Applications of Accelerators to Environmental Protection at the Idaho Accelerator Center

D.P. Wells,
C.R. Segebade

Idaho Accelerator Center

ISU

Applications of Photon Activation and Activation Analysis (PAA) (with environmental implications)

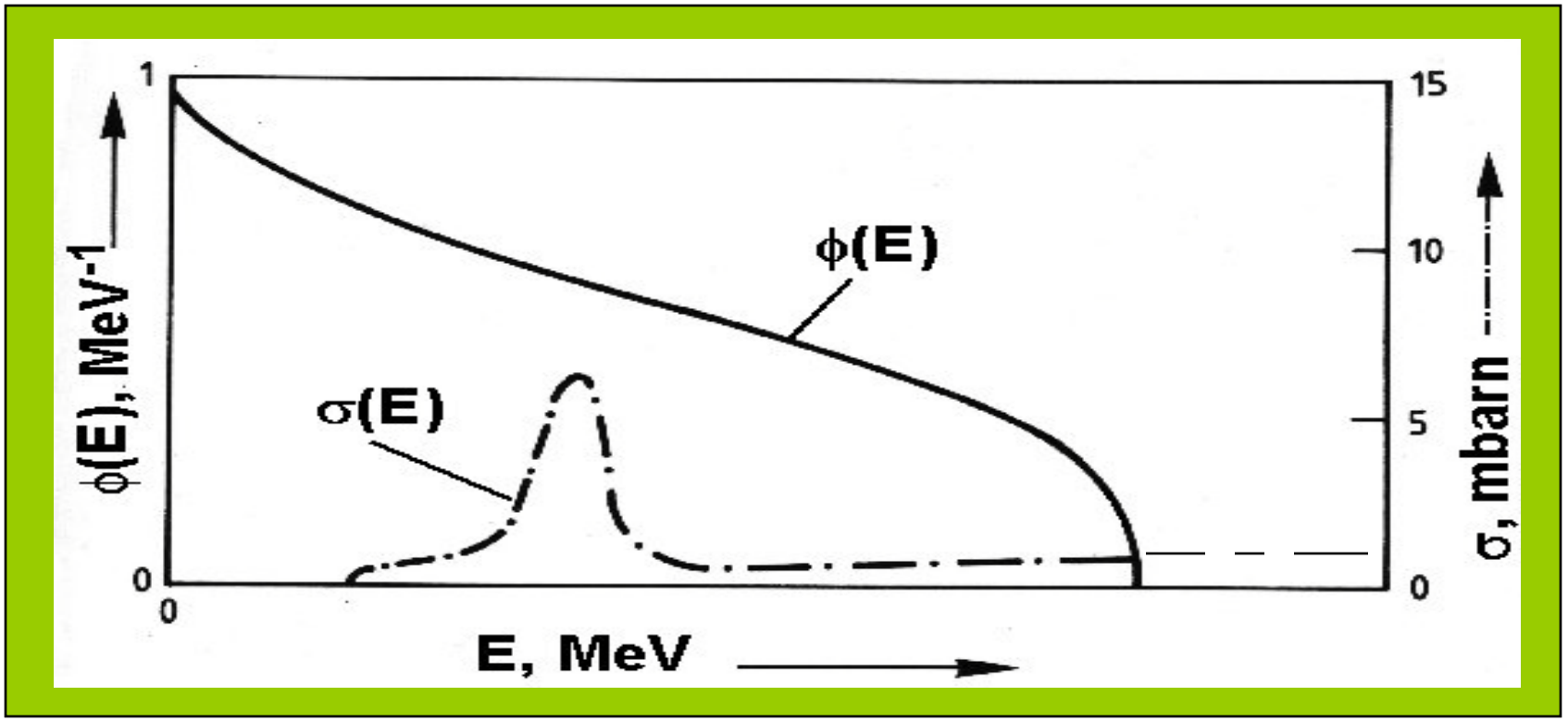
- Trace element analysis of waste streams,
- Trace element analysis of environmental samples (air, water/ice, soil/rock, biological),
- Isotope Production and the avoidance of nuclear waste,
- ‘Burnup’ of nuclear waste with ‘coupled’ accelerator-reactor systems.

Why PAA? Sensitivity and Non-Destructivity:

Absolute Sensitivity – ignoring “matrix effects”,
Relative Sensitivity in most cases is a few ppb.

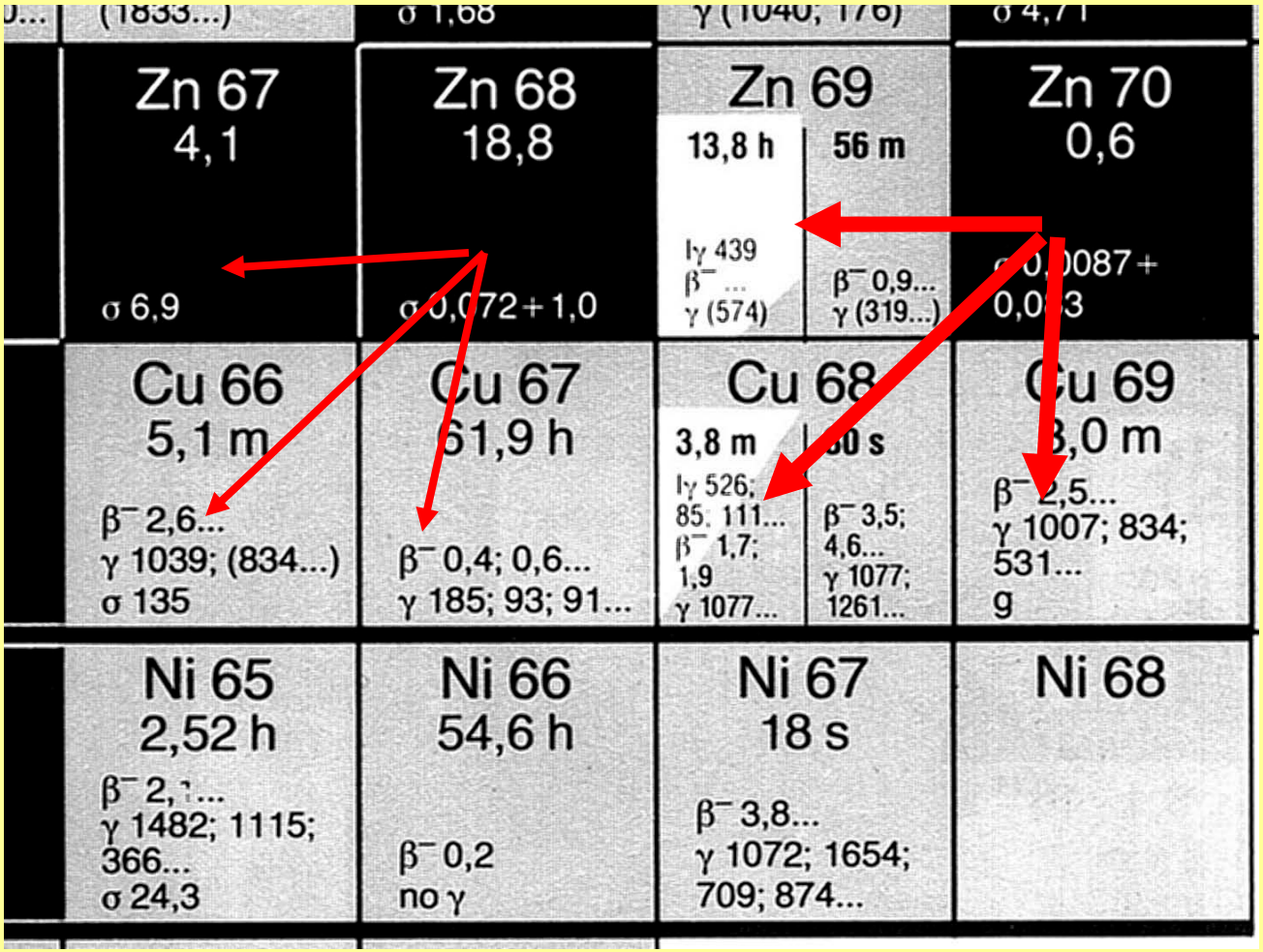
Element	Sensitivity μg
N	0.02
O	0.05
F	0.001
Cl	0.01
Ni	0.6
Sr	0.02
Cd	0.2
Au	0.02
Tl	0.4
Pb	1.0
U	0.0005

How does it work?
Yield = Radionuclide production rate (neglecting decay)



$$Y \propto M \int_{E_S}^{E_{\max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$

The general principle



Reaction types:

- γ, γ´
- γ, xn
- γ, p
- γ, np
- γ, α
- γ, αn
- γ, f
- γ, ...

Example: Waste Assay

- Why PAA?
- What are its advantages?
- Is it practical?

The Problem of Large Amounts
in chemical analysis

Chr. Segebade[1], P. Bode[2] and W. Goerner[1]

[1]Federal Institute for Materials Research and Testing (BAM), Berlin
[2]IRI Interfaculty Reactor Institute ,Delft University of Technology, Delft, the Netherlands

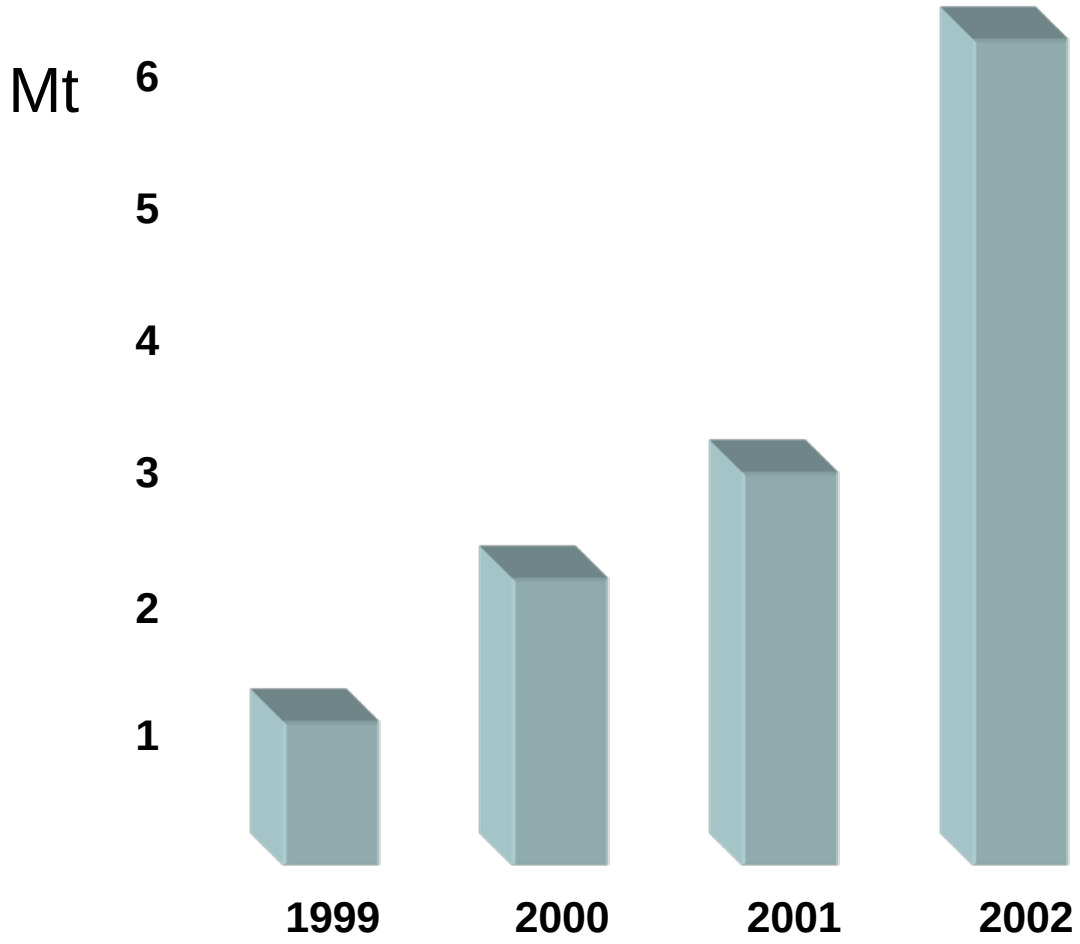
The current “modern trends” in chemical analysis...

Almost ignored, up to present:

The characterisation of large amounts / volumes of material

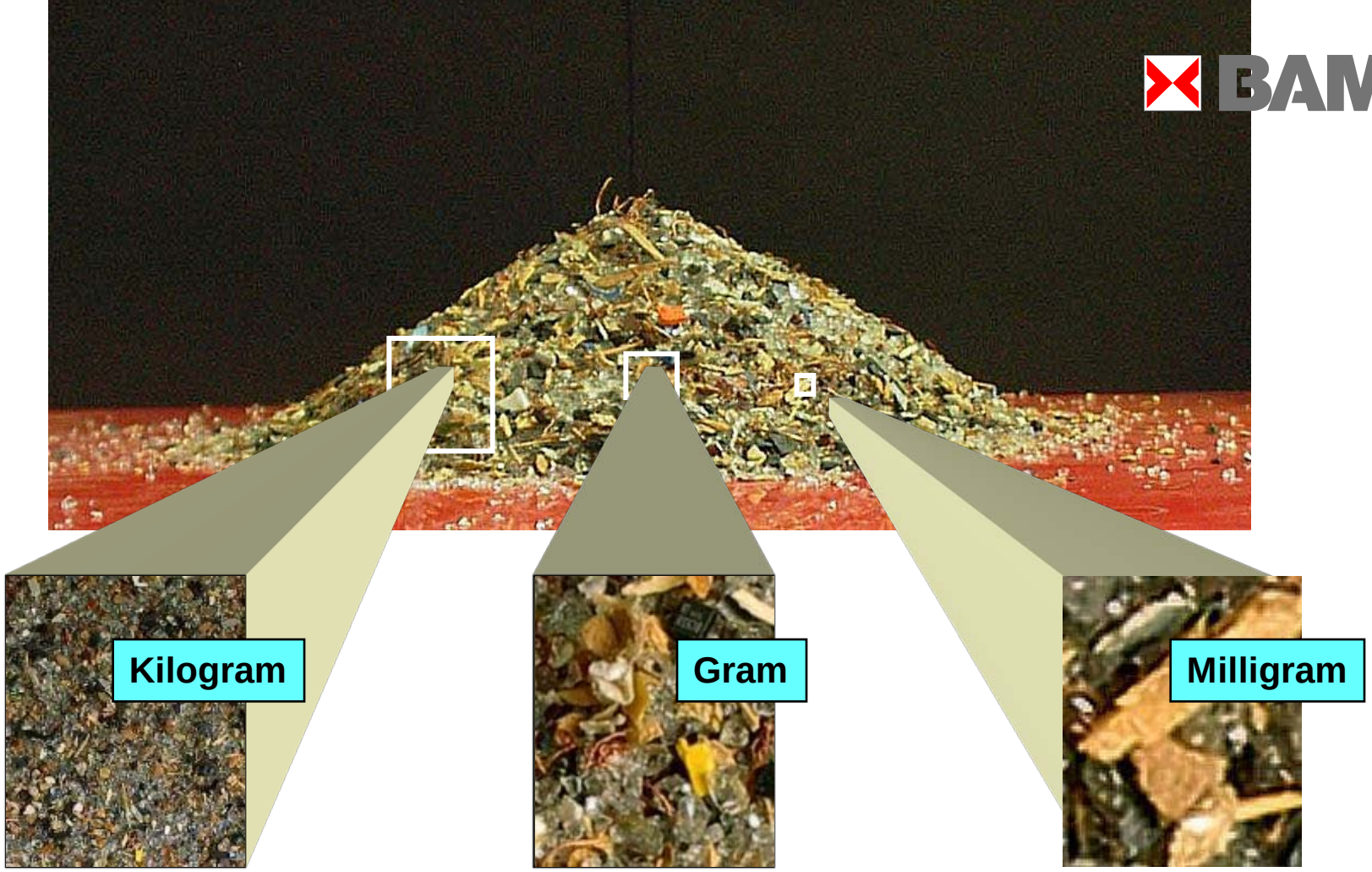
Example: electronic waste; catalysts

Example: electronic waste in Germany



Hazardous:
- Chromium
- Nickel
- Arsenic
- Bromine
- Cadmium
- Lead
- Bismuth
- etc.

Useful:
- Copper
- Selenium
- (PGE)
- Silver
- Indium
- Tin
- Gold
- etc.



Precondition:

<u>Analysis of large samples</u> (5 g up to tens of kg)

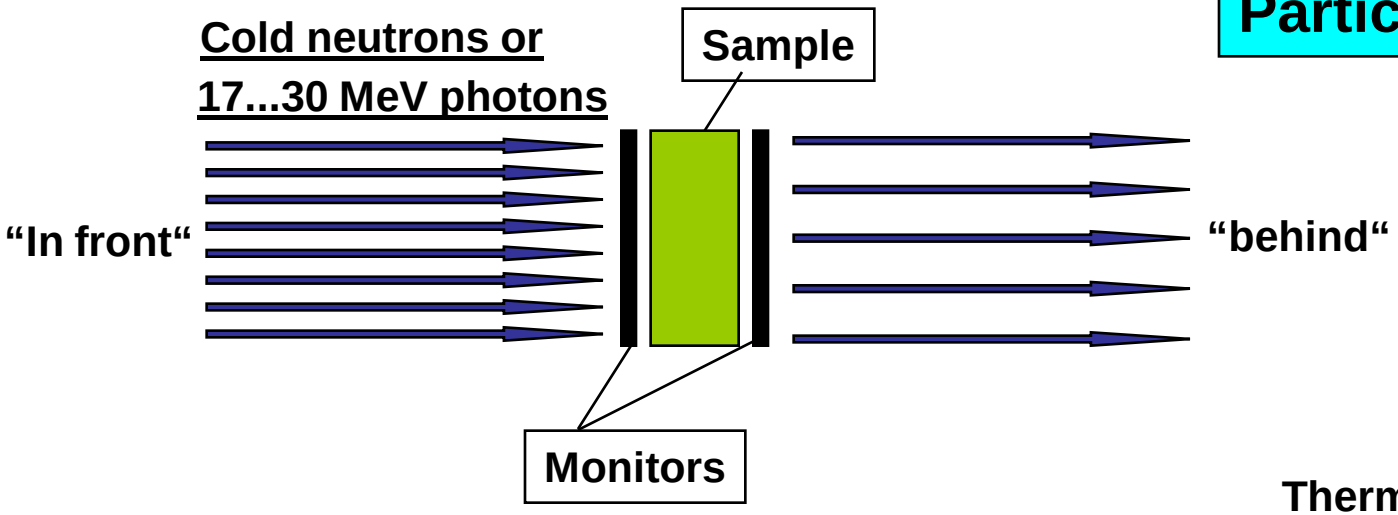
Potentially Suitable analytical methods

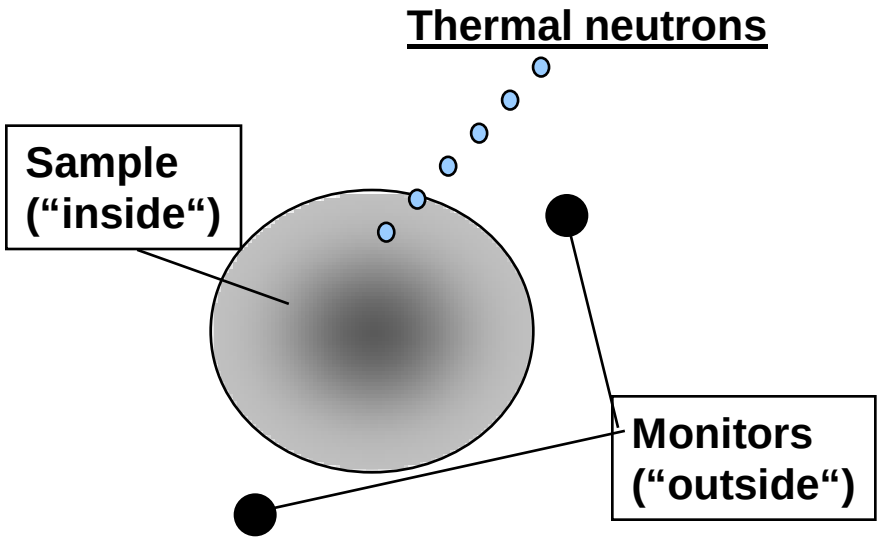
- **X-ray fluorescence analysis**, continuous

- **Activation analysis**
  - with photons (PAA)
  - with neutrons (NAA)

PHOTON BEAM GEOMETRY vs. REACTOR GEOMETRY

Particle consumption





PHOTON BEAM GEOMETRY vs. REACTOR GEOMETRY

Particle consumption

Cold neutrons or photons

- Linear (exponential) interpolation possible

Thermal neutrons

- Extrapolation from “outside“ to “inside“

Flux depression and self-shielding

- Matrix Z dependent
- Strongly matrix dependent

PHOTON BEAM GEOMETRY vs. REACTOR GEOMETRY

Flux gradient

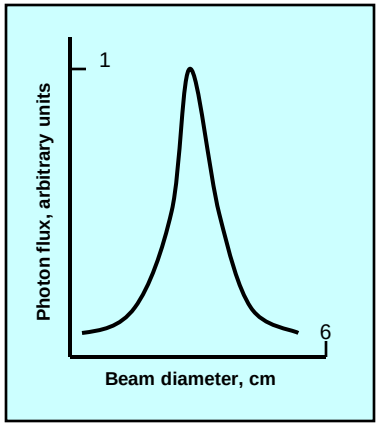
Cold neutrons or photons

- lateral: high
  axial: low

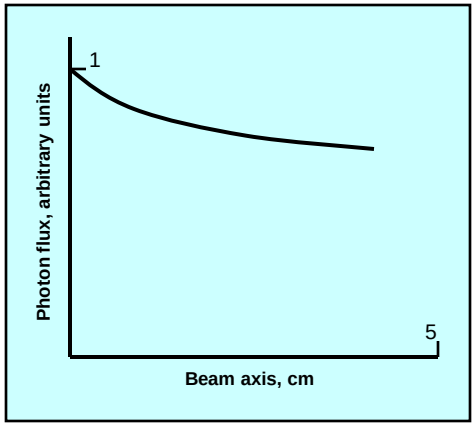
Thermal neutrons

- radial: high
  circumferential: low

Example: 30 MeV bremsstrahlung



Lateral

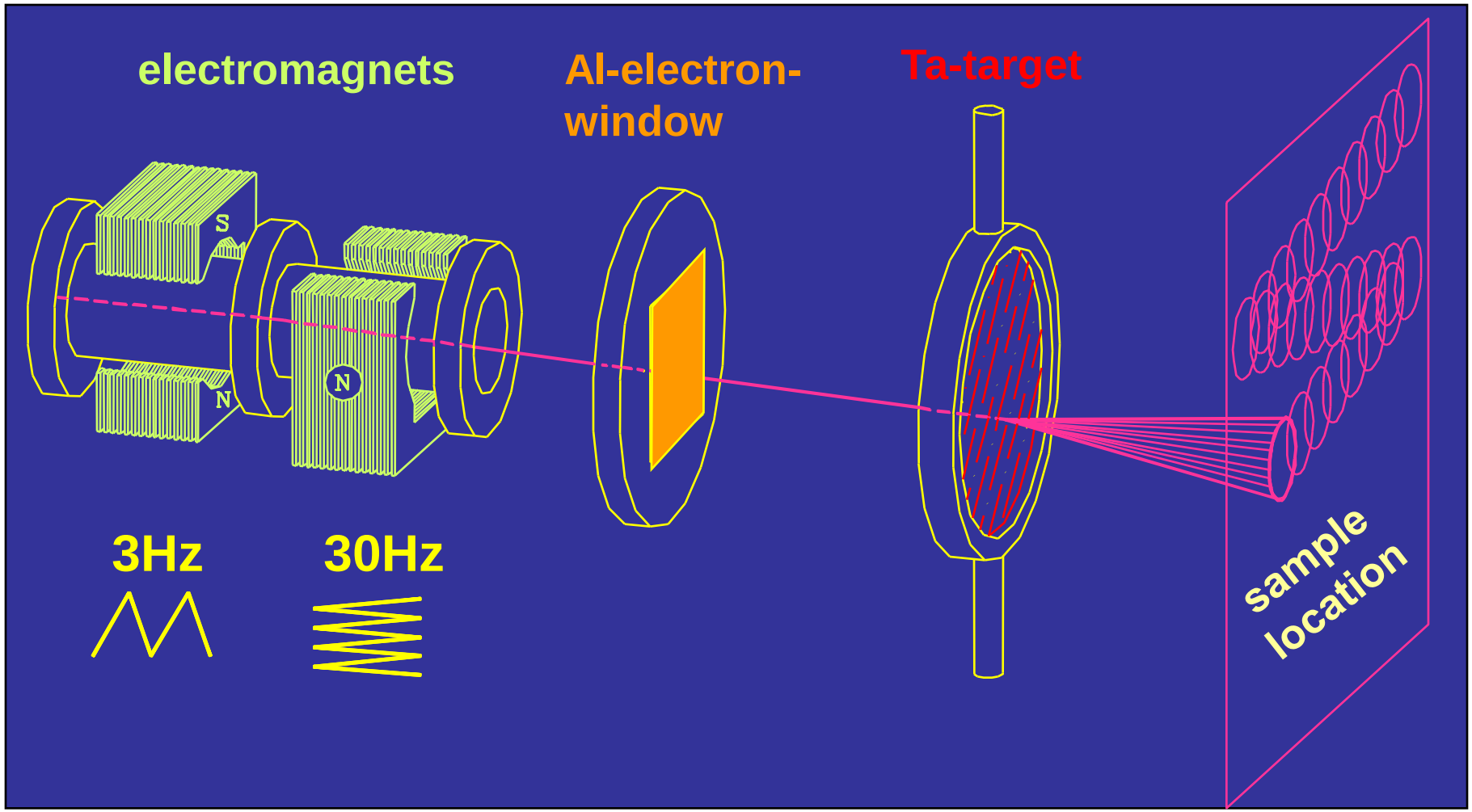


Axial

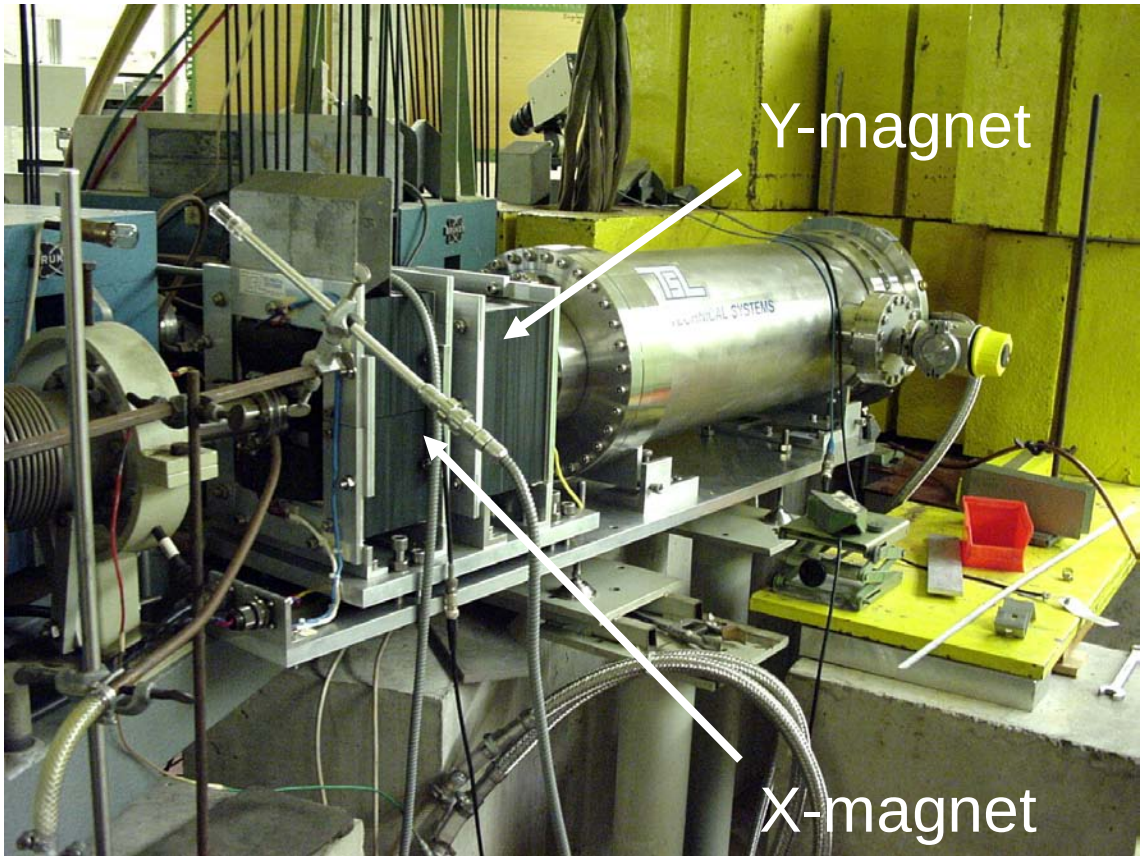
FWHM ca. 1.5cm

Ca. 18% flux decrease @ 4.5cm

THE SOLUTION: **High Energy Photon Screen (HEPS)**



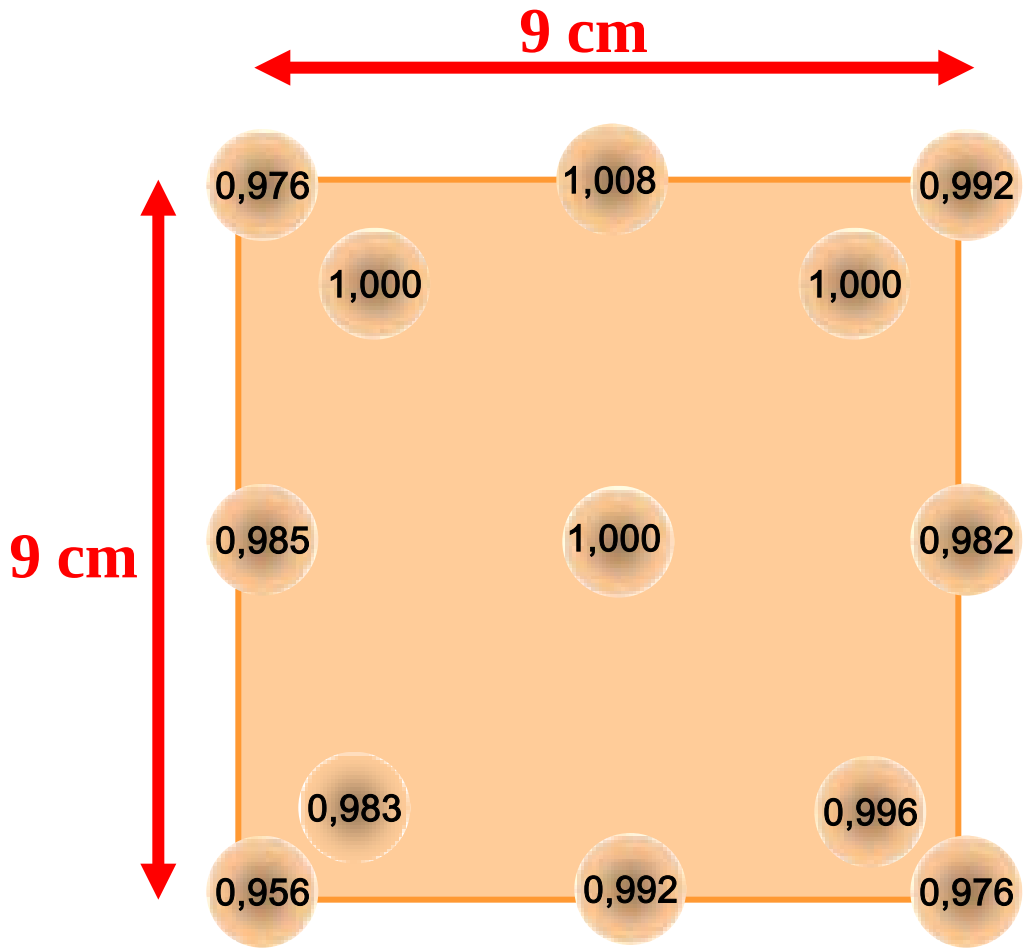
End of beamline with x,y-scanning magnets



Areal homogeneity of the scanning bremsstrahlung field

Copper sheet matrix

Areal homogeneity of the scanning bremsstrahlung field

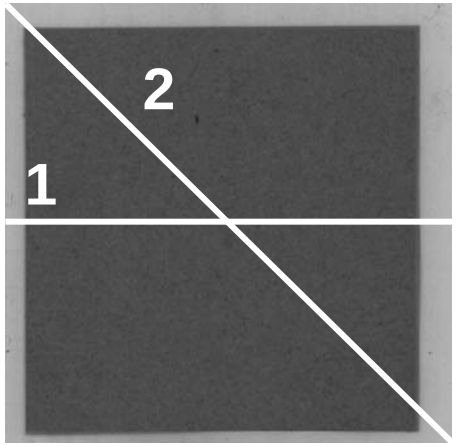


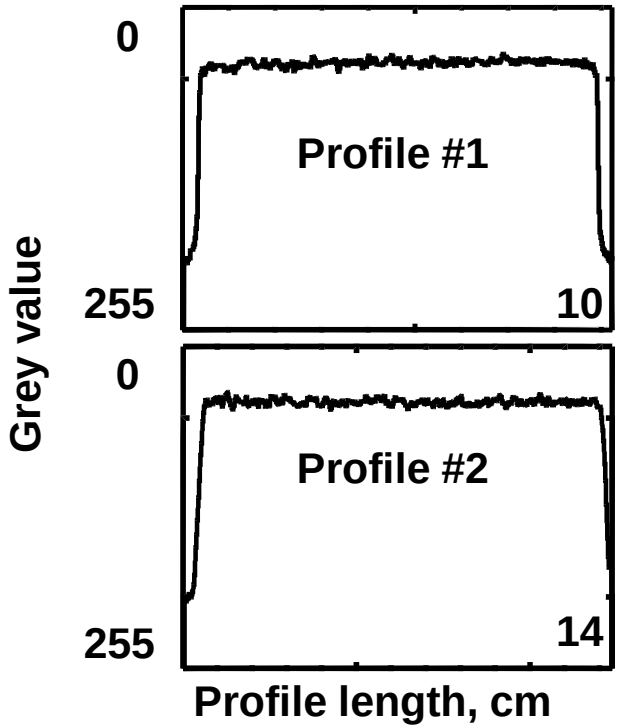
Normalized ^{64}Cu pulse rates after a 1 hour meander irradiation of 300 mg Cu discs at 30 MeV electron energy

High Energy Photon Screen (HEPS)

- **Areal homogeneity of the scanning bremsstrahlung field**

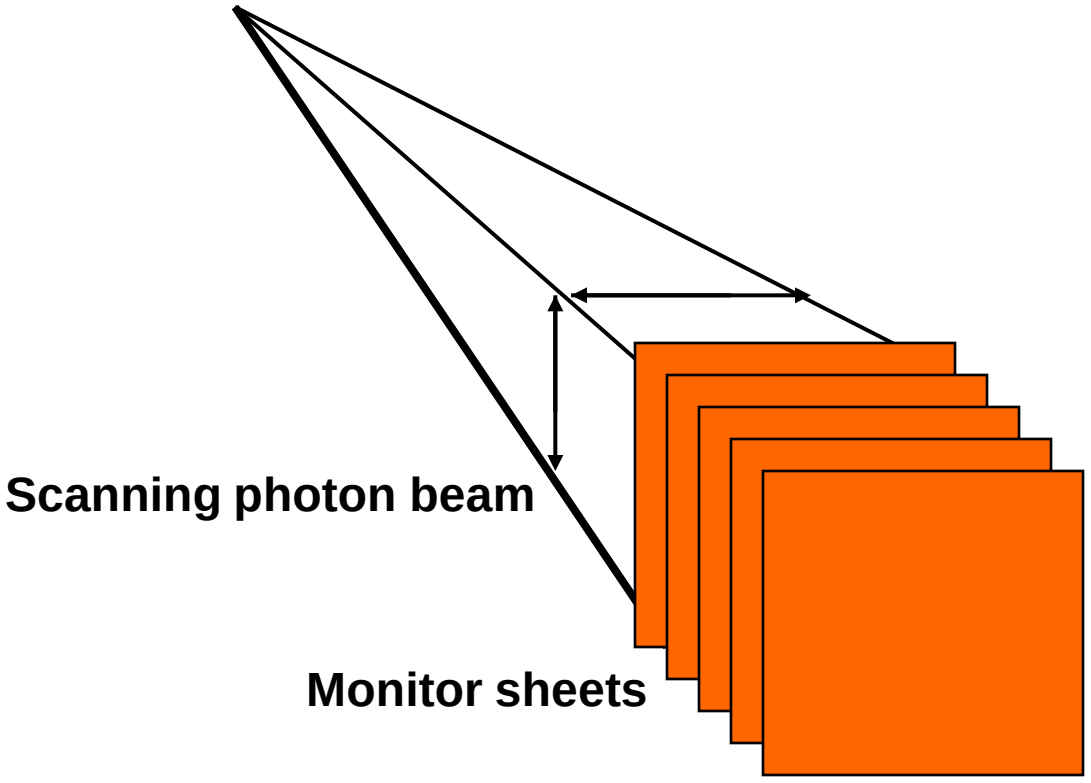
Autoradiographic approach





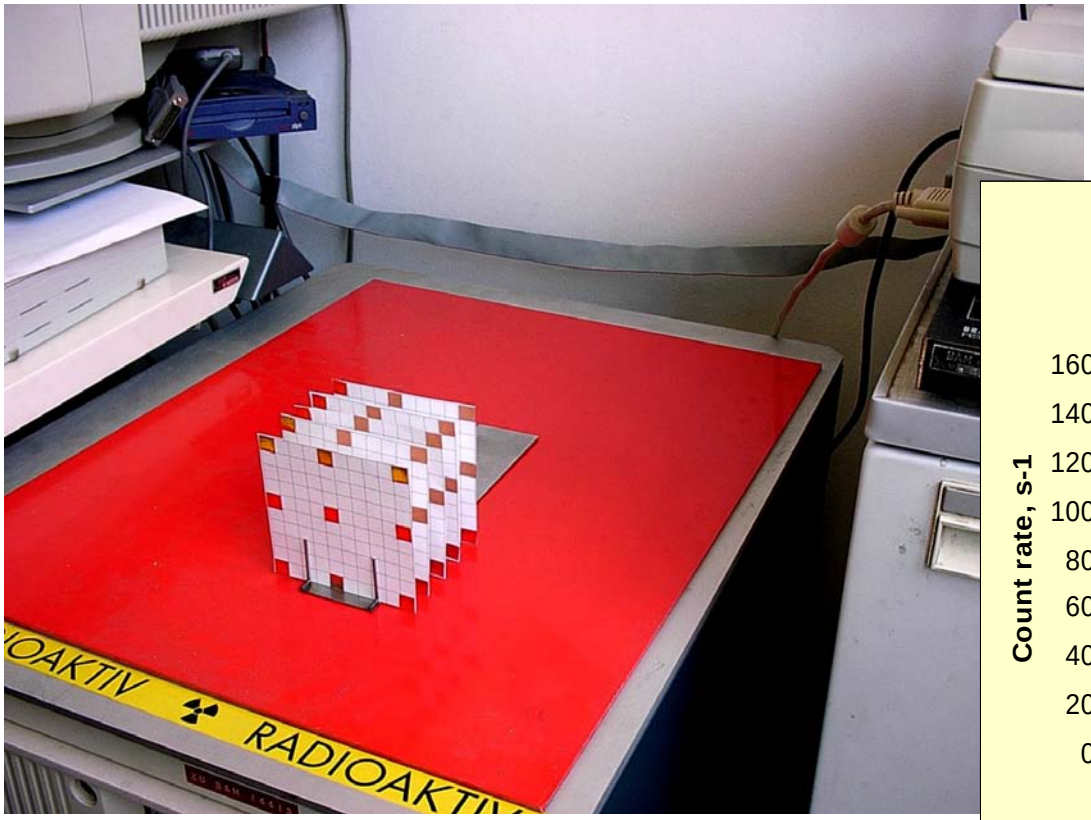
High Energy Photon Screen (HEPS)

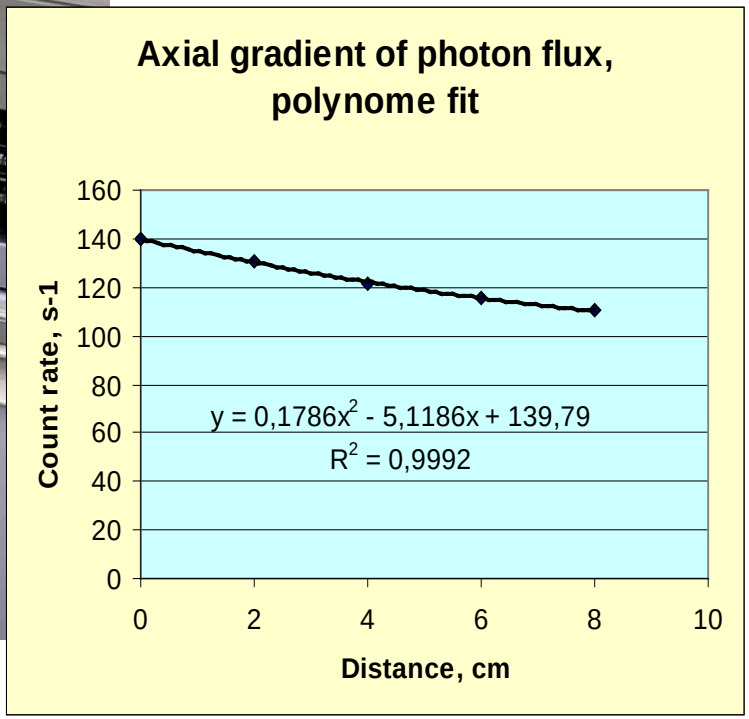
- **<u>Axial</u> homogeneity of the scanning bremsstrahlung field**



High Energy Photon Screen (HEPS)

- **Axial homogeneity of the scanning bremsstrahlung field**





Results

Contents, mg/kg (± 1σ, rel.%)

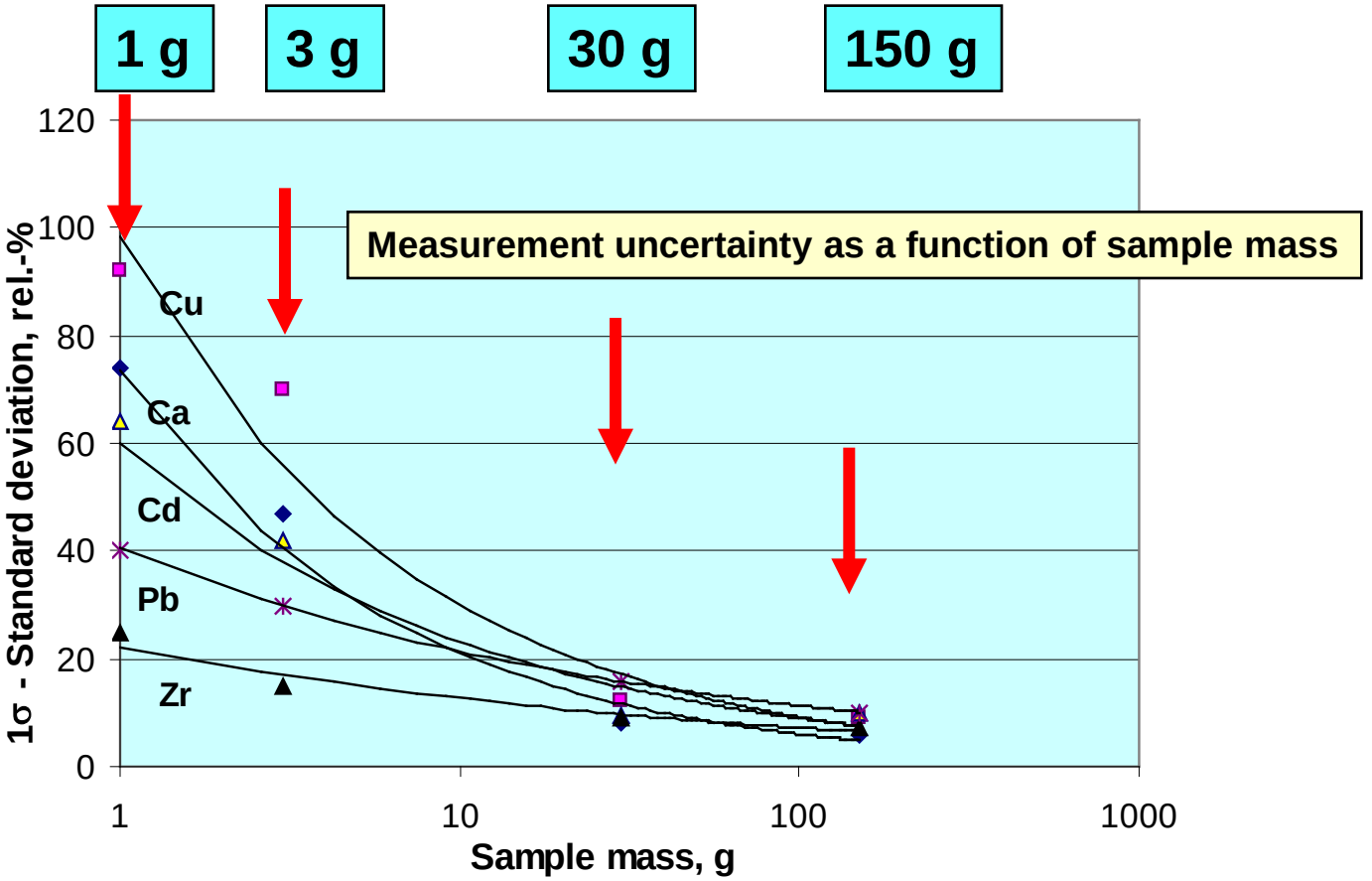
Element	PAA[1]		NAA[2]	
Sodium%	4.8	(16)	4.6	(3.0)
Calcium	652	(6)	n.d.[3]	
Titanium	183	(13)	n.d.	
Chromium	31	(26)	48.8	(9.2)
Manganese	151	(14)	n.d.	
Nickel	134	(20)	n.d.	
Copper%	4.8	(9)	5.36	(8.3)
Zinc%	0.167	(16)	0.162	(3.8)
Zirconium	971	(7)	n.d.	
Silver	14	(28)	25.7	(8.4)
Cadmium	87	(23)	n.d.	
Tin%	0.127	(5)	0.122	(35)
Antimony%	0.200	(8)	0.247	(1.1)
Barium%	4.15	(11)	4.5	(2)
Cerium%	0.23	(16)	0.218	(2.8)
Gold	n.d.		1.91	(2.1)
Lead%	0.107	(10)	n.d.	
Uranium	5.4	(13)	n.d.	

[1]Average out of 5 single samples @ 150 g

[2]One sample @ 1.22 kg

[3]n.d.: not detected

Results


1 g
3 g
30 g
150 g
Measurement uncertainty as a function of sample mass
1σ - Standard deviation, rel.-%
120
100
80
60
40
20
0
Cu
Ca
Cd
Pb
Zr
1
10
100
1000
Sample mass, g

Conclusion

- Large samples can be analysed
- Significantly reduced working effort
- Improved representativeness

Example: Isotope Production

Why use Photon Activation Methods?

Why is it better or worse than reactor methods?

How does it work?

Is it practical?

Current reactor production technique:

fission in a reactor produces a variety of isotopes (about 6% of the time it makes 99Mo).

$$n^o + {}^{235}U \rightarrow {}^{99}Mo + Sn + x\,n^o \quad (\approx 6\%)$$

^{99m}Tc – metastable Technetium-99

- Nice thing about ^{99}Mo – it has a half life of 2.7 days and can be shipped around.

- It decays to ^{99m}Tc which has a half life of 6 hours. This is used on patients. Approximately 1.6M North Americans per month get a ‘treatment’.

- ^{99m}Tc is extracted from ^{99}Mo by “milking the cow”.

Problems with current technique

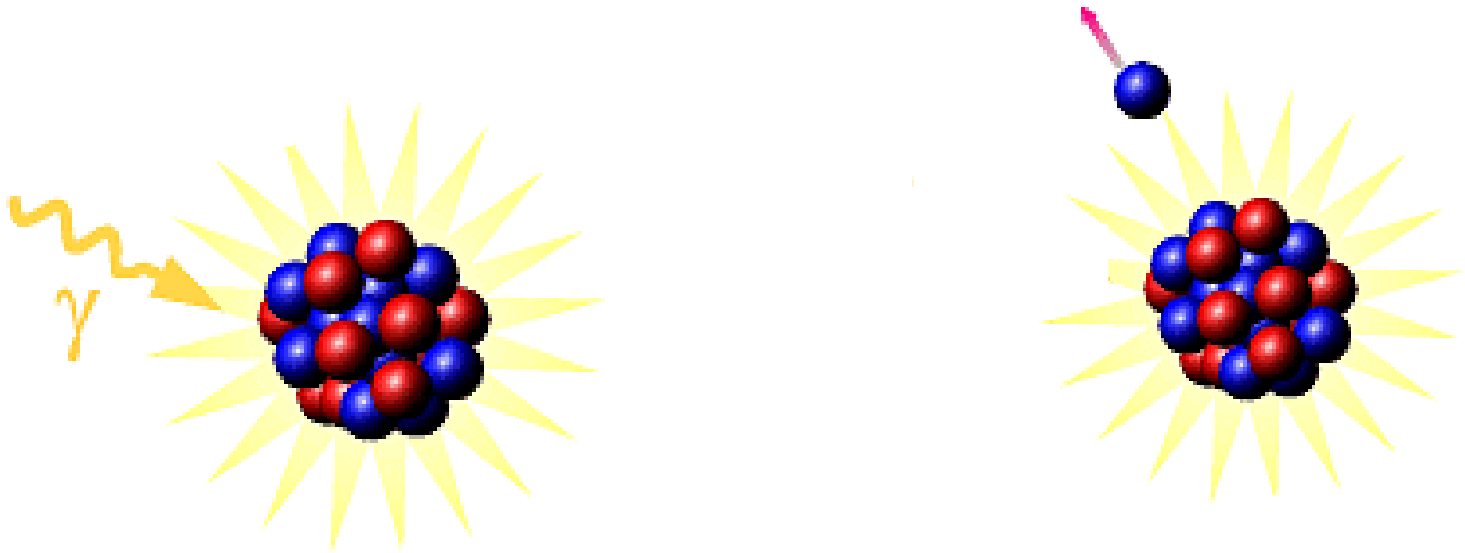
- High level waste
  - Worldwide, 10's of kilograms of (HEU) HLW waste each year.

- Nuclear proliferation
  - U.S. exports 15.5 kg of HEU (highly enriched uranium) to Canada each year.
  - 93% enrichment HEU made for nuclear warheads.

- Reliability of supply
  - 2007 supply crisis.
  - Future problems importing radioactive materials.

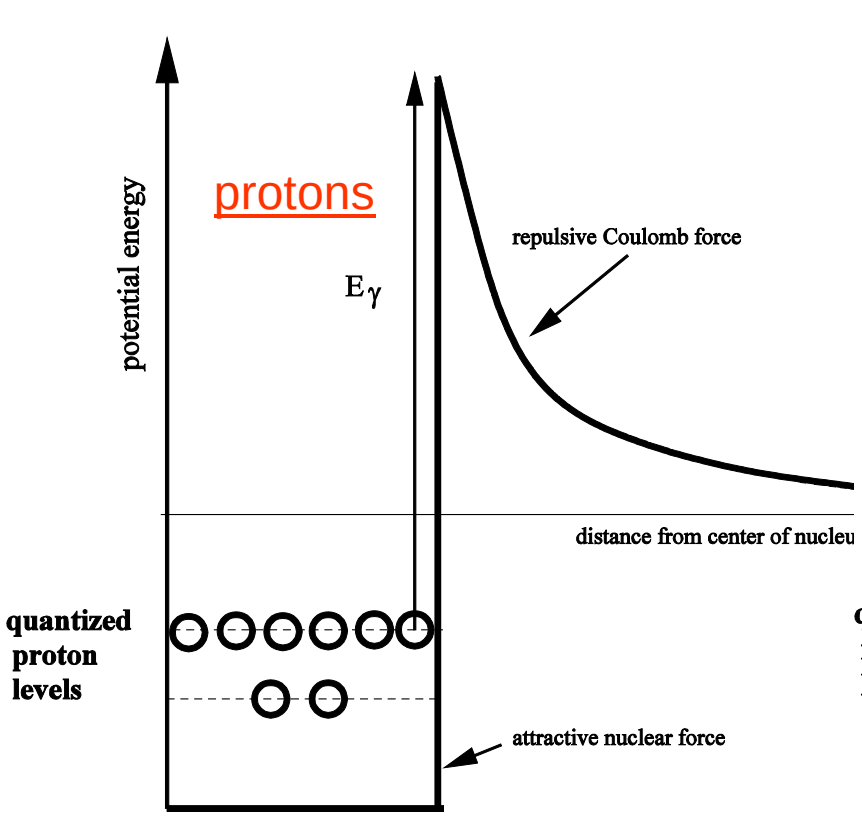
(γ,n) production technique

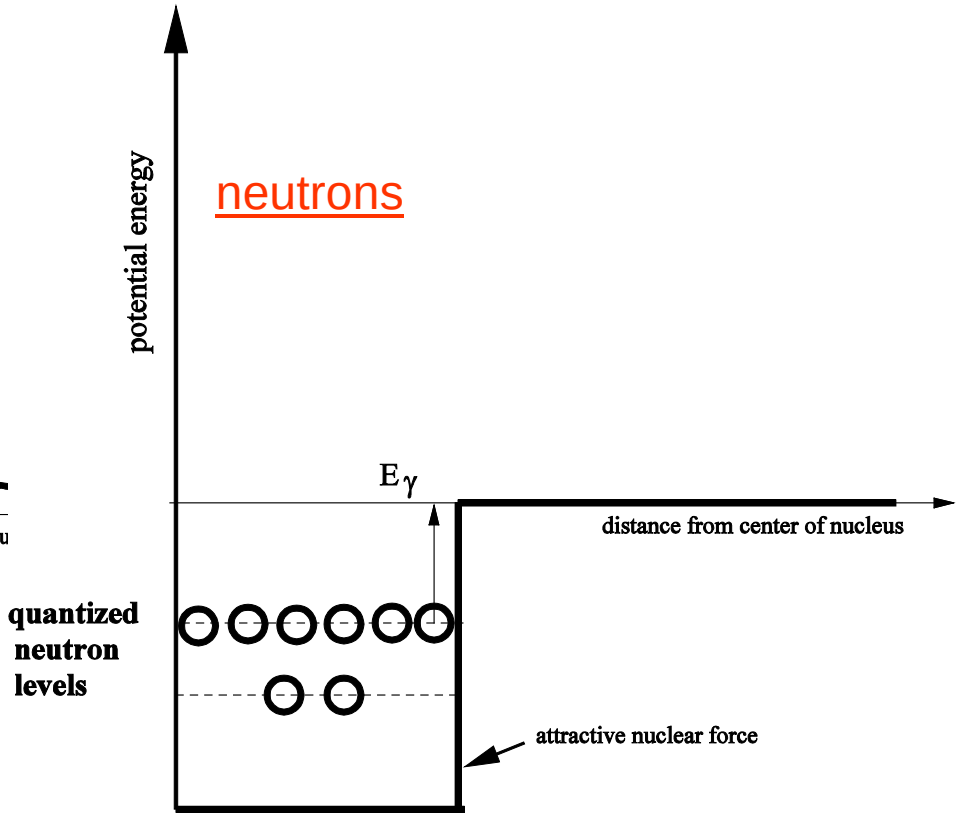
- Photon excites ^{100}Mo nucleus.
- Knocks out a neutron leaving ^{99}Mo.



$$\gamma + {}^{100}Mo \rightarrow {}^{99}Mo + n^{o}$$

In general, photons make ***proton rich*** isotopes

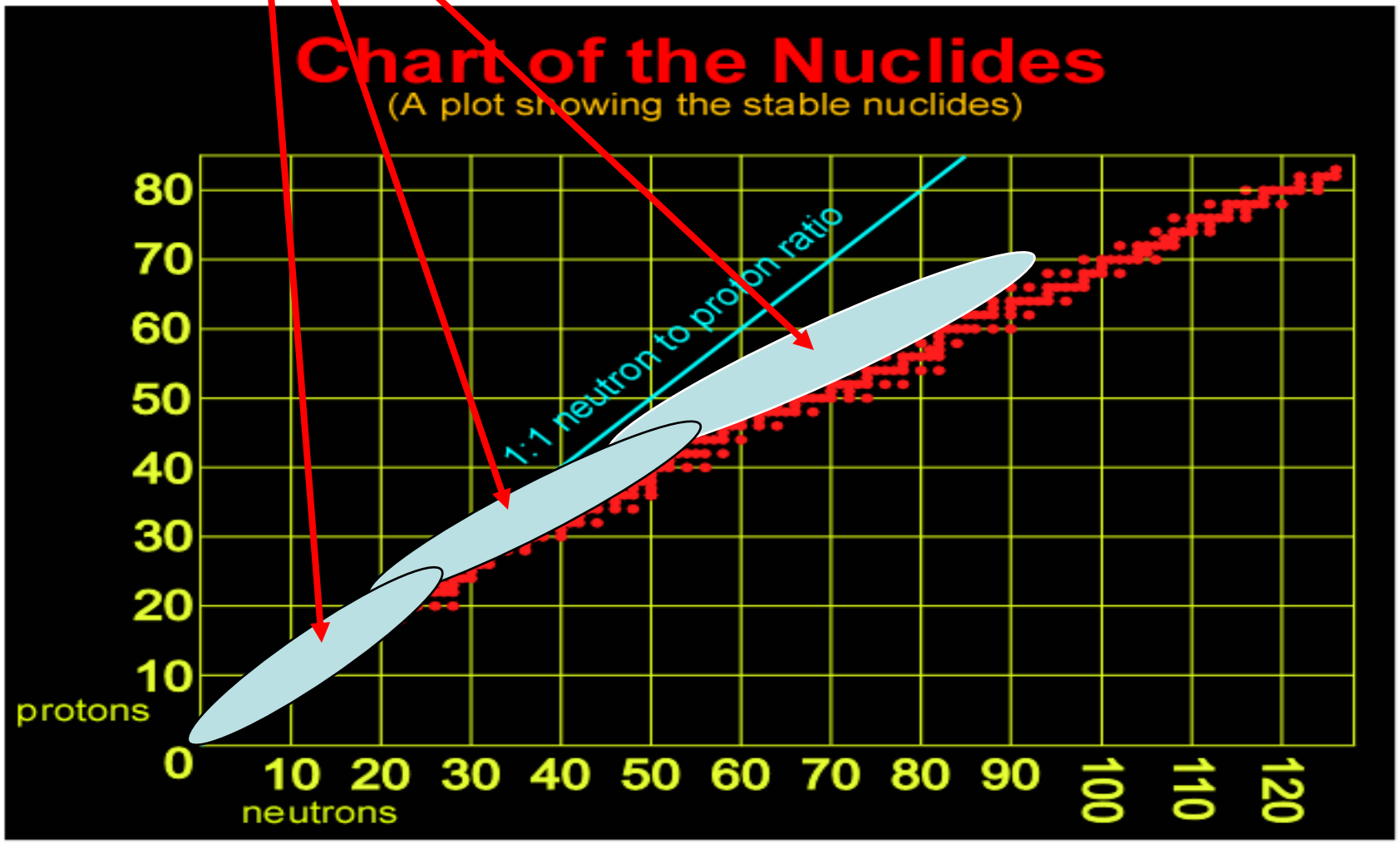




Coulomb energy barrier for protons …

but not for neutrons.

(gamma, x) reactions efficiently populate moderately proton-rich nuclei (and some neutron-rich nuclei)



Isotope	Crude yield per 100 kW per week from 100g targets at 50 MeV electron beam energy
F-18*	9 kCi/wk
Cu-64*	10 kCi/wk
Ba-131*	15 kCi/wk
Mo-99*	2 kCi/wk
In-111*	2 kCi/wk
Y-88*	6 Ci/wk
Se-75*	6 Ci/wk

* - Taken from measured yield at 2 kW using IAC linac and scaled by power, mass, isotopic enrichment, geometric corrections, etc. Note that these rates are consistent with Segebade et al. and the Mo-99 estimates of TRIUMF.

So why hasn't Photon Activation been exploited?

In two words:

Specific Activity
(or lack thereof)

This is the Achilles heel of Photon Activation.

So why hasn't Photon Activation been exploited?

Example: ^{99}Mo –

The maximum Specific Activity of ^{99}Mo in a post-gamma-irradiated pure ^{100}Mo target is in the 1-10 Ci/g range (see Bennett, 1998 or TRIUMF, 2008).

But if one separates the ^{99}Mo from a post-irradiated pure ^{100}Mo target, the answer is very different. Advances in materials science have enabled us to produce

better than 1 kCi/g specific activity of ^{99}Mo.

Issues and Questions for Scale-up:

1) ISU experiments were done with 1-2 kW electron beams and 1 g targets: scale-up to 100 g targets at 100 or more kW electron beams raises significant questions about heat transfer, efficiency of separation, etc.

2) Costs, optimization, target designs, multi-isotope production, FDA approvals, NRC licensing (of the material, not the accelerators), and many others all pose challenges.

IF high specific activity can be produced from Photon Activation, shouldn't this be exploited?

Advantages:

1) No HEU, LEU or uranium of any kind ➔ no safeguards or proliferation risks.

2) No HLW. And no major nuclear wastes of any kind.

3) No major decommissioning costs associated with accelerator facility (when compared to reactor decommissioning costs).

4) Such a facility could simultaneously produce a large number of isotopes that are currently in short supply.

5) Such a facility(s) would diversify isotope production, research and education capacity and help insure a reliable supply.

Limiting ourselves to photon activation, for the moment, what can we make?

Via (γ, n): 64Cu, 26Al, 131Ba/131Cs, 88Y, 197Hg, 192Ir, 203Hg, 99Mo, 75Se, 11C, 13N, 15O, 18F

Via (γ, p): 67Cu, 47Sc, 177Lu, 90Y, 110mAg, 166Ho, 57Co, 77Se

Via (γ, 2n): 77Br, 67Ga, 7Be, 111In

Via (γ, np): 82mRb, 64Cu

Thank You.

- Acknowledgements:

This work was supported, in part, by a grant from the U.S. DoD.