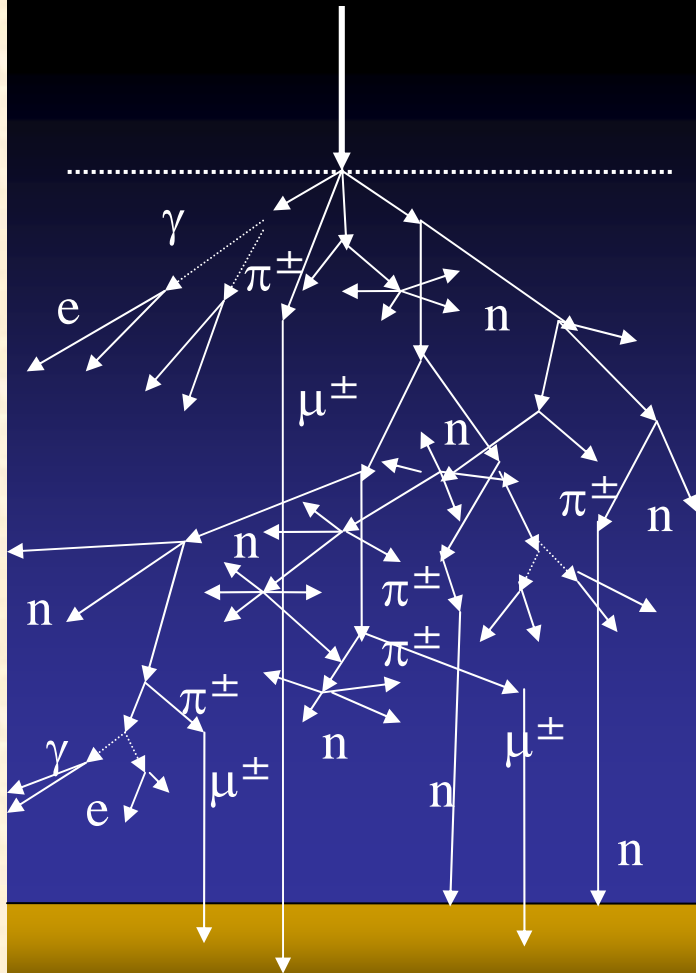
$$^{36}\text{Cl}/^{35}\text{Cl} = 5.90 \times 10^{-11}$$

It takes 3 minutes for each measurement.

The standard deviation of the fluctuation of the $^{36}\text{Cl}/^{35}\text{Cl}$ ratio is kept within $\pm 3\%$.

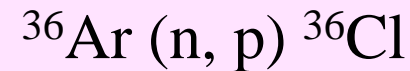
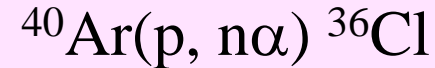
^{36}Cl production

Primary Cosmic Ray



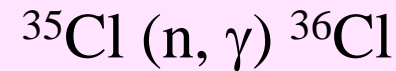
^{36}Cl ($T_{1/2}=301$ kyr): β -decay ^{36}Ar (98 %)

Natural — Cosmic rays



~ 20 atoms/ m^2/s

Neutron capture (U, Th)



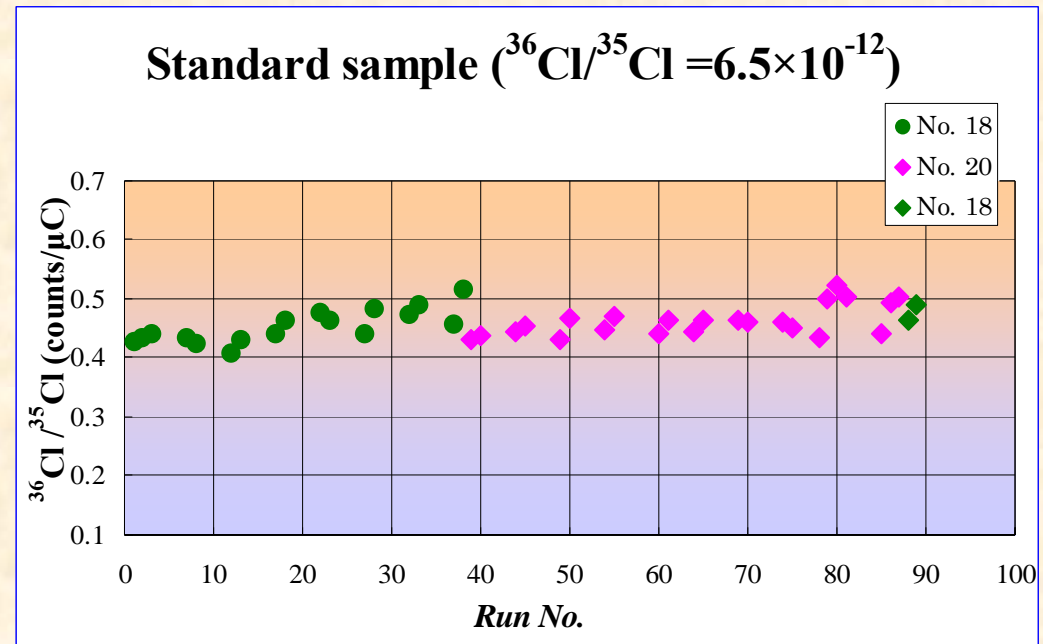
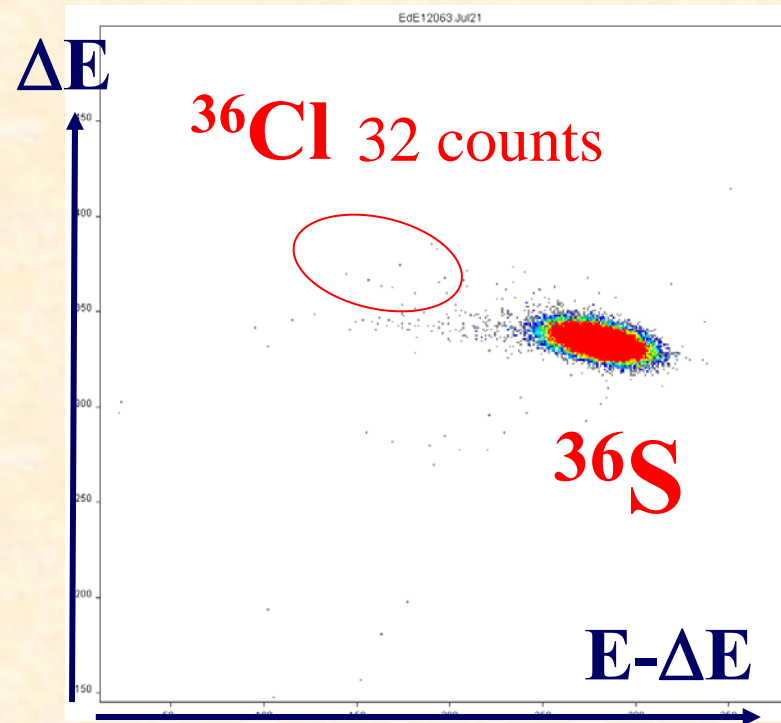
Human activities



Nuclear explosion tests in the sea
(1952-1958)

Atomic power plant,
Nuclear fuel cycle facilities, etc.

^{36}Cl -AMS



Reproduction in replicated measurements

^{36}Cl -AMS (Vt:10MV)

Beam energy: $^{36}\text{Cl}^{14+}$ 100MeV

Background: $^{36}\text{Cl}/\text{Cl} = \sim 1 \times 10^{-15}$

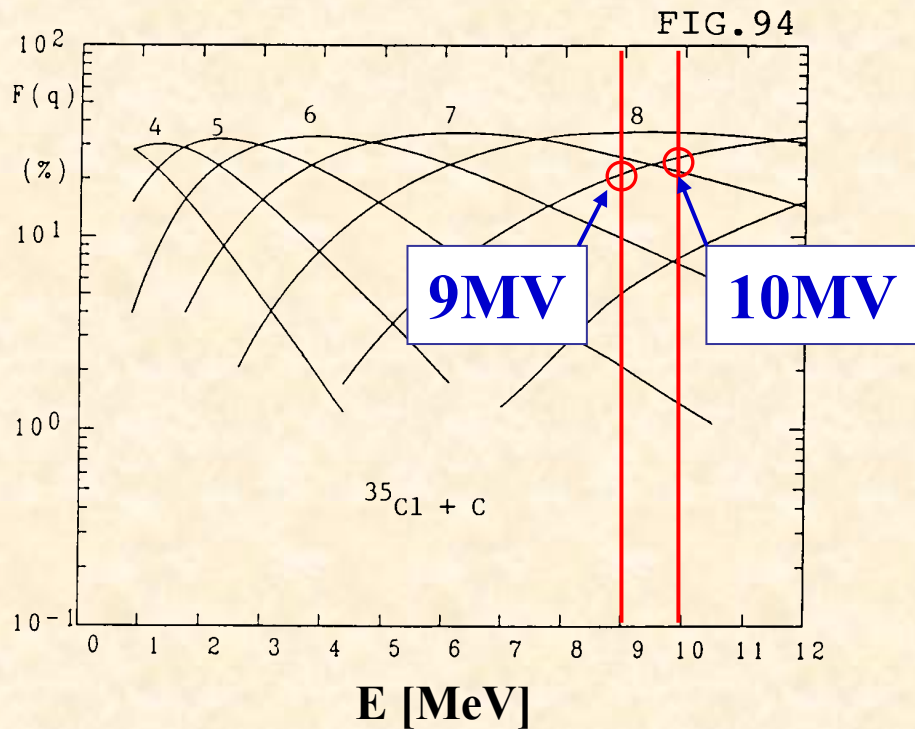
Detection range: $^{36}\text{Cl}/\text{Cl} = 10^{-10} \sim 10^{-14}$

Accuracy: $\pm 3\%$ ($^{36}\text{Cl}/\text{Cl} \sim 10^{-12}$)

Charge state distribution of Cl ions

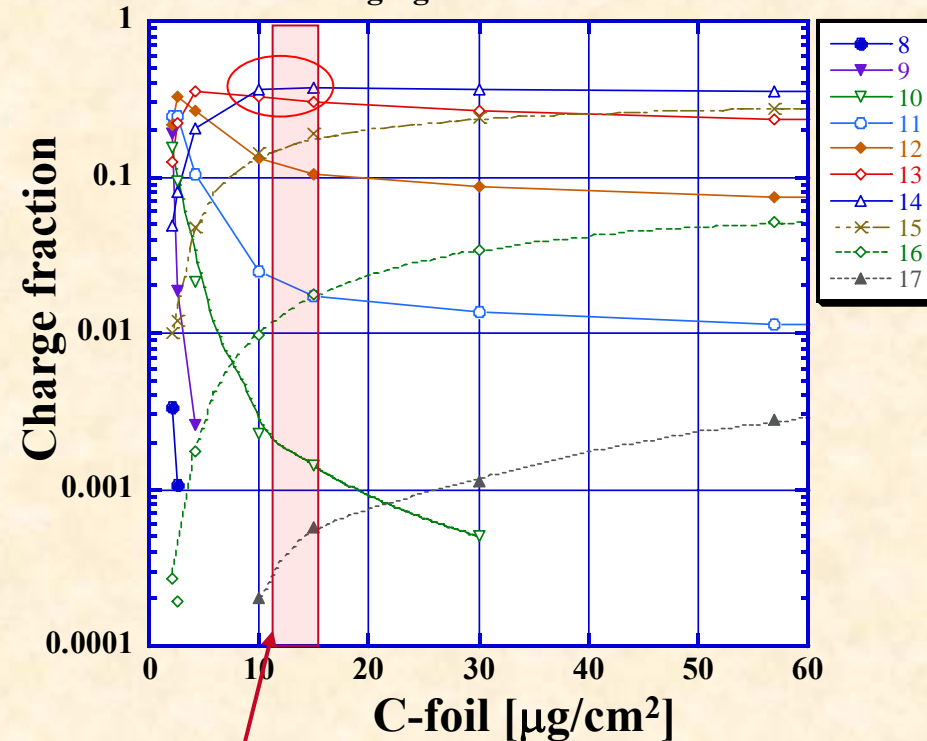
Charge state distribution of ^{35}Cl and ^{37}Cl ions after the passage through a carbon foil.
[by Dr. Shima et al.]

1st stripper foil



2nd stripper foil

Charge fraction $F(q)$ of $90\text{MeV } ^{35}\text{Cl}^{9+}$ emerging from a carbon foil



Charge fraction $F(q)$ of $^{35}\text{Cl}^{9+}$ increased 1.5 times from 20% to 30%.

The optimum thickness of 2nd stripper foil: $12\text{-}15\mu\text{g}/\text{cm}^2$ [$F(14+)$]

Sample preparation

Sample (10–20 mL)
← 30% H₂O₂ (1 mL)
← 13 M HNO₃ (1 mL)
← 0.3 M AgNO₃ (1 mL)
Centrifugation (2500 rpm, 10 min)

Supernatant Precipitate (AgCl)

Sulfur Reduction (2 times)

← 3 M NH₄OH (2 mL)
← sat. Ba(NO₃)₂ (3 mL)

Filtration (Filter Paper)

Filtrate ([Ag(NH₃)₂]⁺ Cl⁻)

Precipitate (BaSO₄)

← 13 M HNO₃ (0.5 mL)

Centrifugation (2000 rpm, 15 min)

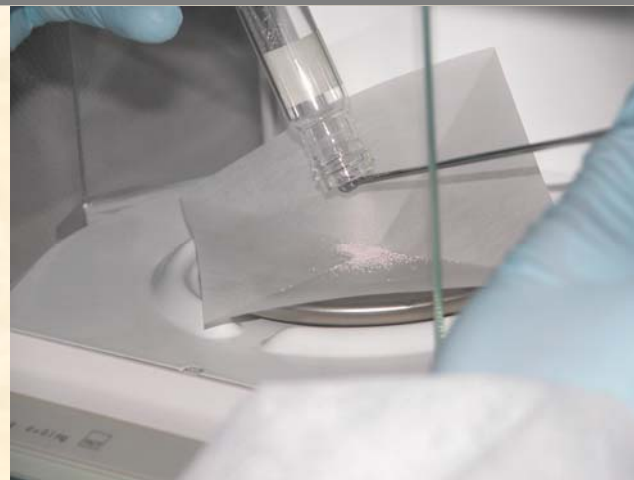
Supernatant

Precipitate (AgCl)

Washing (with 0.01 M HNO₃, 2 times)

Washing (with 99.5% C₂H₅OH)

Drying (130°C, 3 h)



Sample: AgCl 1~5mg



A benzene saturated solution of fullerene (C₆₀) for ¹²C₃⁻ pilot beam. [7.6 μl / AgCl: 1 mg]