PAUL SCHERRER INSTITUT

# Paul Scherrer Institut

Wir schaffen Wissen – heute für morgen

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# Nuclide inventory in proton irradiated lead – comparison of simulation and measurement

# Contents

- Motivation and overview of the PSI accelerator facility
- Pb-samples from the SINQ target4
- Experimental results
- Method of the activity calculation
- Method to gain statistics on residual nuclide distribution
- Results from MCNPX + Cinder'90v7.4.2 (FISPACT/EAF2007):
  BERTINI-Dresner-RAL, ISABEL-ABLA, CEM3.02
- Dose rates around SINQ target4
- Summary and Remarks

# Motivation

Components around the beam line get activated
... need to be removed eventually

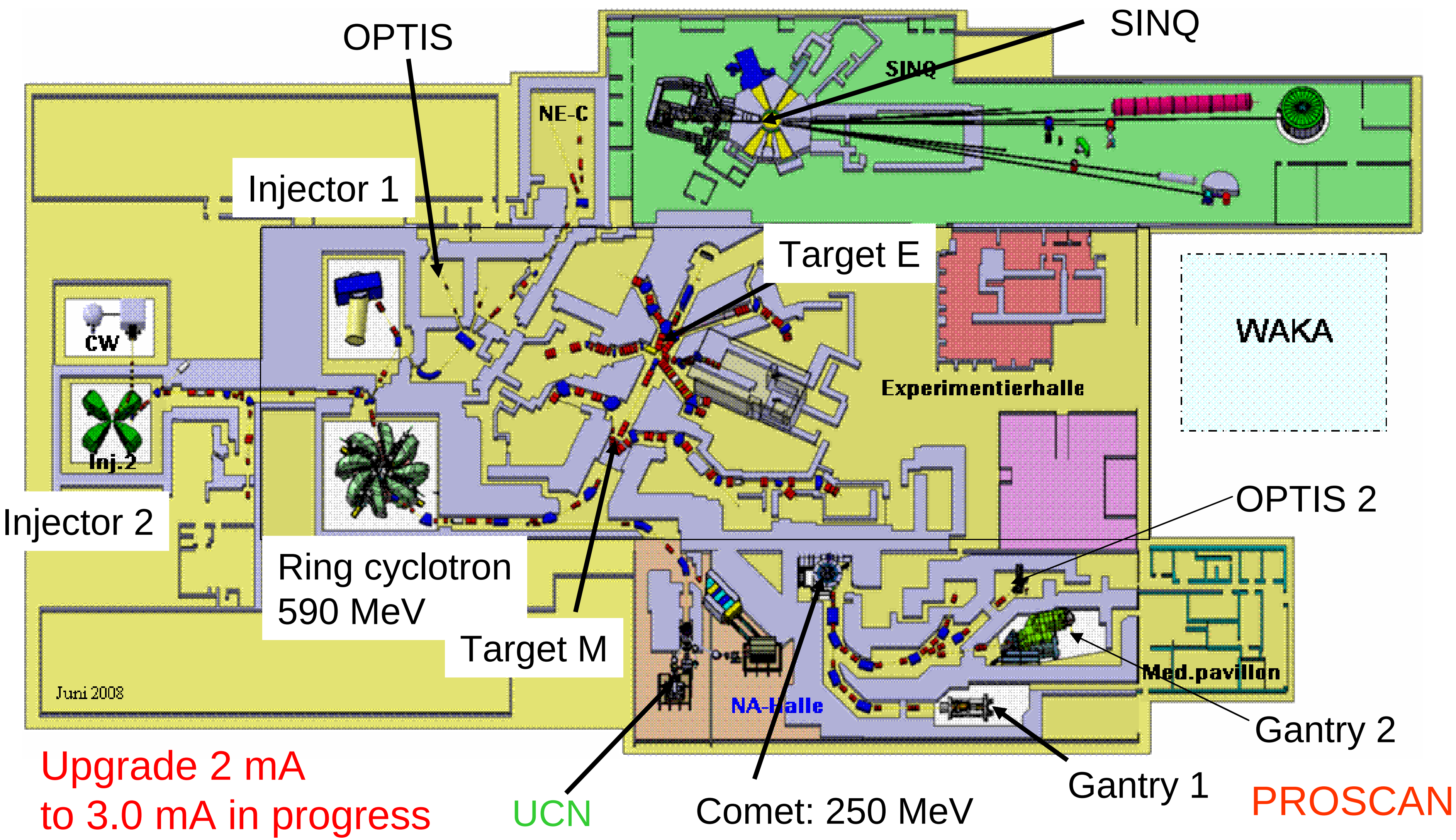
## Calculations are necessary

- for replacement → waste
  - Nuclide inventory needed before disposal as required by swiss authorities
  - periodical validations are required: comparison to experimental data

- for future (planned) installations/facilities:
  - estimation of the amount of total waste after operation
  - dose rates needed for construction of shielding

- for repair/dismantling → planning of work procedures
  dose rate estimation needed

# PSI Proton Accelerator Facilities



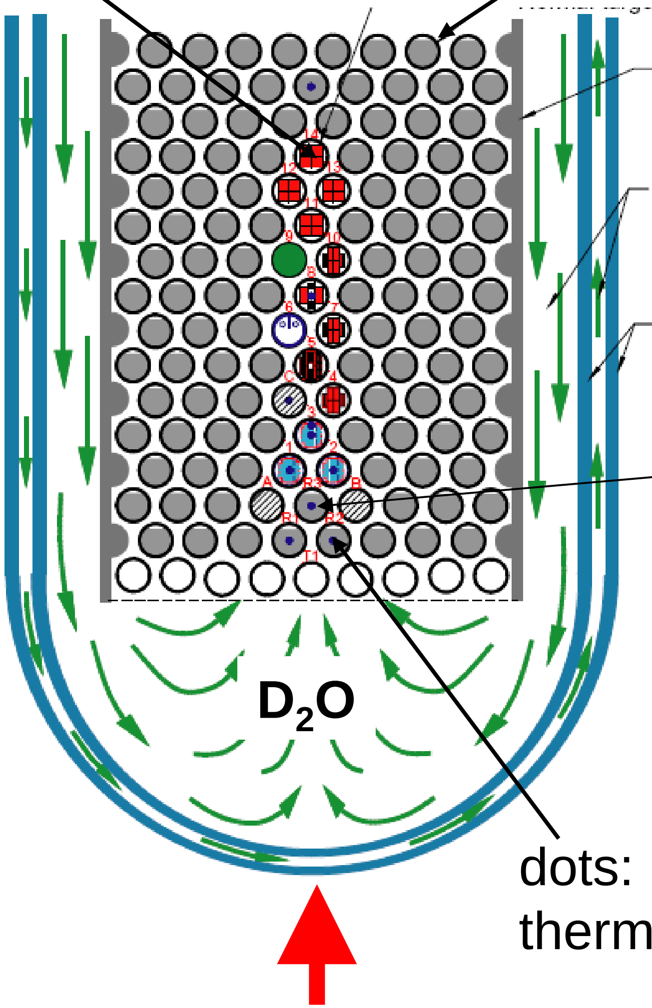

Upgrade 2 mA
to 3.0 mA in progress

# SINQ facility: neutron spallation source

Target 4: March 2000-Dec. 2001 → 10.03 Ah

Stip probe

Pb-rod with SS316L tube




Zr-2 frame

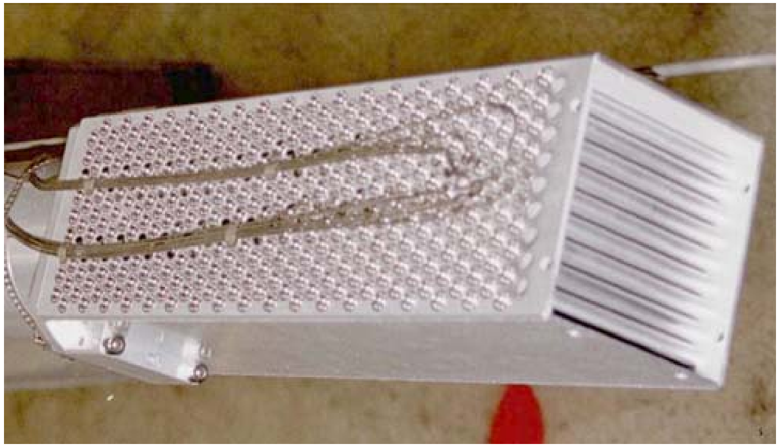
AlMg(3%) container

Rod3: Pb

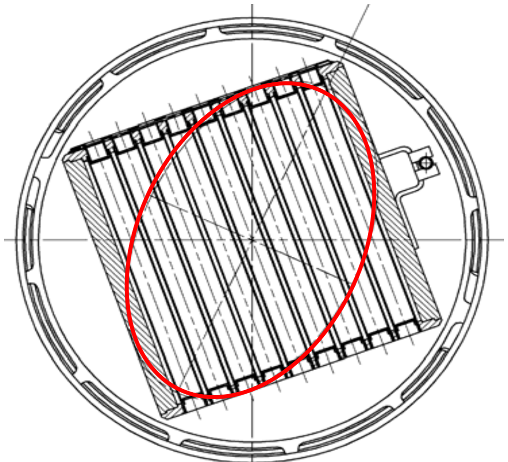
dots: thermocouple

proton beam: 590 MeV

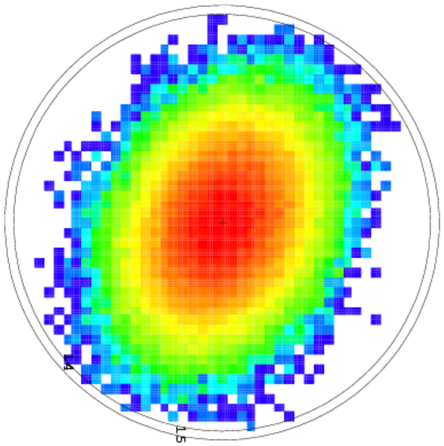
spallation target:



footprint on target: gaussian



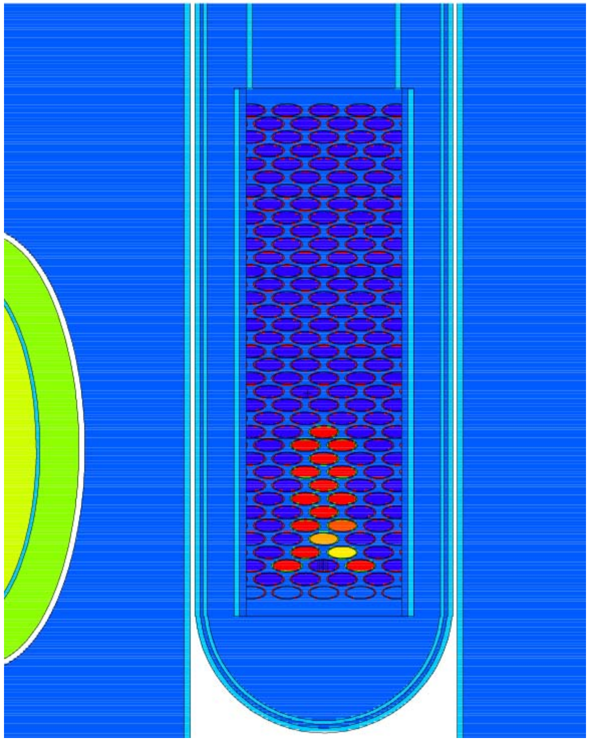
$\sigma_x$=2.1 cm, $\sigma_y$= 3.0 cm



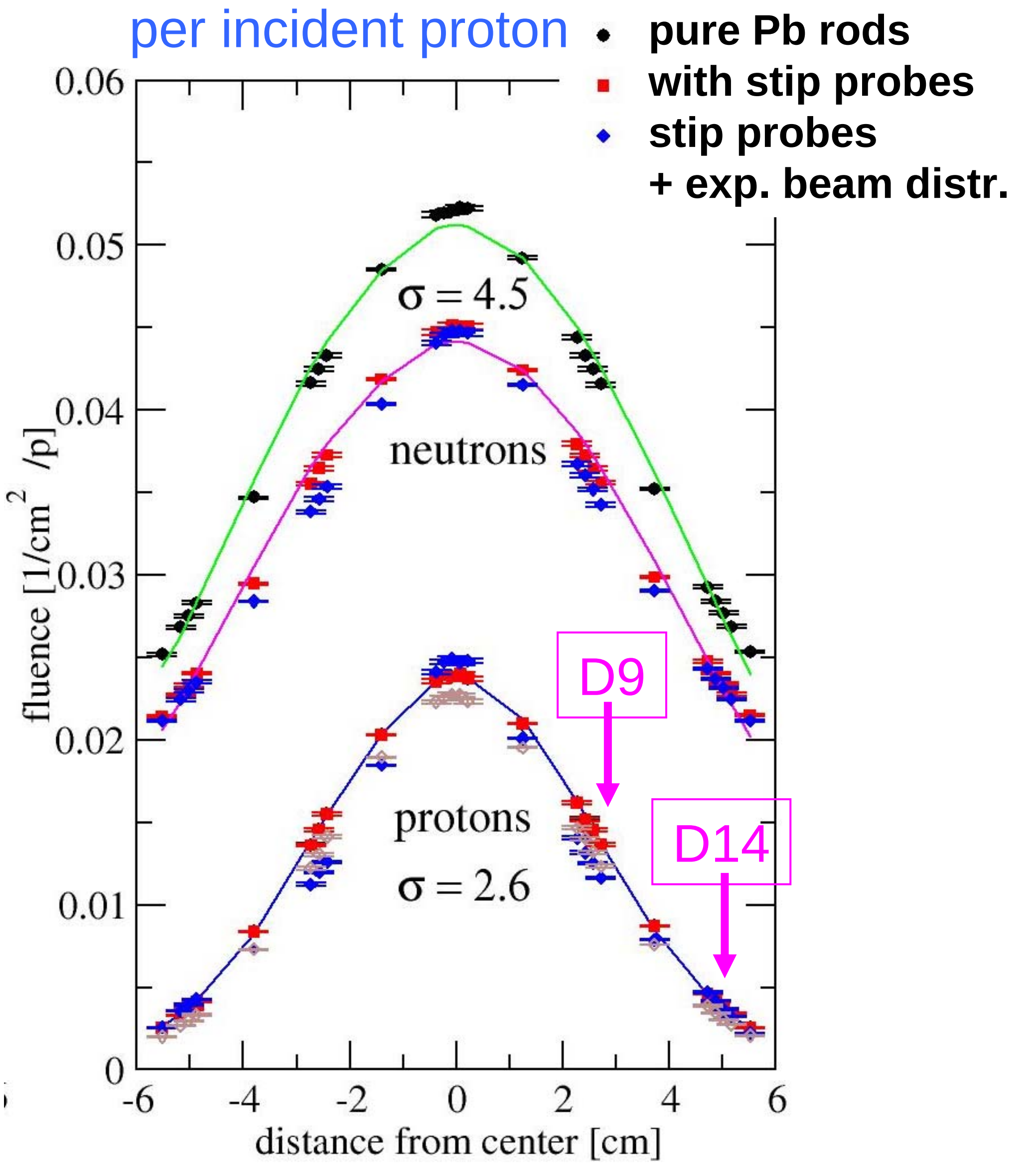
MCNPX

# Proton and neutron flux along Rod3 (in 2. row)

MCNPX: cut from model

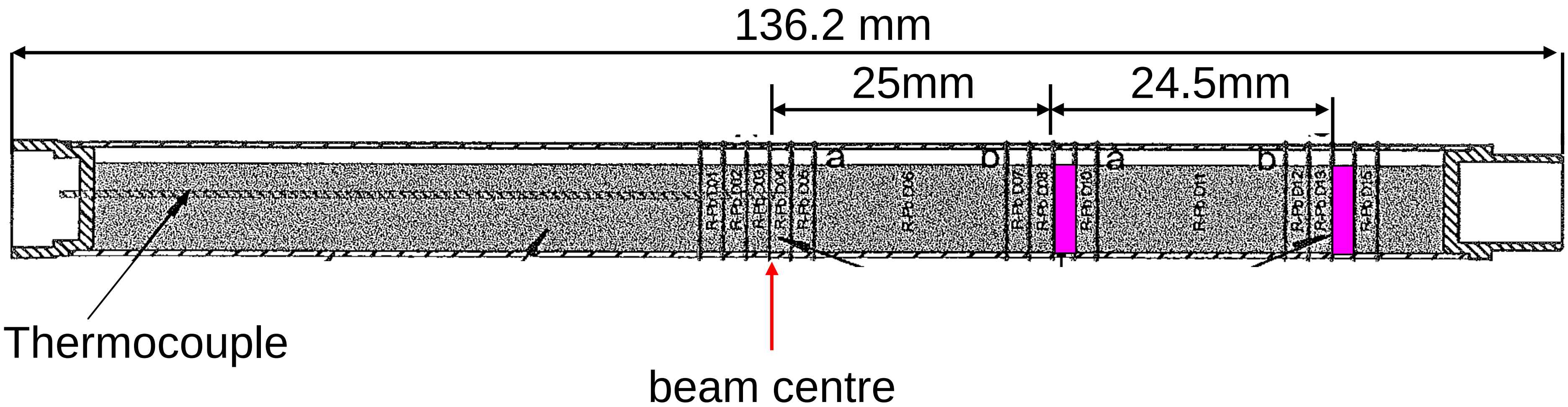


per incident proton




- Stip probes reduce flux by 20 %
→ included in the model
- experimental measured beam profile very close to gaussian
- neutron flux distribution is wider

# D9 and D14 Pb-samples in Rod3




sizes of samples: ∅= 10 mm, d = 1.5 mm
→ very small

radiochemistry uses small probes
→ high sensitivity of the method
→ less dose rate for working personal
→ sample well localized
←→Monte Carlo Method needs good statistics (especially for calc. of residuals)
→ computer power: 512 CPU, ~20 h x 3
→ biasing method: importances: 5
} no success: check with neighboring cells
→ larger regions to average over

# Results of the measurement (~Sept. 2006)

| | D9 [Bq/g] | D14 [Bq/g] | | D9 [Bq/g] | D14 [Bq/g] |
|---|---|---|---|---|---|
| $^{207}$Bi | $3.00 \cdot 10^{7}$ | $1.01 \cdot 10^{7}$ | $^{106}$Ru | $4.83 \cdot 10^{6}$ | $3.91 \cdot 10^{6}$ |
| $^{172}$Lu/$^{172}$Hf | $2.00 \cdot 10^{7}$ | $5.41 \cdot 10^{7}$ | $^{110m}$Ag | $1.29 \cdot 10^{6}$ | $3.93 \cdot 10^{5}$ |
| $^{173}$Lu | $2.76 \cdot 10^{7}$ | $4.30 \cdot 10^{7}$ | $^{125}$Sb | $1.32 \cdot 10^{6}$ | - |
| $^{194}$Au/$^{194}$Hg | $1.86 \cdot 10^{7}$ | $3.13 \cdot 10^{6}$ | $^{133}$Ba | $2.8 \cdot 10^{6}$ | $7.94 \cdot 10^{5}$ |
| $^{102m}$Rh | $5.53 \cdot 10^{6}$ | $1.44 \cdot 10^{5}$ | $^{44}$Sc/$^{44}$Ti | $8.00 \cdot 10^{4}$ | $2.84 \cdot 10^{4}$ |
| $^{202}$Tl/$^{202}$Pb | $4.80 \cdot 10^{5}$ | $1.87 \cdot 10^{5}$ | $^{108m}$Ag | $3.75 \cdot 10^{5}$ | $1.56 \cdot 10^{4}$ |
| $^{60}$Co | $3.67 \cdot 10^{6}$ | $1.40 \cdot 10^{6}$ | $^{194m}$Ir | $2.61 \cdot 10^{4}$ | - |
| $^{54}$Mn | $2.29 \cdot 10^{5}$ | $7.01 \cdot 10^{4}$ | $^{26}$Al (AMS) | 0.5 | 0.2 |
| $^{58}$Co | $1.55 \cdot 10^{6}$ | $9.47 \cdot 10^{5}$ | $^{36}$Cl (AMS) | $9.5 \cdot 10^{1}$ | $4.8 \cdot 10^{1}$ |

AMS: Accelerator mass spectrometry at ETH Zürich
very sensitive → low activity measurement

# Calculation of the Activation: Method

Monte Carlo particle transport program **MCNPX** : n,p,$\gamma$,$\alpha$,$\pi$,d,$^3$H....

Input: dedicated geometry, material compositions,
cross sections: for n<20MeV ENDF-B-VI.6, otherwise models

Output: n-fluxes (E<20 MeV), residual nuclei production rates

via Perl script

decay codes: **Cinder'90** or **SP-Fispact** or **Orihet3**

Input: irradiation and cooling history
cross sections: Cinder library, EAF 2007
decay properties of isotopes

Output: nuclide inventory, photon rate

via Perl script

used as $\gamma$–source in **MCNPX** → dose rate

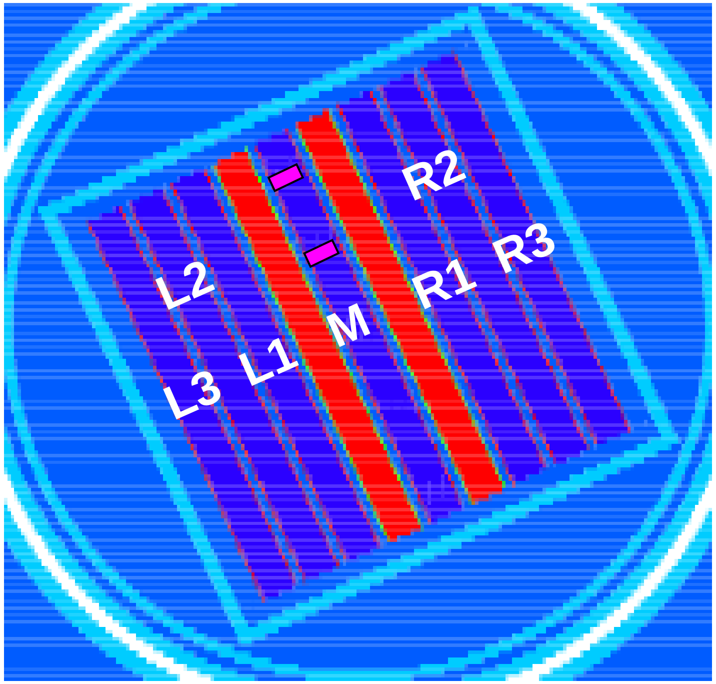
M. Wohlmuther et al., Proc. AccApp *2007*, Pocatello, p. 226.
F.X. Gallmeier, dito, p. 207

# Physical models used in MCNPX

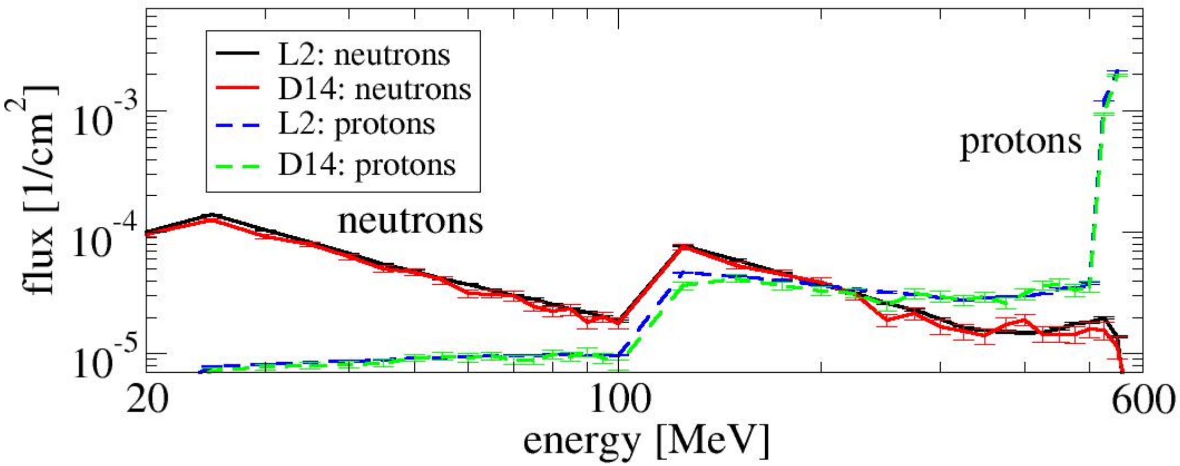
- BERTINI (INC) + Dresner (evaporation) + RAL (fission): MCNPX2.5.0 default option in MCNPX
  Bertini: based on Serber's model, spacelike realization
  Dresner: EVAP-A, based on Weisskopf's statistical model
  RAL (Atchison): fission in between of the evaporation stages
- ISABEL (INC) + ABLA (evaporation + fission): MCNPX2.5.0
  Isabel: primary particle is followed through the nucleus, classical collisions
  based on Serber's model, timelike realization
  ABLA (old version): break-up, fission,
  particle emission (based on Weisskopf-Ewing)
- CEM3.02: MCNPX2.7.c
  - based on the standard (non-time dep.) version of the Dubna cascade
  - three stages: INC + pre-equilibrium + evaporation/fission
  - recent on-going developments (Mashnik et al.)
    e.g. improved decription of complex particle production

# Method applied to gain more statistics

- sum up the residual nuclide production rates $P_R$ from larger regions (statistics for n-fluxes (< 20 MeV) very good)
- scale with the flux in D-samples




Comparison of the n- and p- fluxes for D14 and L2

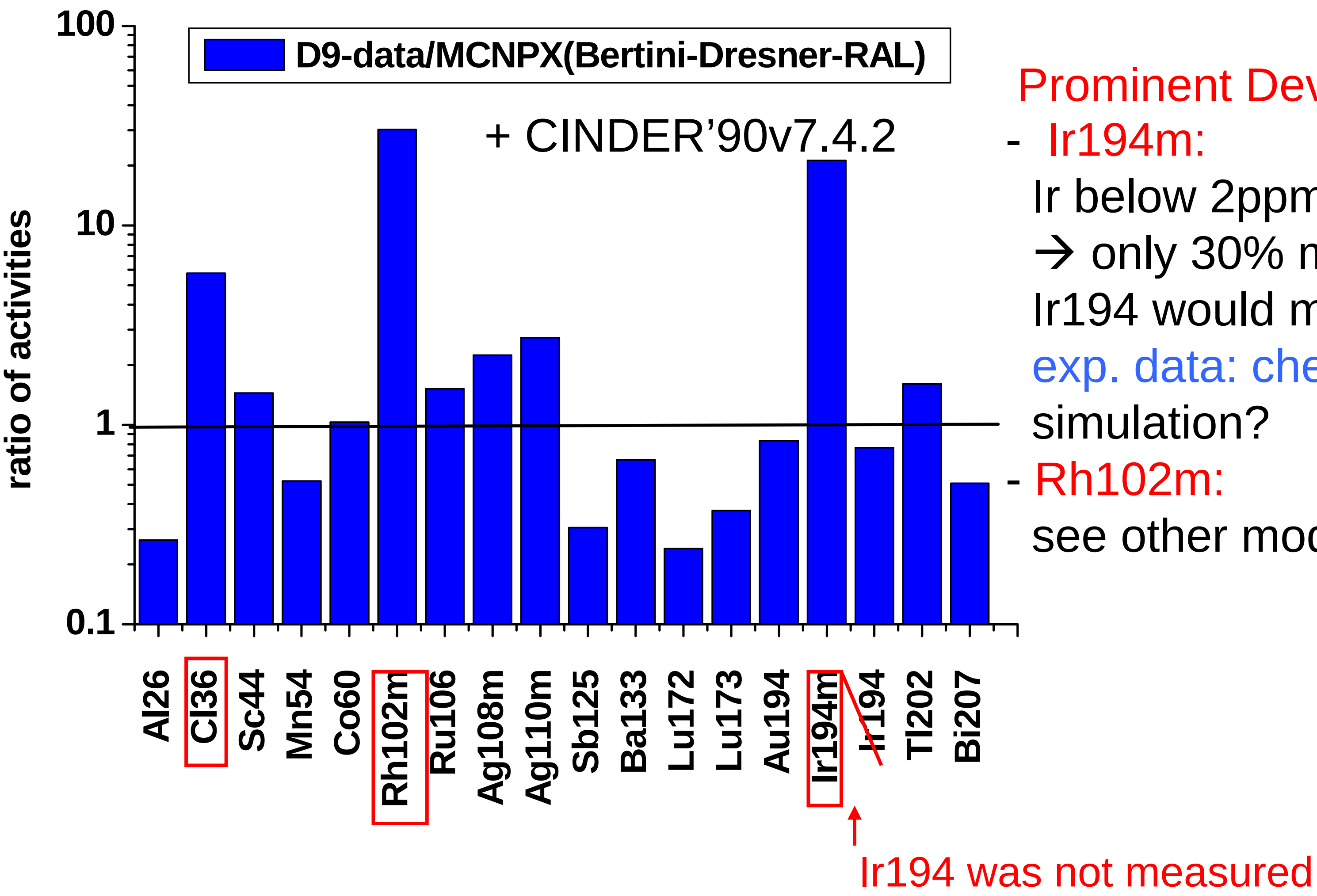



$$P_R(D14) = P_R(L2+R2)\ \frac{\text{p-flux}(D14)\ V(D14)}{\text{p-flux}(L2+R2)\ V(L2+R2)}$$

$$P_R(D9) = P_R(M)\ \frac{\text{p-flux}(D9)\ V(D9)}{\text{p-flux}(M)\ V(M)}$$

$$\frac{V(M)}{V(D9)} = 80$$

# Results for D9-sample: BERTINI-Dresner-RAL

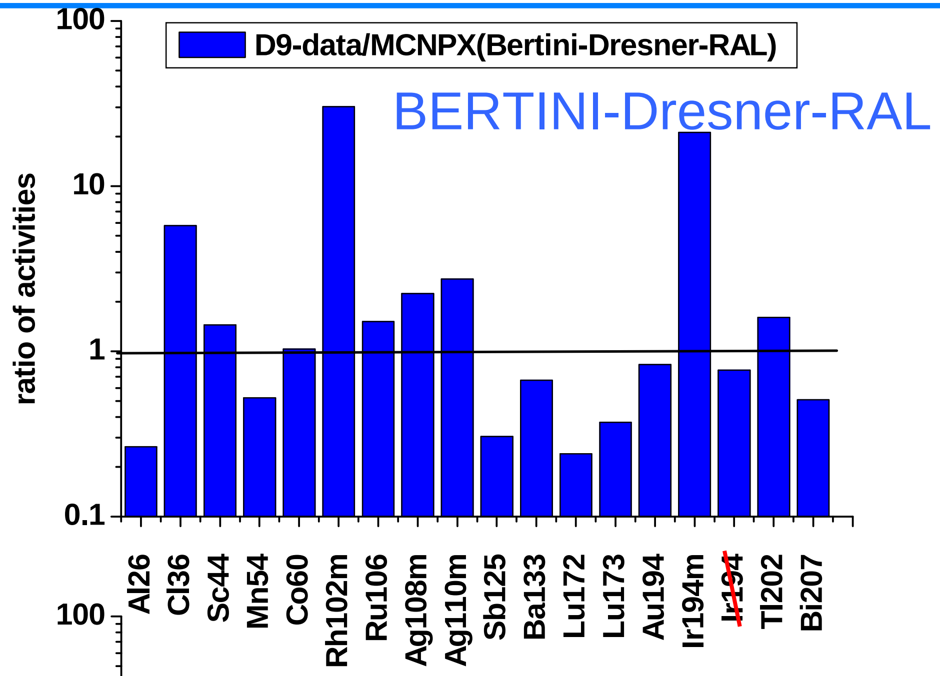


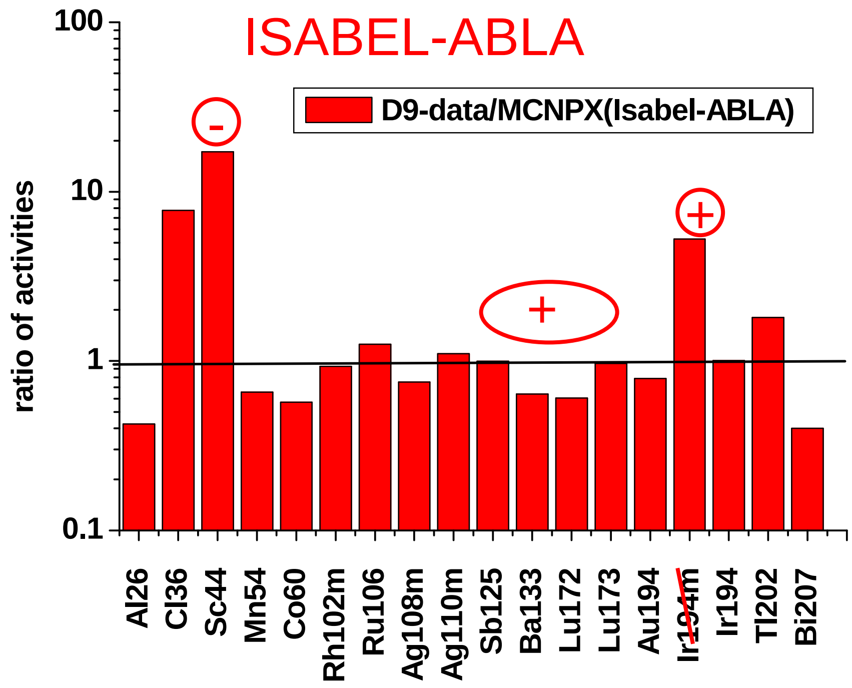

Prominent Deviations:

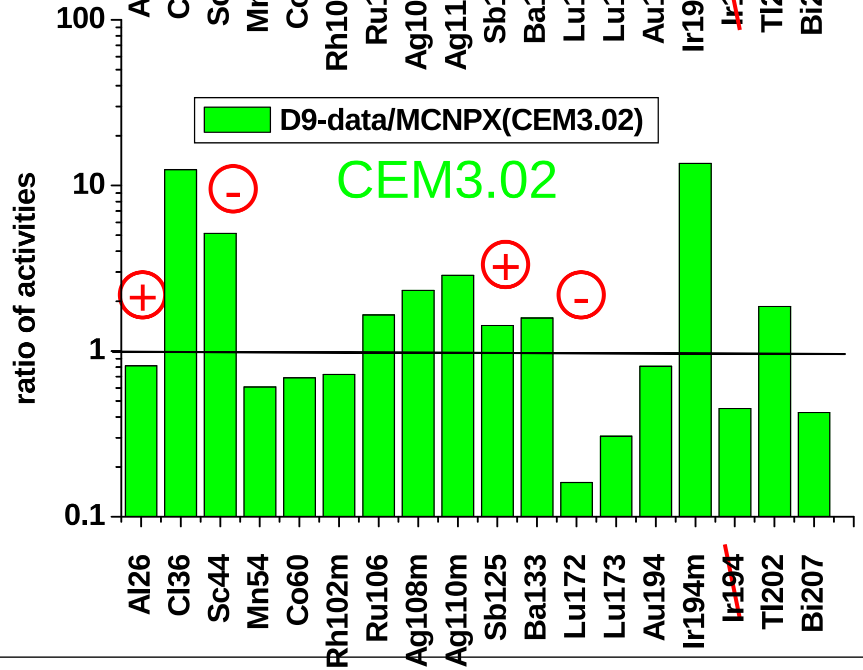
- Ir194m:
  Ir below 2ppm detection limit
  → only 30% more production
  Ir194 would match
  exp. data: checked, no mix-up
  simulation?
- Rh102m:
  see other models

- Cl36: main production via Cl35(n,$\gamma$)Cl36 but missing in material definition
  reason: cannot be detected by applied method (ICP-OES via dissolved probe)

# Results for D9-sample: different models



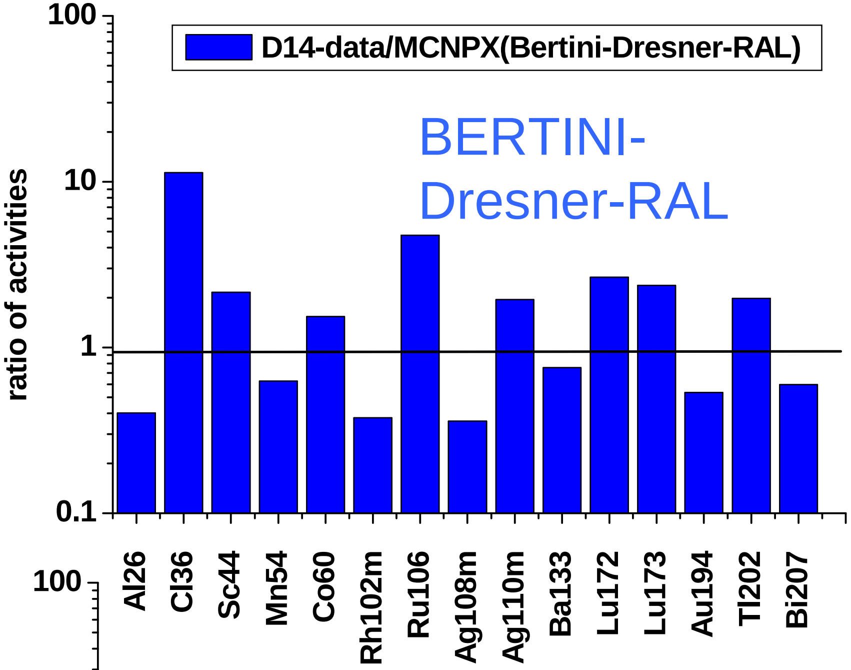

ISABEL-ABLA:

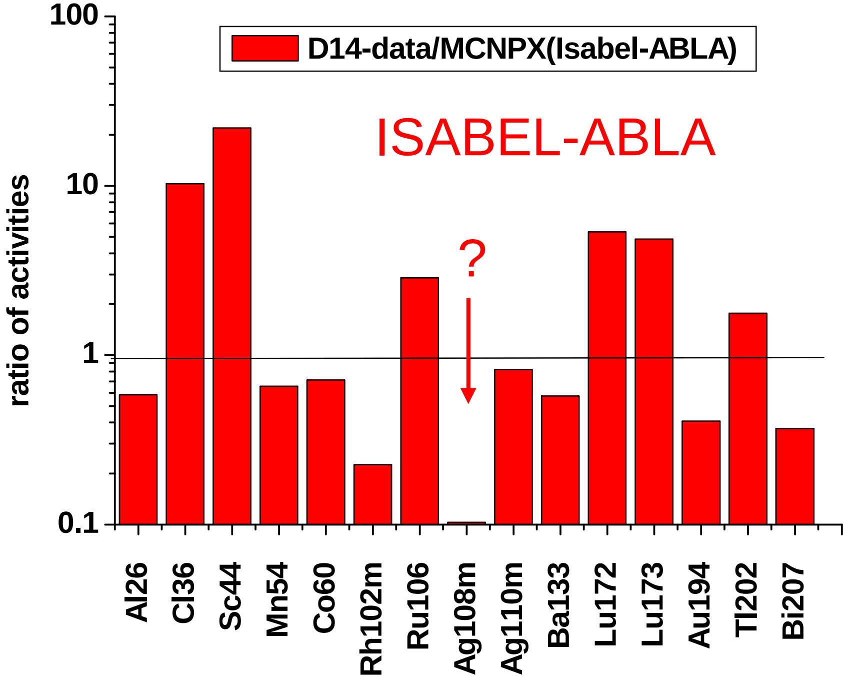
- improvement in the deep spallation zone as well as for Rh102m
- understimation of Sc44

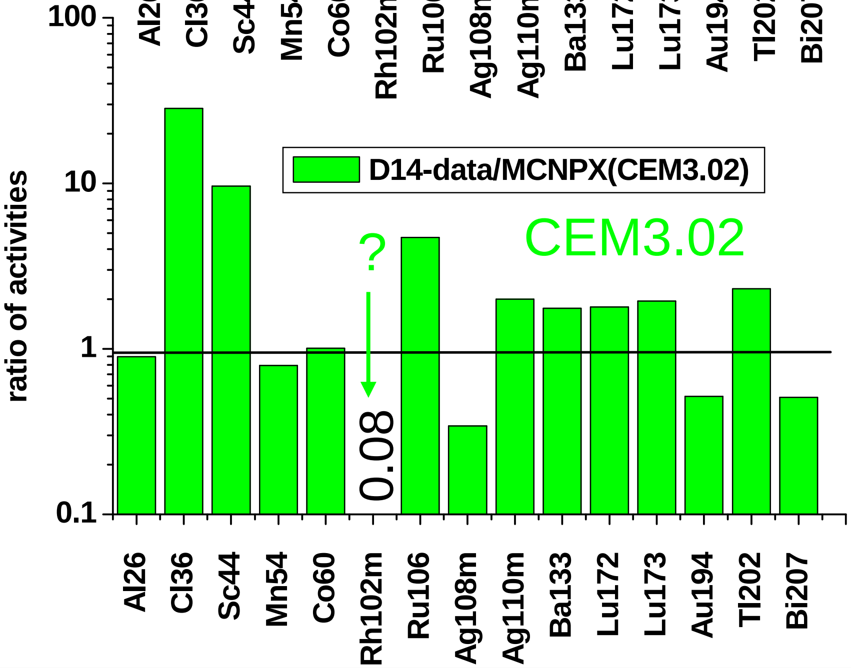
CEM3.02:

- improvement for Al26, Rh102m
- overestimation of Lu172/173

# Results for D14-sample: different models




ISABEL-ABLA:

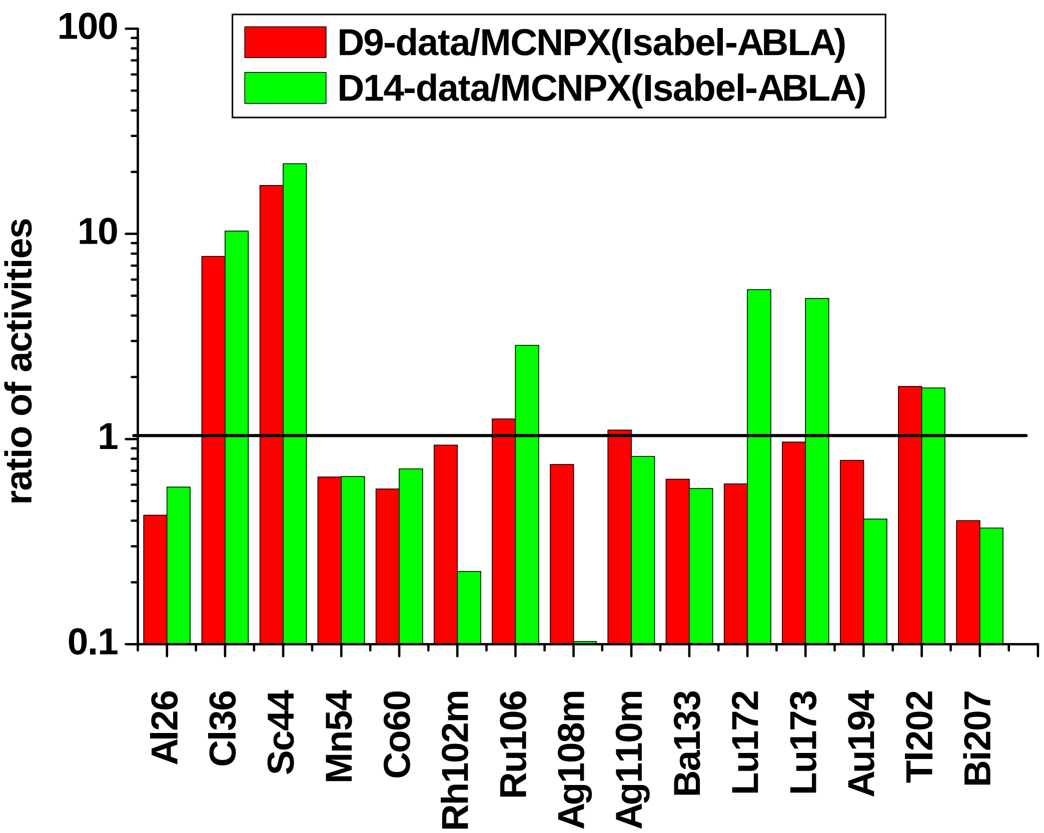
-large deviation for Ag108m

-understimation of Sc44

CEM3.02:

- improvement for Al26
- understimation of Sc44, Rh102m

# Comparison D9 and D14-sample: Isabel-ABLA



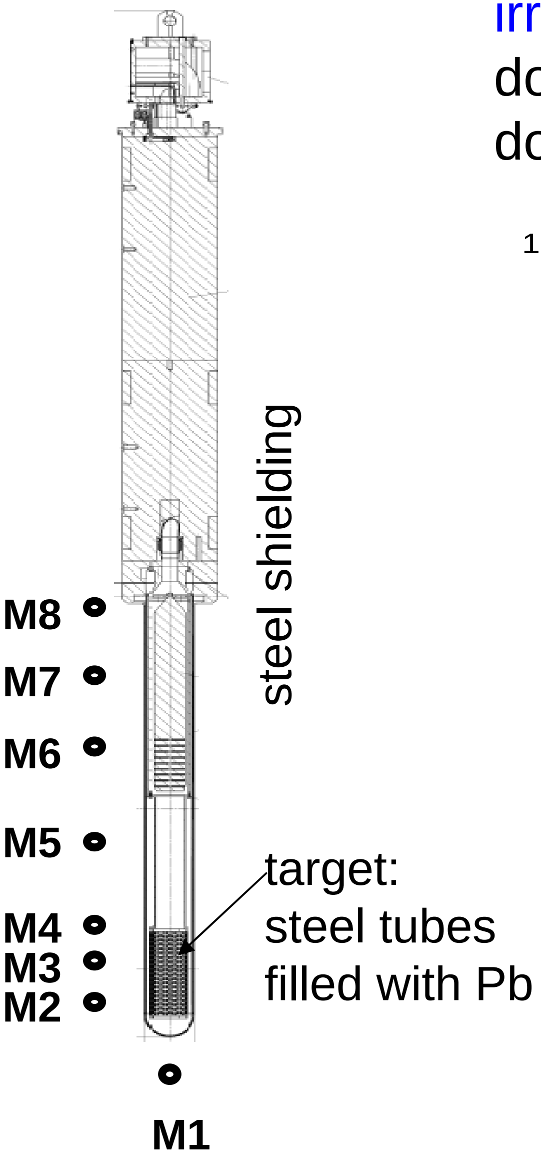

for most isotopes consistent predictions except for: Rh102m, Ag108m, Lu172, Lu173

different composition of n- and p-flux in D9 and D14

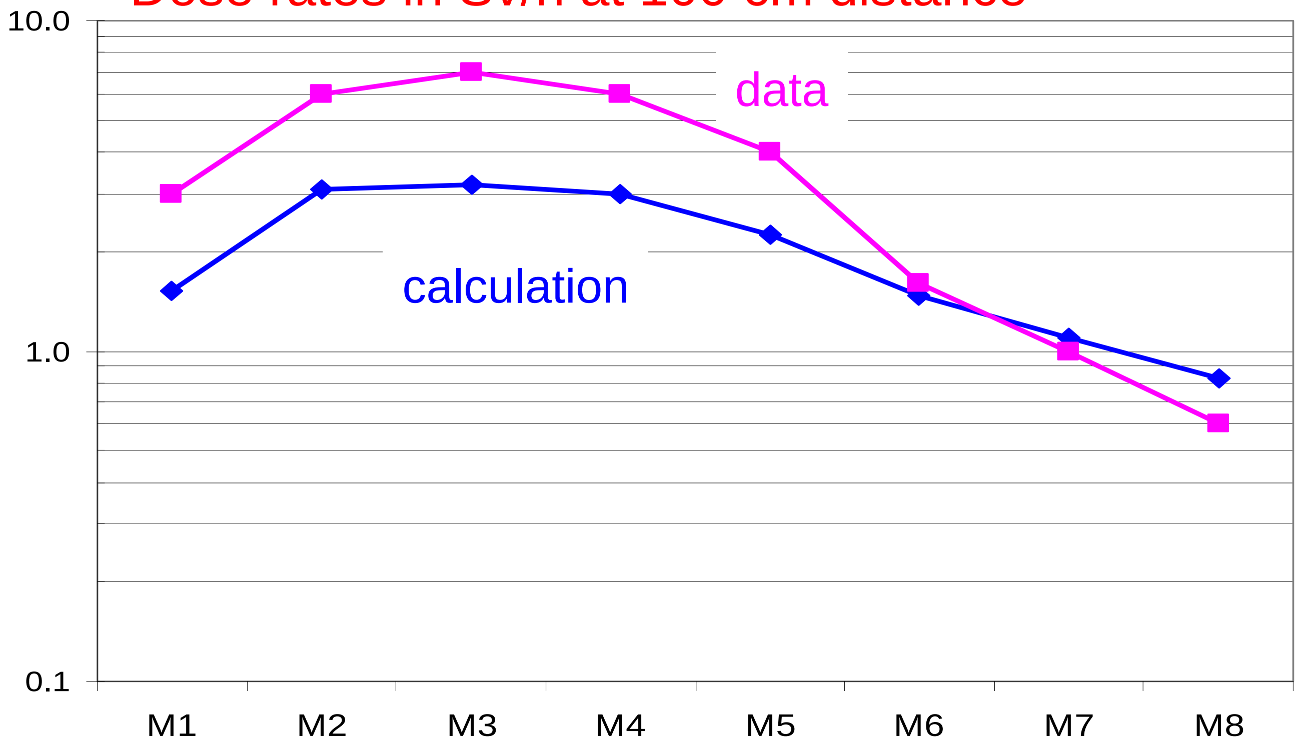
| | p-flux | n-flux>20 MeV | sum | n-flux<20 MeV |
|---|---|---|---|---|
| D9 | 0.014 | 0.0027 | 0.0167 | 0.035 |
| D14 | 0.0036 | 0.0013 | 0.0049 | 0.023 |

# Dose rates at SINQ Target 4

irradiation: 2 years with 10 Ah, cooling: 10 months
dose rates obtained at 3cm, 30cm, 100cm
dose rates calculated with MCNPX + Cinder'90




Dose rates in Sv/h at 100 cm distance




- deviations within a factor of 2
- more precise predictions require finer devision of regions

# Summary and Remarks

- Activity of 2 Pb samples from SINQ target4 were determined experimentally
- D9 and D14 have different compositions of p- and n-fluxes (> and < 20 MeV)
- Statistics gained by using average residual distribution in neighbouring rods with very similar n- and p-fluxes

  remark:

  nuclide inventory (activities) are almost the same if all rods are used, even the p-fluxes are different (n-fluxes are quite similar)

  reason:

  cross sections are almost flat for > 200 MeV

  exceptions:

  for nuclides close to the target nucleus → direct reactions

  → < 20 % deviation for A>194

- Comparison of measured activity to MCNPX + CINDER'90v7.4.2 using:
  - BERTINI + Dresner + RAL
  - ISABEL + ABLA
  - CEM3.02

  } should be preferred (ISABEL + ABLA, CEM3.02) remark: FISPACT/EAF07 coupled to MCNPX/CEM3.02

Lu172: 2 x less

Lu173: 3 x less

Ti44/Sc44: 30 % less

no Ir194m produced

- Dose rates at SINQ target4:
  - good agreement with data within a factor of 2 at d = 100 cm
  - at shorter distance regions have to be subdivided due to inhomogeneous activitation

# Material composition of Pb

| element | Pb (w-ppm) |
|---|---|
| Na | 28 |
| Mg | 0.4 |
| Al | 1.5 |
| P | 17.5 |
| S | 16.5 |
| Ca | 2.6 |
| Ti | 0.4 |
| Cr | 1.8 |
| Fe | 9.2 |
| Ni | 6 |
| Cu | 467 |
| Zn | 5.8 |
| Ge | 24.1 |
| Mo | 2.5 |
| Ag | 45.4 |
| Sn | 130 |
| Sb | 1039.5 |
| Ba | 0.3 |
| Tl | 4 |
| Pb | 998000 |
| Bi | 220.5 |

> 50ppm: Sn,Bi, Cu, Sb
+ several traces and impurities
→ difficult to get statistics

Modified version of MCNPX2.5.0:
active card (by F. Gallmeier)
for cross sections from models (like htape)
→ 1) particle is scattered at all isotopes of the material definition for each event
2) weigthing according to presence are applied
→ helps to gain statistics if impurities are present and important

drawback: old version

material analysis from ICP-OES at PSI