Measurement of Radio-nuclides in Radioactive aerosols produced in a 120-GeV Proton Target Station

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JASMIN Collaboration (Japanese-American Study of Muon Interactions and Neutron detection) project
T-993, T-994

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-Outline-

1. Introduction "Radioactive aerosols in accelerator target room"
2. Motivation "Why we focus on radio-nuclides and P-32"
3. Experimental procedure "Aerosol-sampling, Measurement of P-32"
4. Results "Activity levels of P-32 and other radio-nuclides"
5. Discussion "Dose of P-32 and other radionuclides, Source materials"
6. Summary

JASMIN Collaboration (**J**apanese-**A**merican **S**tudy of **M**uon **I**nteractions and **N**eutron detection) project
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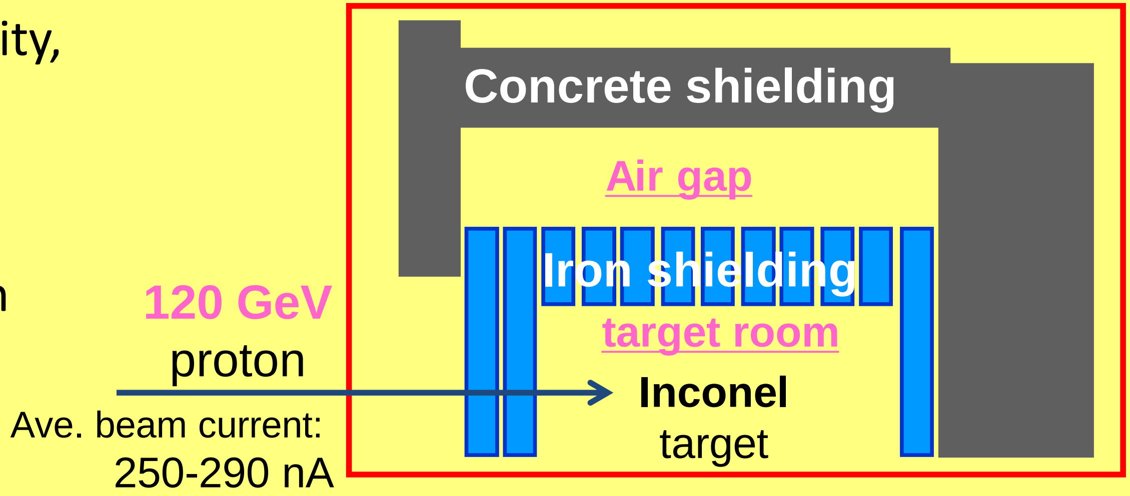
Introduction

In target area of accelerator facility,
"**<u>High-energy proton beams"</u>**
travel in air, and
<u>"beam sprays"</u>
from the interaction of the beam
with the target
and related target
station components also
travel in air.



Anti-proton target station (AP0) @Fermilab

Radioactive aerosols are produced by
-**direct activation** by the beam and its secondaries
-the subsequent **attachment of the various radio-nuclides** produced by other nuclear **spallation** reactions <u>in the **target**</u>, <u>in the **instruments**</u> around the target, <u>**air**</u> and <u>other sources</u>.

In terms **of radiation control**,
it is important to know **the radio-nuclide composition** of
the **radioactive aerosols** in an accelerator target area.

Introduction

In target area of accelerator facility,
"**<u>High-energy proton beams</u>**"
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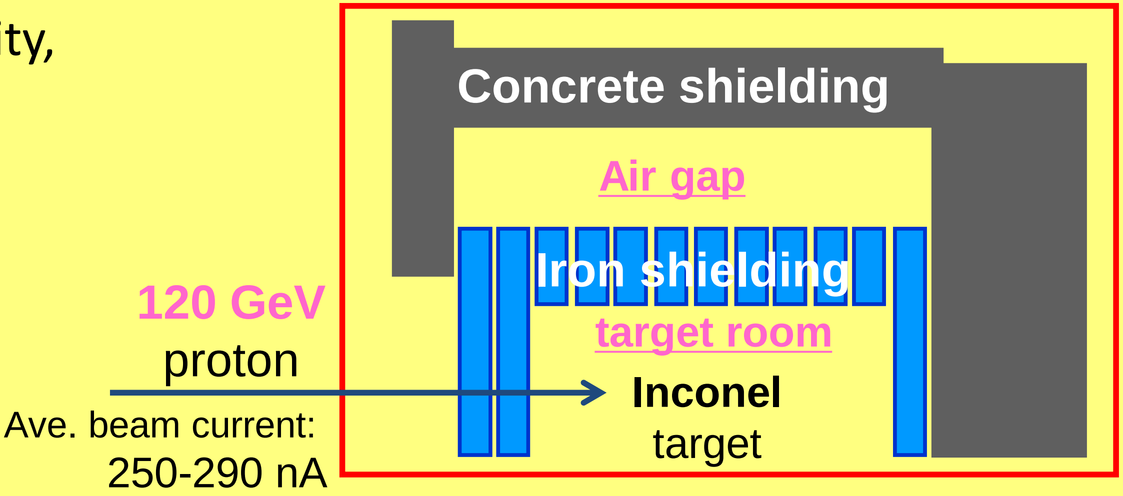


Anti-proton target station (AP0) @Fermilab

Motivation -1-

Especially, in **ultra high energy** proton target station,
the **radio-nuclide composition** of radioactive aerosols
and their **source materials** have scarcely been studied previously.

In order to control external and internal exposure of the workers in a high-energy accelerator target area, it is important to measure the produced **amount of radio-nuclides** in the **radioactive aerosols**.

Motivation -2- (Why we focus on P-32)

The pure beta emitters such as **P-32** as well as **gamma rays emitting nuclides** are also important.

^{32}P ($T_{1/2}$ = 14.26 d, no-γ)

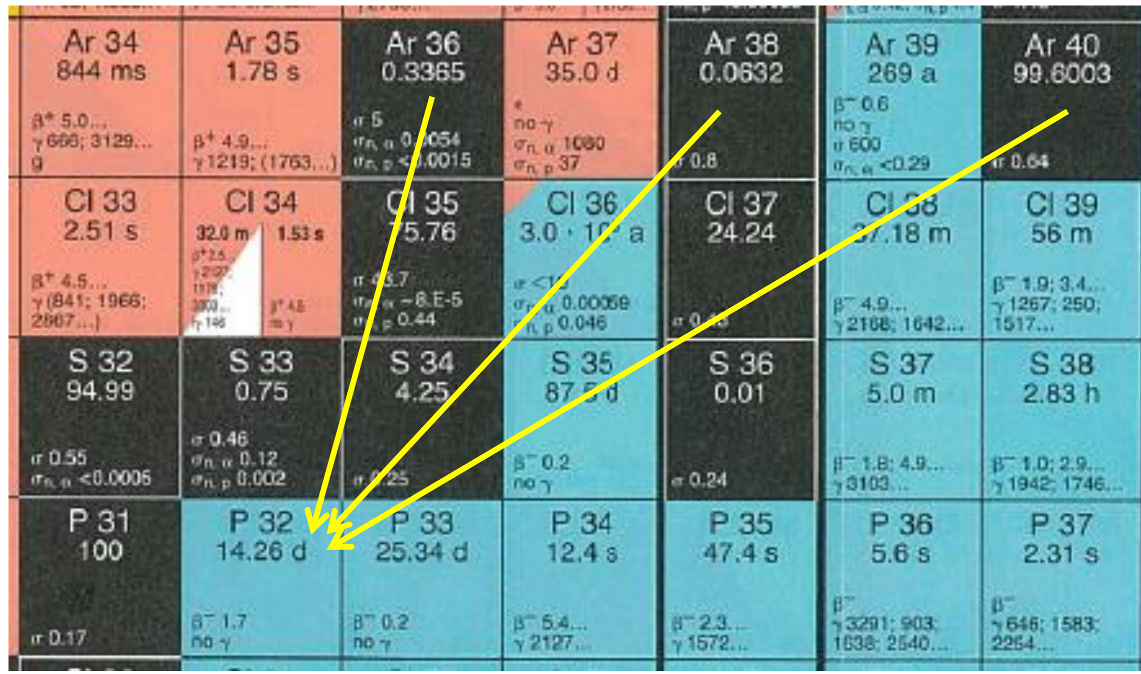
-**pure beta emitter**

→cannot be determined by **conventional method** used in radiation control

-**from argon (Ar) in air**

→ close to Ar

→ P-32 from Ar >> trace



In spite of **target materials**, it is essential to determine **activity levels of ^{32}P** for the radiation control purposes, especially for evaluating the internal exposure of the workers.

Production of ^{32}P in target area have not been studied previously,
because of difficulties in detecting and characterizing pure beta-emitters compared to gamma-ray emitters.

Motivation -2- (Why we focus on P-32)

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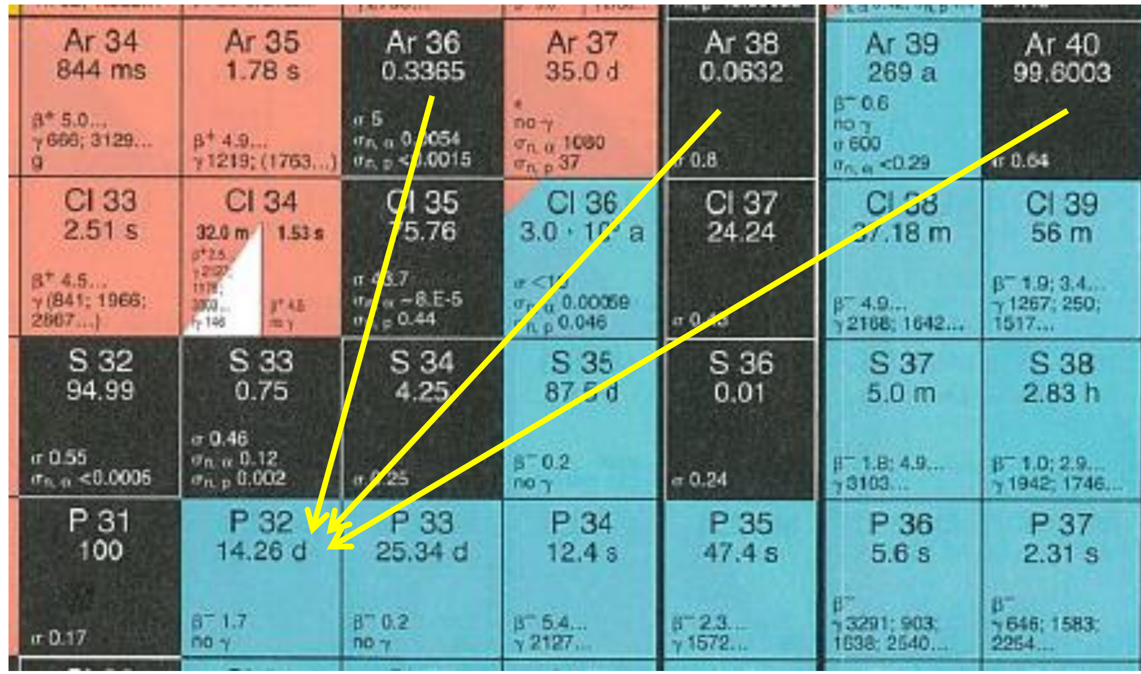


This work

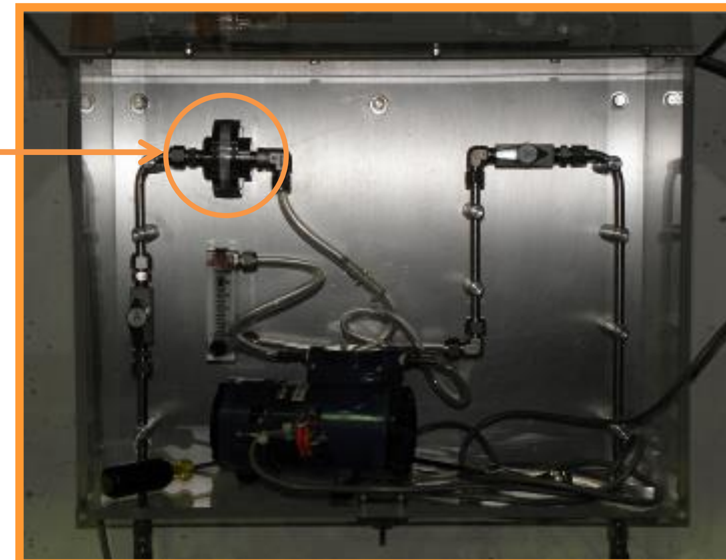
What kinds of radionuclides were produced in Anti-proton target station?

Measuring the beta emitting radio-nuclide **^{32}P**

Determination of **the activity levels** # Estimation of internal dose

Discussion on their **source material**

Experimental (Aerosol-sampling)



The radioactive aerosols, (which were produced from air, target material, or the other instruments,) were withdrawn by a pump and collected on **filter paper**.

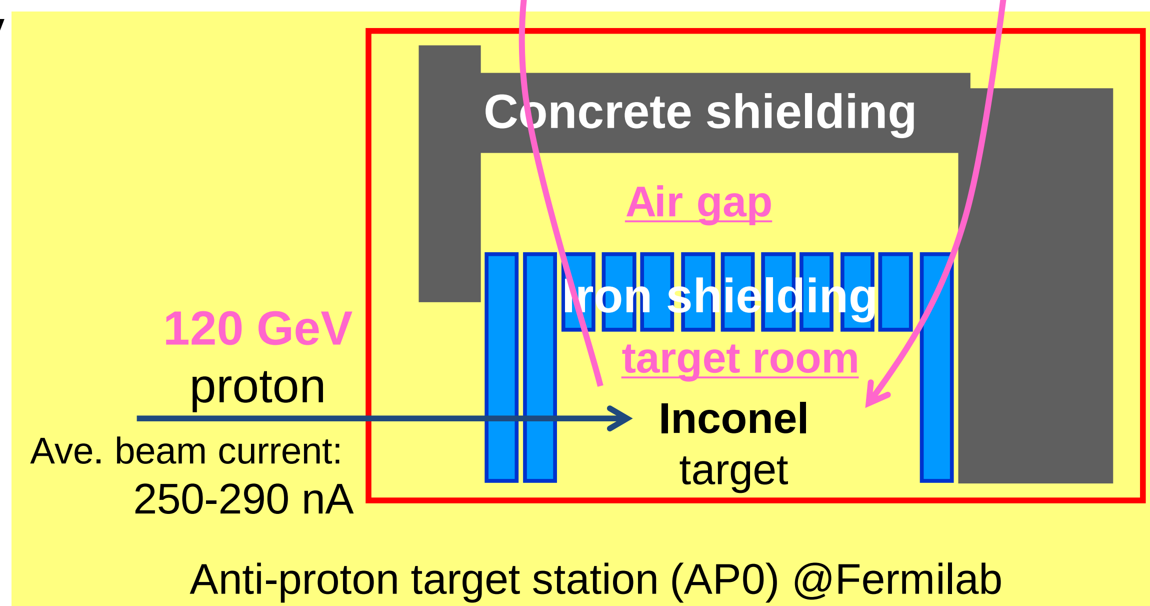
↓

gamma-ray spectrometry

↓

chemical treatment (to separate P)

Sampling condition:
9.5 L of air / min
26 h-sampling
(≈ 15 m^3 of air in total)



Experimental (Chemical treatment to separate P& LSC)

The phosphorus compound $MgNH_4PO_4 \bullet 6H_2O$ was extracted from the filter paper by a chemical separation procedure*. (@Hot laboratory in KURRI)

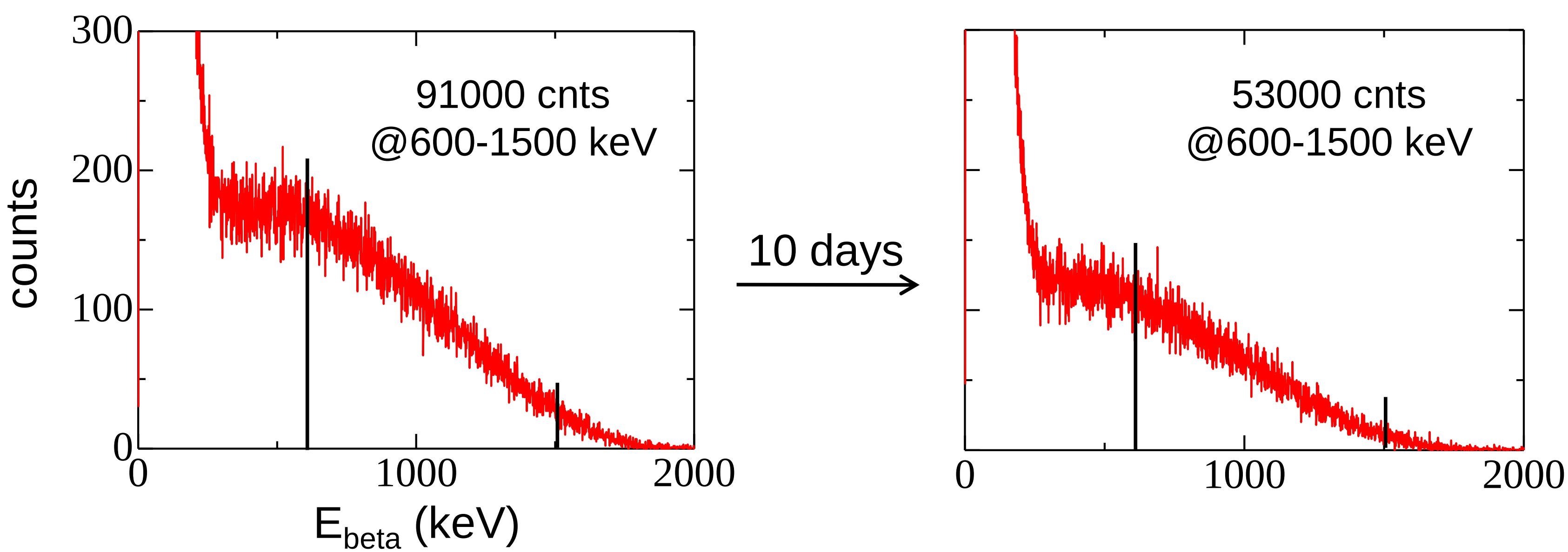
filter paper collecting radioactive aerosols

↓

$MgNH_4PO_4 \bullet 6H_2O$ (white solid of 20.4 mg)

Chemical yield: 51% (estimated from P-carrier)

Beta-ray spectra from liquid scintillation counter (LSC)



*W. T. Mullins and G. W. Leddicotte, “The Radiochemistry of Phosphorus” National Academy of Science (1962)

Results (At the time just after aerosol-sampling from target room)

Nuclide	half-life (d)	Activity (Bq)
P-32	14.26	26 ± 3
Be-7	53.29	880 ± 6
Na-22	950.095	2.0 ± 0.1
Sc-46	83.82	8.4 ± 0.2
V-48	15.97	31 ± 1
Cr-51	27.7	80 ± 3
Mn-52	5.6	34 ± 2
Mn-54	312.2	15 ± 1
Co-56	77.26	34 ± 1
Co-57	271.79	64 ± 1
Co-58	70.86	110 ± 2
Fe-59	44.503	4.4 ± 0.3
Co-60	1924.28	1.6 ± 0.1
Se-75	119.64	1.4 ± 0.1
Y-88	106.6	0.44 ± 0.09
Ag-110m	249.9	11 ± 1
Os-185	94	2.1 ± 0.2
Au-195	186.09	6.8 ± 0.8
Au-196	6.2	160 ± 2
Au-198	2.696	2800 ± 40

P-32 → Chemistry+LSC

Be-7 to Au-198: Gamma-ray spectrometry

Discussion: Estimation of internal dose* (ICRP publication 72)

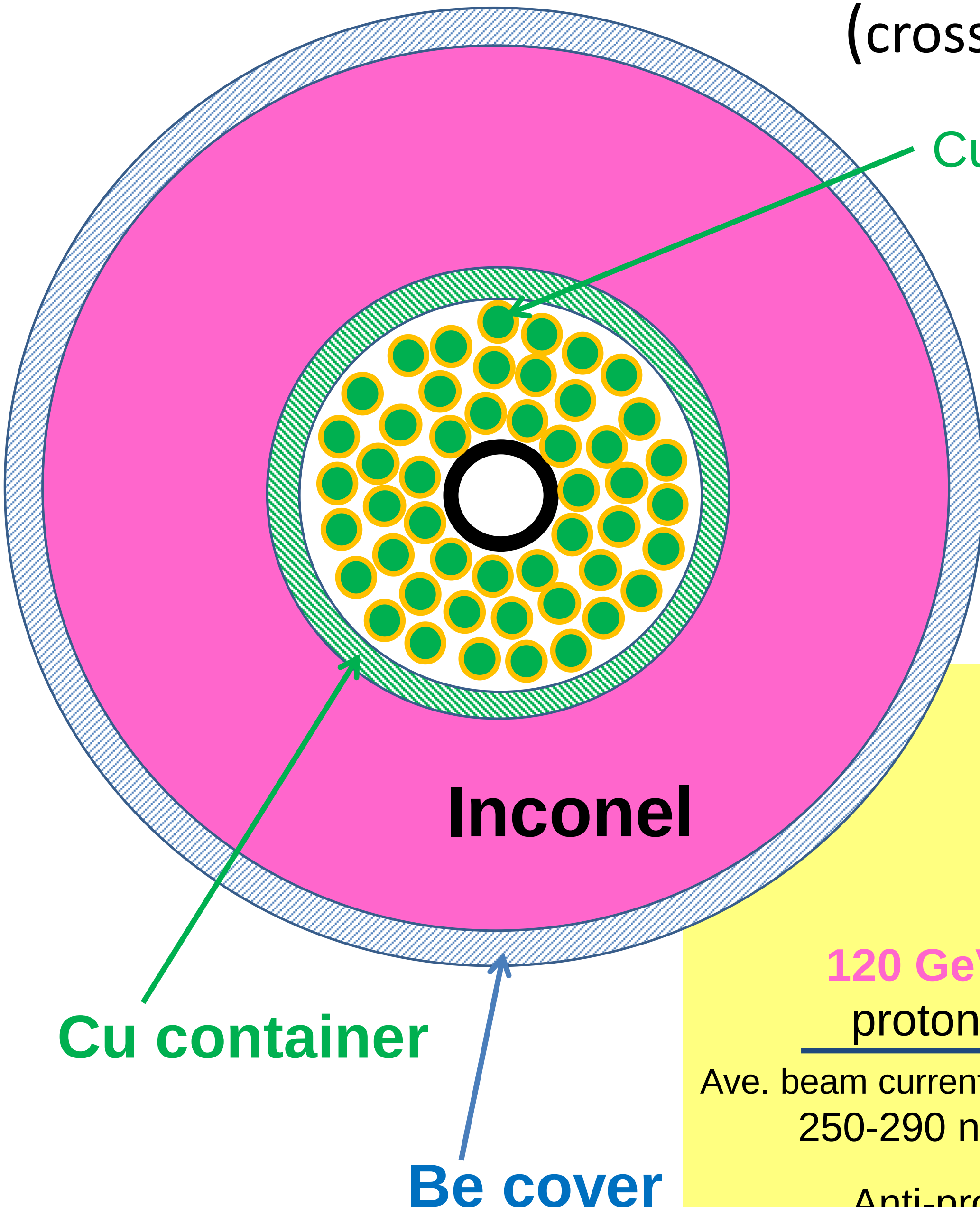
Nuclide	half-life (d)	Activity (Bq)			μ Sv/h	Ratio (%)
P-32	14.26	26	±	3	**0.0071**	2.5
Be-7	53.29	880	±	6	0.0039	1.4
Na-22	950.095	2.0	±	0.1	0.0002	0.1
Sc-46	83.82	8.4	±	0.2	0.0046	1.6
V-48	15.97	31	±	1	0.0060	2.1
Cr-51	27.7	80	±	3	0.0002	0.1
Mn-52	5.6	34	±	2	0.0038	1.4
Mn-54	312.2	15	±	1	0.0018	0.6
Co-56	77.26	34	±	1	0.0182	6.5
Co-57	271.79	64	±	1	0.0051	1.8
Co-58	70.86	110	±	2	0.0185	6.6
Fe-59	44.503	4.4	±	0.3	0.0014	0.5
Co-60	1924.28	1.6	±	0.1	0.0040	1.4
Se-75	119.64	1.4	±	0.1	0.0001	0.1
Y-88	106.6	0.44	±	0.09	0.0002	0.1
Ag-110m	249.9	11	±	1	0.0106	3.8
Os-185	94	2.1	±	0.2	0.0003	0.1
Au-195	186.09	6.8	±	0.8	0.0009	0.3
Au-196	6.2	160	±	2	0.0000	0.0
Au-198	2.696	2800	±	40	**0.1926**	68.9

Aggregate dose: 0.28 μSv/h

*Inhalation dose coefficients: **to age 70 y** for adults

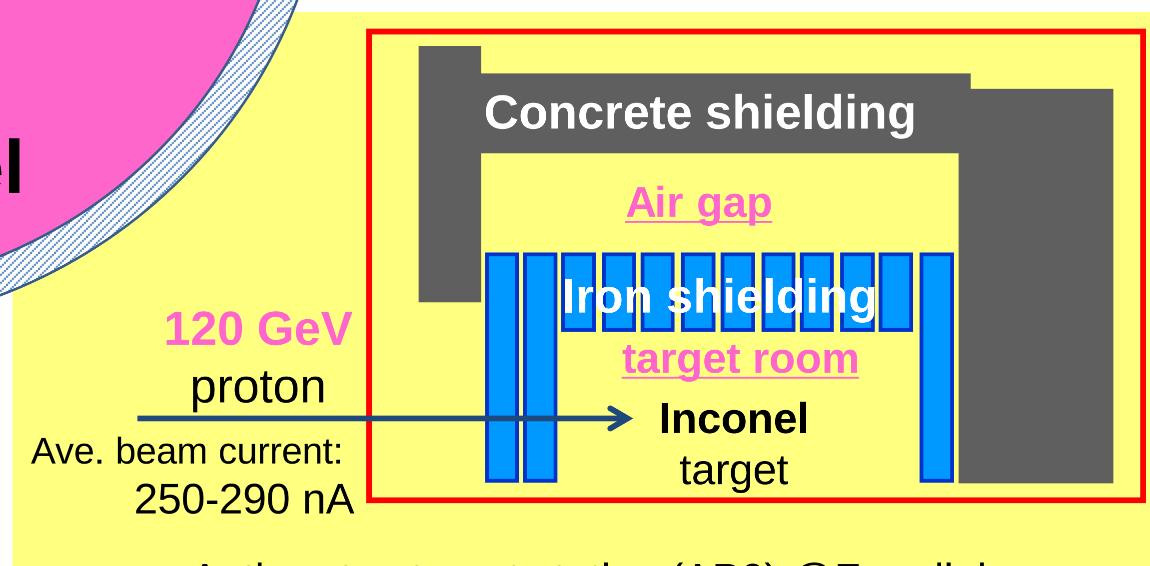
Discussion “Target materials in detail”

(cross-section drawing of target parts)



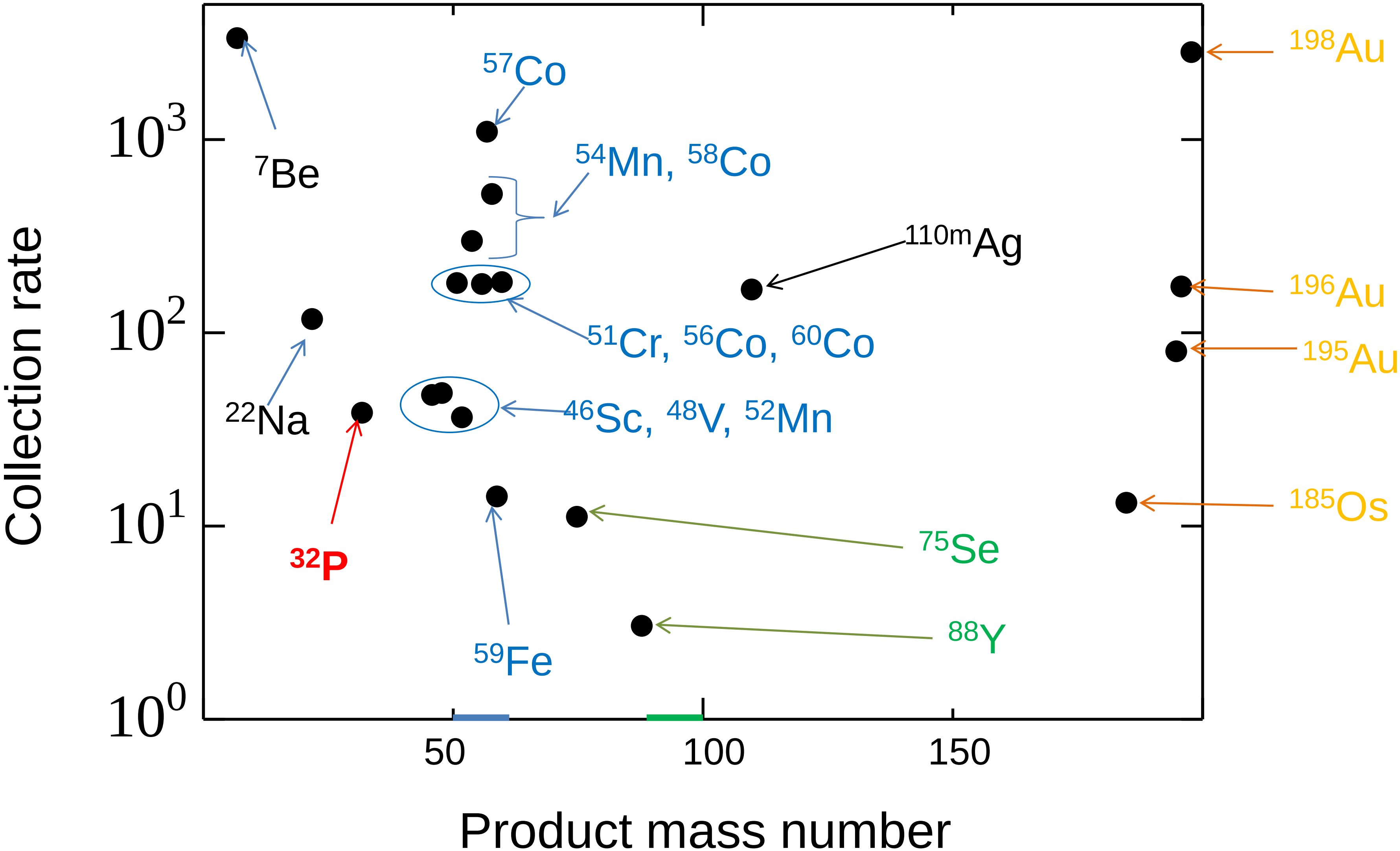
To cool down the Inconel target, Cu container and Cu balls are installed in the Inconel-target-hole.

Inconel: Ni, Fe, Cr, Nb, Mo etc.,



Anti-proton target station (AP0) @Fermilab

Discussion "Which contribute to P-32 production, air, target, etc.?"



Spallation products from Au coating of Cu balls, SP from Nb, Mo in Inconel, SP from Cr, Fe and Ni in Inconel, ^{22}Na, ^{7}Be: SP from Inconel, ^{32}P: Ar in air

Discussion: Source material?

Nuclide	half-life (d)	Activity (Bq)	plausible **main** source
P-32	14.26	26 ± 3	Ar in air
Be-7	53.29	880 ± 6	spallation product (SP) from target
Na-22	950.095	2.0 ± 0.1	spallation product (SP) from target
Sc-46	83.82	8.4 ± 0.2	SP from Cr in Inconel
V-48	15.97	31 ± 1	SP from Cr in Inconel
Cr-51	27.7	80 ± 3	Cr in Inconel
Mn-52	5.6	34 ± 2	SP from Fe, Ni in Inconel,
Mn-54	312.2	15 ± 1	SP from Fe, Ni in Inconel,
Co-56	77.26	34 ± 1	SP from Ni in Inconel,
Co-57	271.79	64 ± 1	SP from Ni in Inconel,
Co-58	70.86	110 ± 2	SP from Ni in Inconel,
Fe-59	44.503	4.4 ± 0.3	SP from Fe, Ni in Inconel,
Co-60	1924.28	1.6 ± 0.1	SP from Ni in Inconel,
Se-75	119.64	1.4 ± 0.1	SP from Nb, Mo in Inconel,
Y-88	106.6	0.44 ± 0.09	SP from Nb, Mo in Inconel,
Ag-110m	249.9	11 ± 1	SP from Au coating of Cu balls
Os-185	94	2.1 ± 0.2	SP from Au coating of Cu balls
Au-195	186.09	6.8 ± 0.8	SP from Au coating of Cu balls
Au-196	6.2	160 ± 2	SP from Au coating of Cu balls
Au-198	2.696	2800 ± 40	SP from Au coating of Cu balls

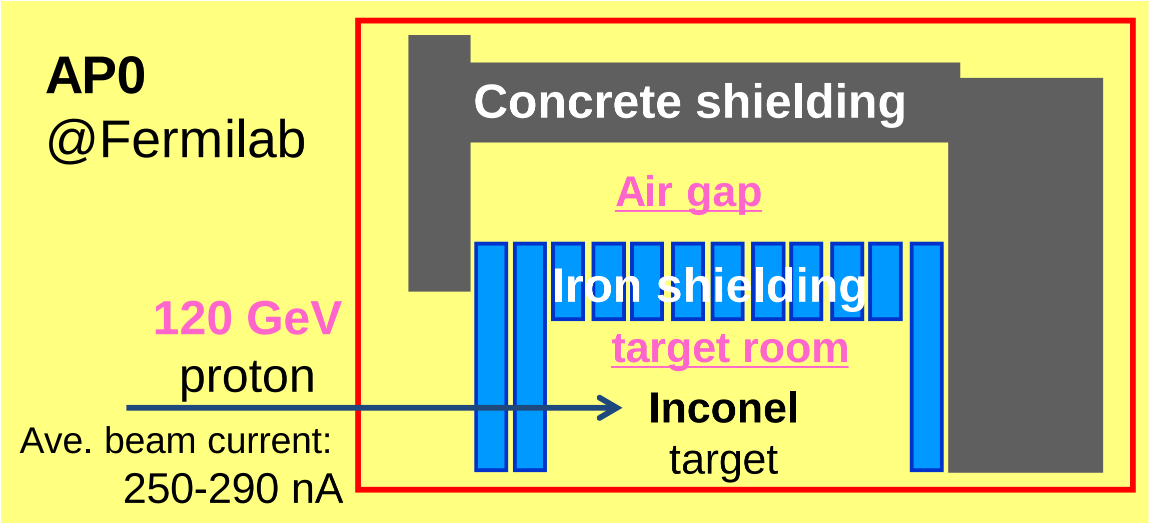
Cu-container & ball (Sc-46 to Co-60)

Discussion: Estimation of internal dose (ICRP publication 72*)

Nuclide	half-life (d)	Activity (Bq)	μ Sv/h	Ratio (%)
P-32	14.26	26 ± 3	0.0071	2.5
Au-198	2.696	2800 ± 40	0.1926	68.9
Aggregate dose			0.28	100

P-32 :**0.014** mSv
Au-198 :**0.4** mSv
Aggregate dose :**0.56** mSv

Adult,
2000 h/year,



*Inhalation dose coefficients:
to age 70 y for adults

Summary

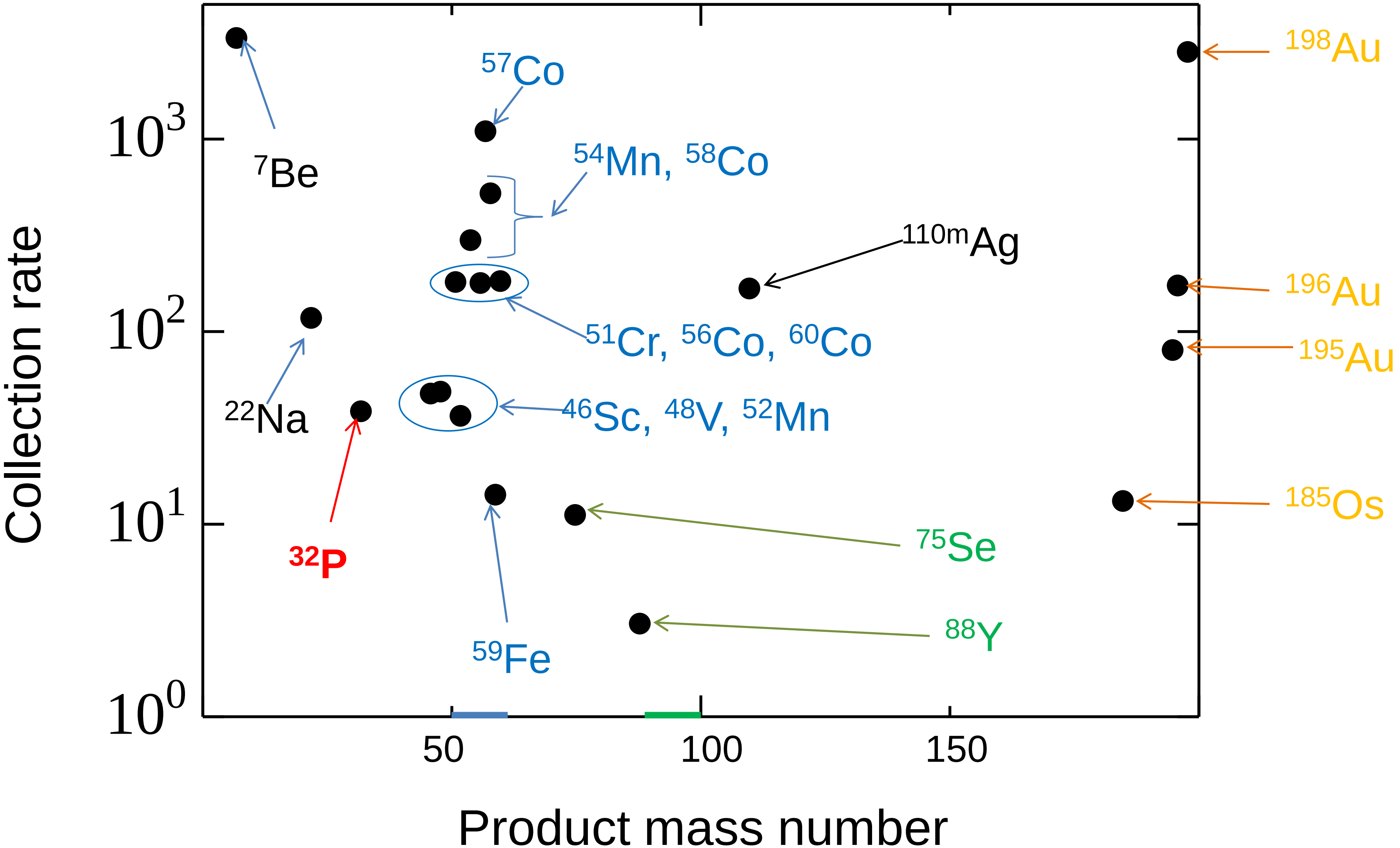
Determination of **the activity levels** of 20 radionuclides produced in 120 GeV proton target station the beta emitting (AP0, FNAL)

Measuring the beta emitting radio-nuclide ^{32}**P**

Estimation of internal dose
→ P-32: 0.007 μSv/h, Aggregates: 0.28 μSv/h

Discussion on their **source material**

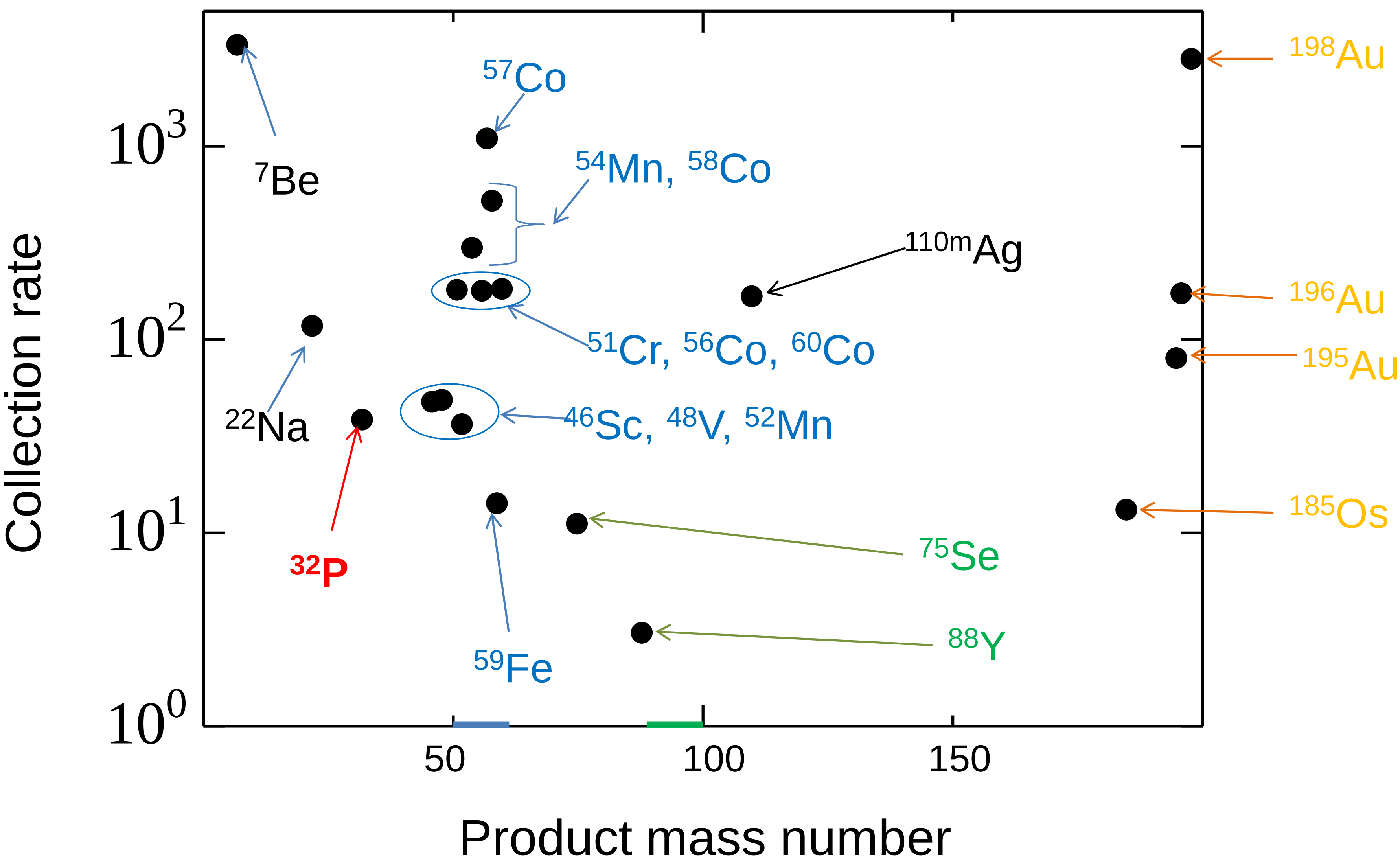
Discussion “Which contribute to P-32 production, air, target, etc.?”



^{7}Be, ^{198}Au**:** high collection rate

^{22}Na & ^{46}Sc, ^{48}V, ^{52}Mn**:** not so higher collection rate

Discussion "Which contribute to P-32 production, air, target, etc.?"



^{32}P: < ^{22}Na, ≈ ^{46}Sc, ^{48}V, ^{52}Mn

→mainly produced from target materials not Ar in air

Experimental (Chemical treatment to separate P)

The phosphorus compound $MgNH_4PO_4 \cdot 6H_2O$ was extracted from the filter paper by a chemical separation procedure*. (@Hot laboratory in KURRI)

filter paper collecting radioactive aerosols

↓ # leached by nitric acid

leaching solution

↓
- # 5 mg of P-carrier ↘
- # Ammonium heptamolybdate sol. ↘
- ← heating and stirring

yellow solid as **$(NH_4)_3P(Mo_3O_{10})_4$**

↓
- # dissolved in ammonia sol.
- # HCl aq. ↘ to slightly acid
- # Citric acid ↘
- ← stirring in ice bath
- # magnesia mixture ($MgCl_2$ + NH_4Cl sol.) ↘
- # ammonia sol. ↘ to slightly basic
- ← filtration

white solid as **$MgNH_4PO_4 \cdot 6H_2O$** (20.4 mg) → liquid scintillation counter

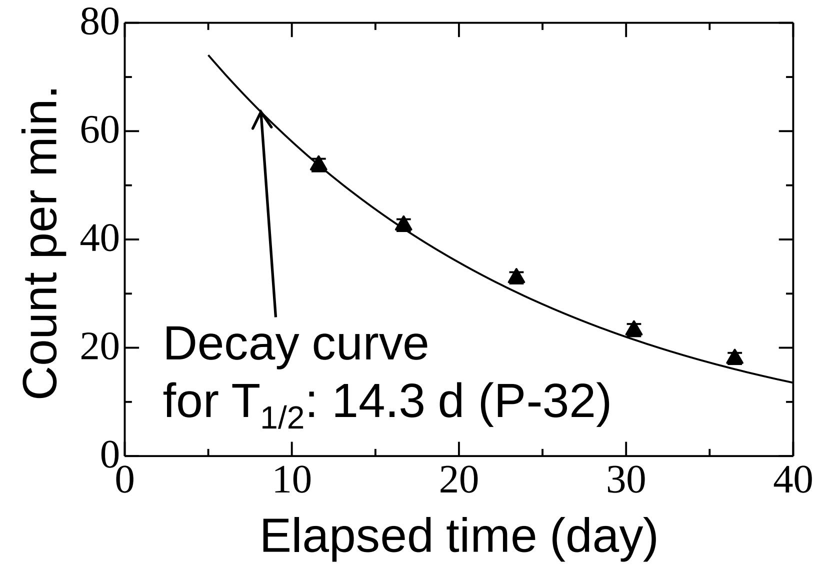
Chemical yield: 51%
(estimated from P-carrier)

*W. T. Mullins and G. W. Leddicotte, "The Radiochemistry of Phosphorus" National Academy of Science (1962)

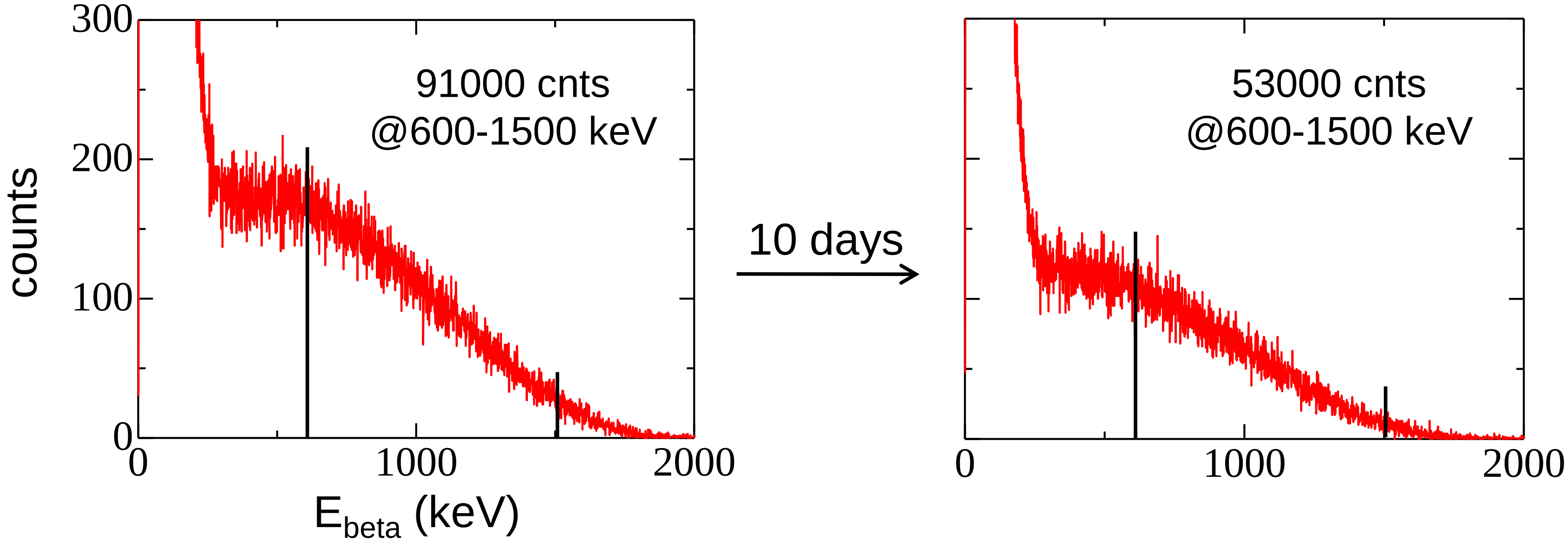
Results "Beta-ray counting and spectrum"

$MgNH_4PO_4 \bullet 6H_2O$

Beta-ray counting
by Geiger-Mueller
counter



Beta-ray spectra from liquid scintillation counter



Results "Activity level of P-32"

$MgNH_4PO_4 \bullet 6H_2O$

Comparing beta-ray spectrum of reference std. of P-32,

26 Bq of P-32

(at the time just after aerosol-sampling from target room)

Beta-ray spectra from liquid scintillation counter