IAEA 60 Years
Atoms for Peace and Development

TALYS for medical isotope production

Arjan Koning, IAEA

Joint ICTP-IAEA Workshop on Simulation of Nuclear Reaction Data with the TALYS Code, October 16 - 20 2023, Trieste

Introduction

- Medical isotopes from nuclear data point of view
- Global assessment of experimental data and outlier assignment
- Consistent and automated parameter fitting
- Some challenges for medical isotopes and its impurities
- Special case: Th232(p,x)Ac225
- Conclusions

Currently declared as IAEA medical isotope evaluations

- 21 (p,n) reactions
- 15 (p,2n) reactions
- 7 (p,3n) reactions, etc.
- 4 (d,n) reactions
- 7 (d,2n) reactions
- 5 (d,3n) reactions etc.
- + residual products further away from the target
- Total: about 140 reaction channels

IAEA Medical portal

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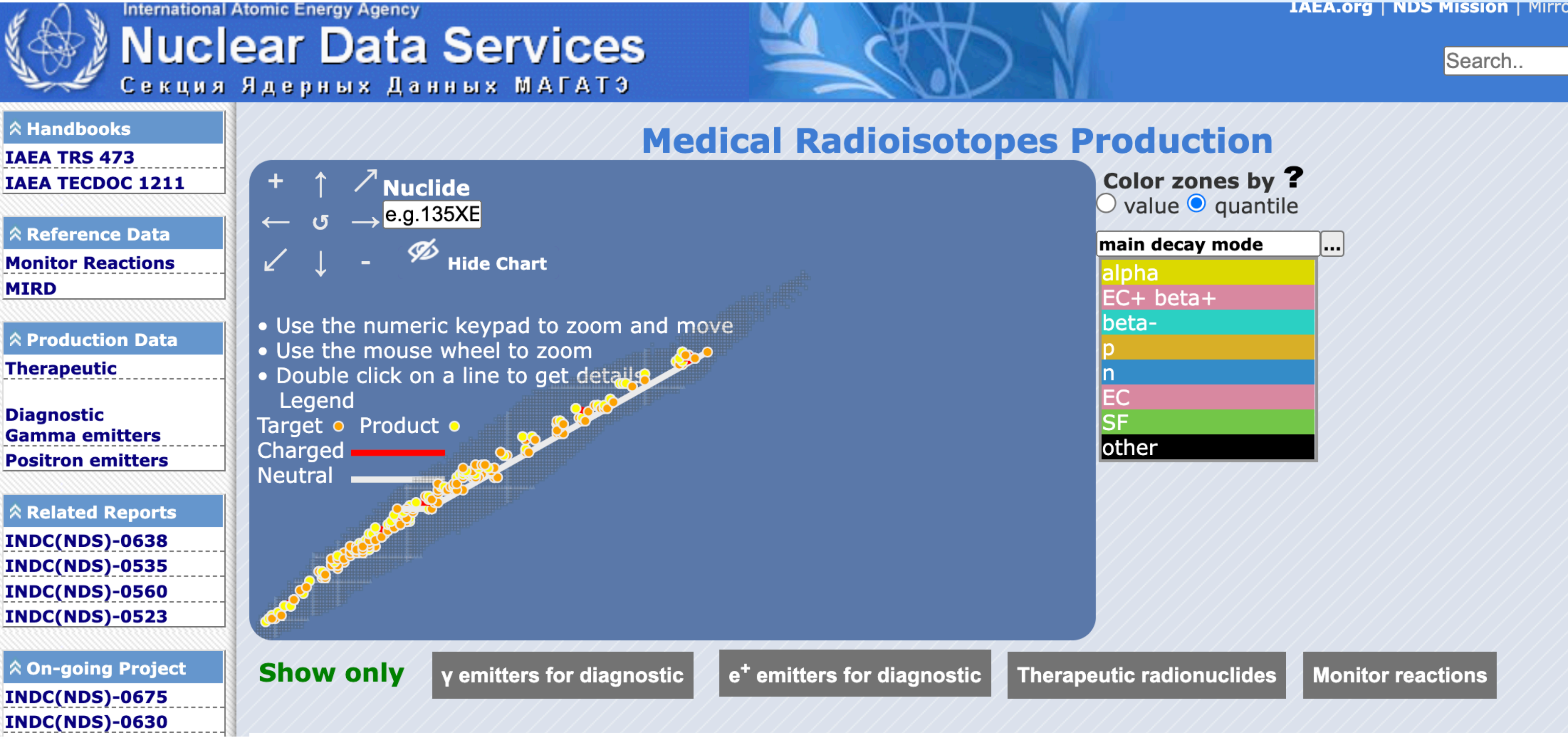
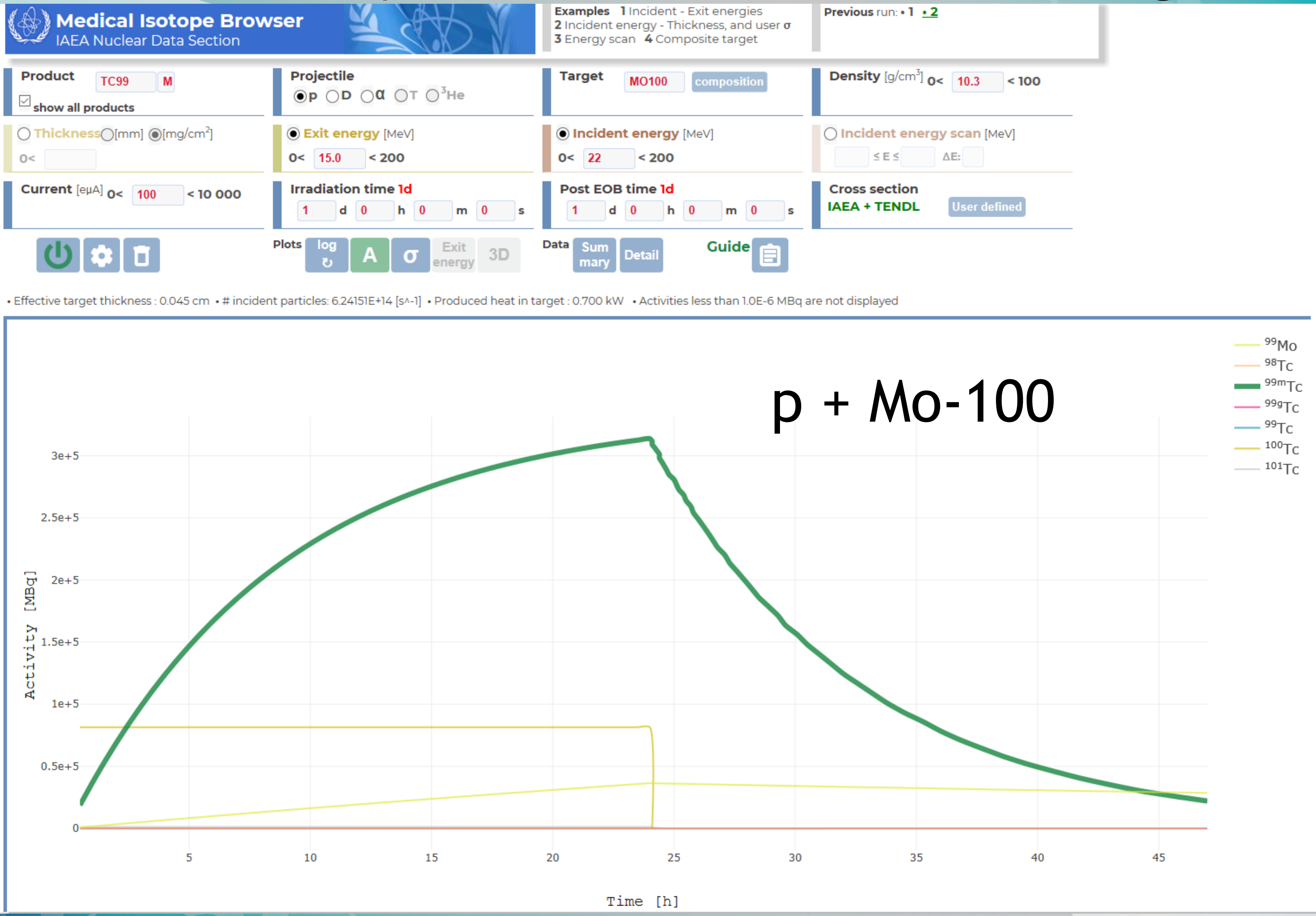


Medical isotope browser: nds.iaea.org/mib



https://nds.iaea.org/talys/

TALYS-Related Software and Databases

TALYS and the TALYS-related packages are open source software and datasets (GPL License) for the simulation of nuclear reactions.

TALYS

Arjan Koning, Stephane Hilaire, Stephane Goriely

Nuclear reaction model code.

- Download TALYS-1.96
- Download previous versions
- Read Tutorial

Created at NRG, cea, ULB Université Libre de Bruxelles, IAEA International Atomic Energy Agency

EXFORTABLES

Arjan Koning

Experimental nuclear reaction database based on EXFOR.

- Download EXFORTABLES-1.0
- Read Tutorial

RESONANCETABLES

Arjan Koning, Dimitri Rochman

Database for thermal cross sections, MACS and average resonance parameters.

- Download RESONANCETABLES-1.0
- Read Tutorial

Created at IAEA International Atomic Energy Agency, Paul Scherrer Institut PSI

Zero-ing in on the truth

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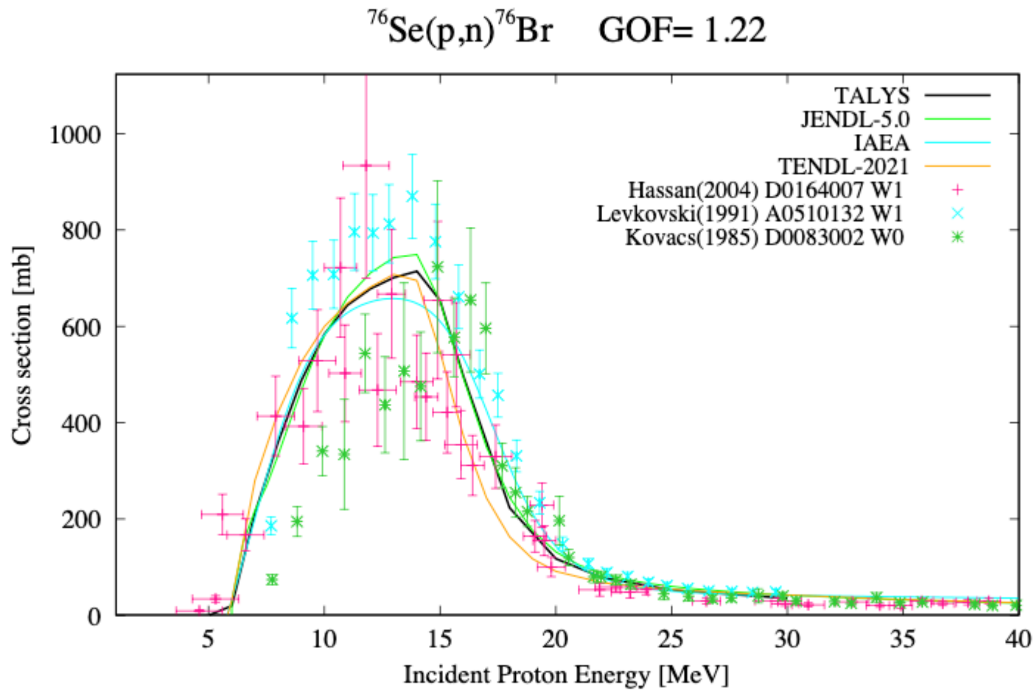
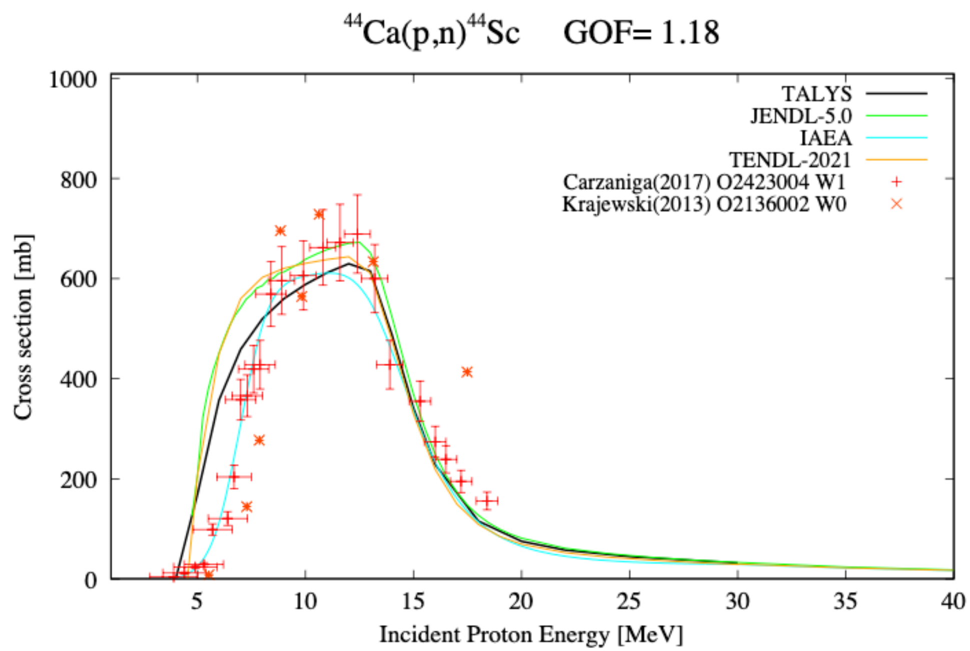
- Run TALYS for all projectiles, nuclides and energies with global settings
- Compare with the **entire** EXFOR database
    - Computational access to EXFORtables: directory-structured database with E-dE-xs-dxs data per measurement (from XC5 file, Viktor Zerkin)
    - Automatic normalisation to new monitor and decay data
    - Assign outliers in EXFOR (Exforcism)
        - comparison with nuclear data libraries,
        - comparison with TALYS
        - comparison with other experimental data sets
        - quantify historical evaluator's opinion in consistent metadata
- Assess predictive power of TALYS as a function of energy, reaction channel and mass range
- Zoom in on specific reaction channel with automated optimisation, varying a restricted set of TALYS parameters



W0: outlier-by-eye

Challenge: EXFOR data may be hidden under different reaction identifiers. Completeness of experimental data from EXFOR to EXFORtables not ensured, some data sets may be missing.



Weighting data sets in EXFOR

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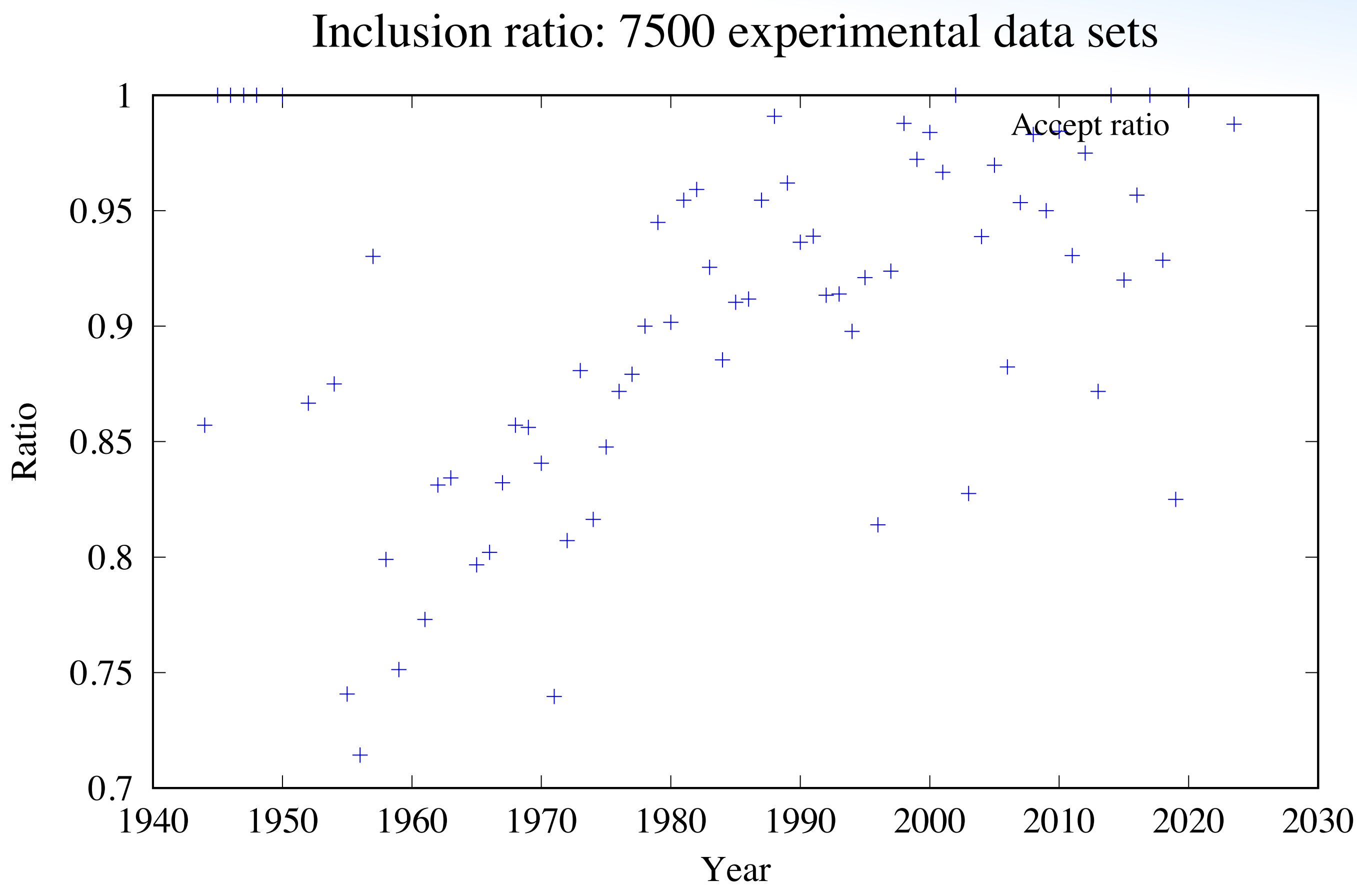
- Weights for ~28000 EXFOR subentries
  - Natalia Dzysiuk for activation c.s. + (all c.s.) Ni: 2336 subentries
  - Erwin Alhassan for proton induced reactions: 166 subentries
  - Natalie Gaughan for proton induced reactions: 103 subentries
  - Arjan Koning for neutron activation cross sections: NEA/DB/DOC(2017)1: 25850 subentries, based on automated chi^2
  - Arjan Koning (2022) for neutron up to alpha-induced cross sections - visual outlier assignment: 8000 subentries
  - Available in JSON files, including all comments etc.

8400 JSON outlier/inlier files, one per EXFOR subentry


```
{
    "Subentry"     : "A0001004",
    "Author"       : "Skakun",
    "Year "        : 1975,
    "Projectile"   : "p",
    "Target Z"     :   48,
    "Target A"     :  111,
    "Target state": "0",
    "X4 Reaction" : "48-CD-111(P,N)49-IN-111,,SIG",
    "Evaluations" :
    [
        {
        "Evaluator"    : "Arjan Koning",
        "Date"         : "2022-06-05",
        "Weight"       :    0,
        "Comment"      : [
                         " Excluded from evaluation: graphical outlier"
                         ]
        },
        {
        "Evaluator"    : "Erwin Alhassan",
        "Date"         : "2019-11-08",
        "Weight"       :    0,
        "Comment"      : [
                         " Erwin Alhassan (PSI, 2018)  0",
                         " (1 -> accept and 0 -> reject)",
                         " Reasons for inclusion/exclusion",
                         " 1) Experimental data set not consistent with other experiments such as Takacs (2005) between about 10 - 15
 MeV (The cross sections are systematically lower)"
                         ]
        },
        {
        "Evaluator"    : "Natalie Gaughan",
        "Date"         : "2019-03-15",
        "Weight"       :    1,
        "Comment"      : [
                         " IAEA-TECDOC-1211 - Data selected"
                         ]
        }
    ]
}
```

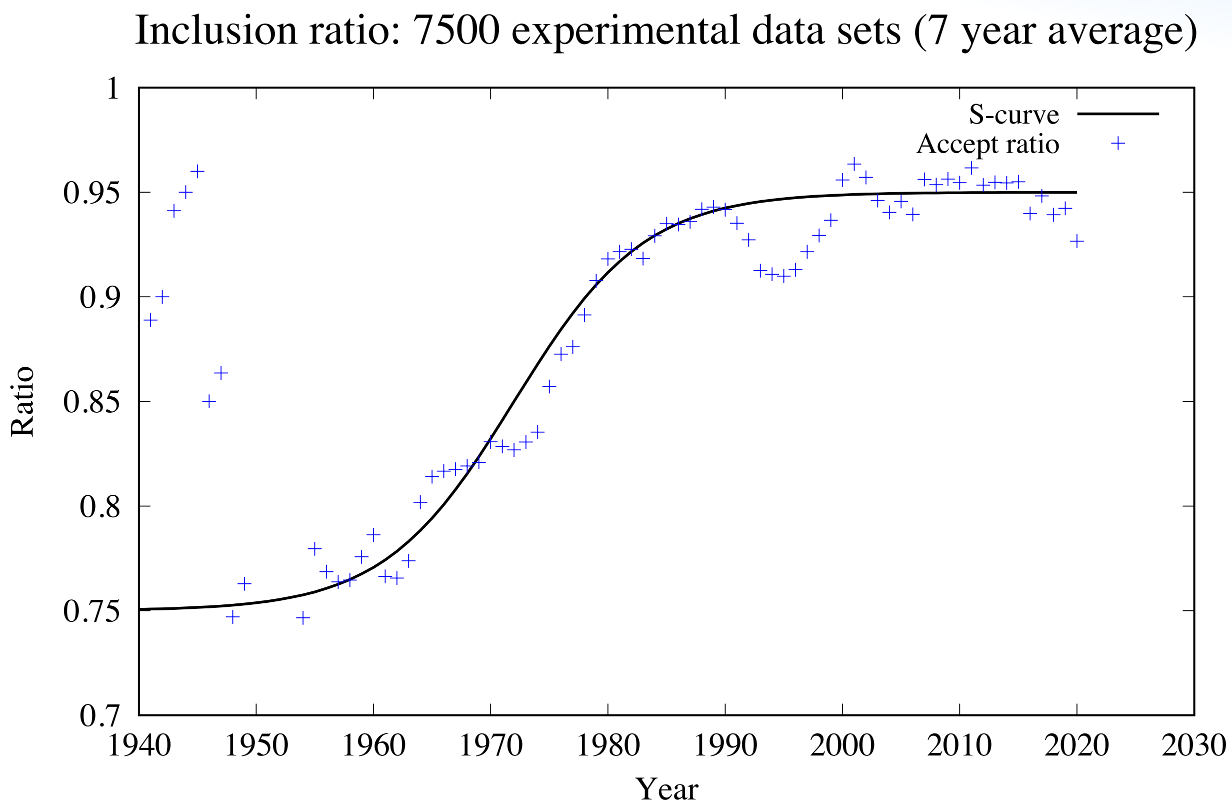

So far: 1 user

EXFOR outlier assignment



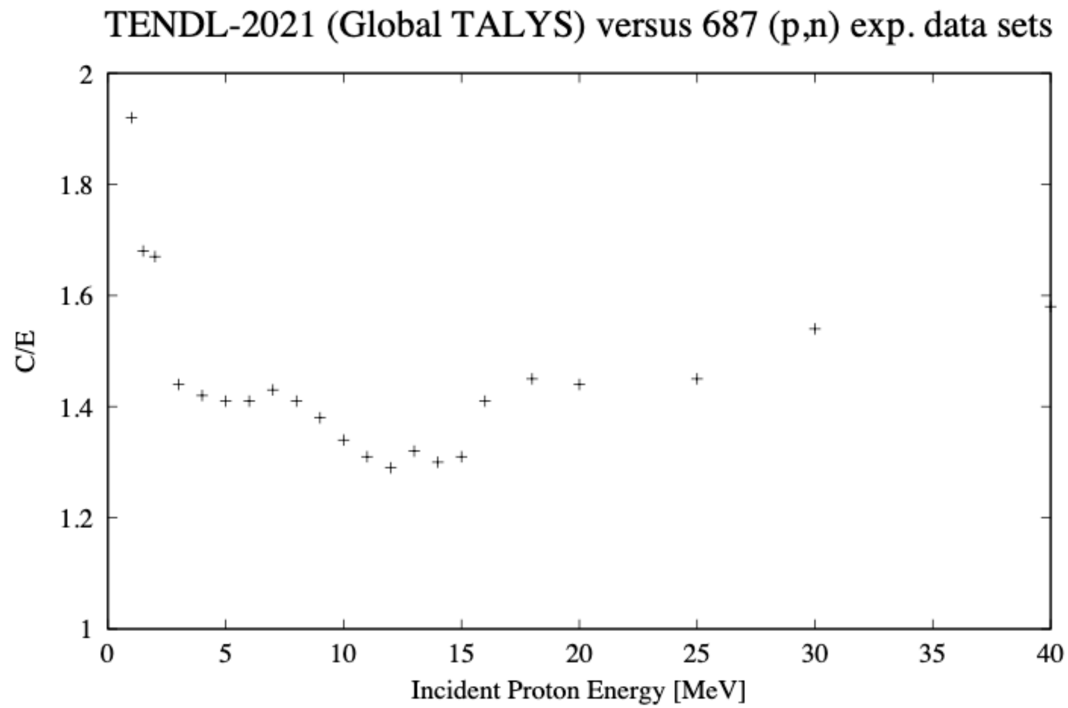
Summed over all (n,g), (n,f), (n,n'), (n,2n), (n,p), (n,a), (p,n), (g,n), (a,n), (d,n) reactions we could mine from EXFOR. 6500 accepts, 1000 rejects

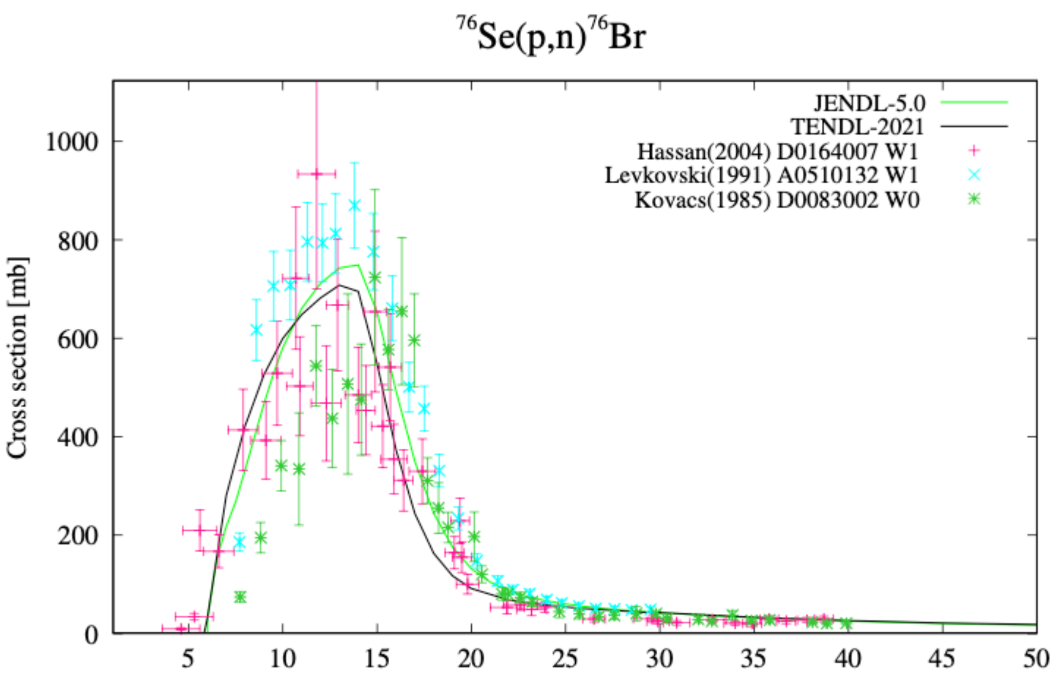
EXFOR outlier assignment: a learning curve?



Other analyses possible:

- per reaction channel
- per author, co-author, lab, etc (probably should not publish THAT)
- per incident energy (e.g. 14 MeV)
- re-insert this as prior in the next Bayesian update





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Global predictive power for (p,n): ~30% around the peak

(p,2n): ~40%
(alpha,n): ~45%
(d,n): ~60%

Common trend for all threshold reactions. Relative deviation is
- Large near threshold (> 2)
- Small near peak
- Larger in the tail

Global predictive power is energy dependent

Systematic approach for TALYS

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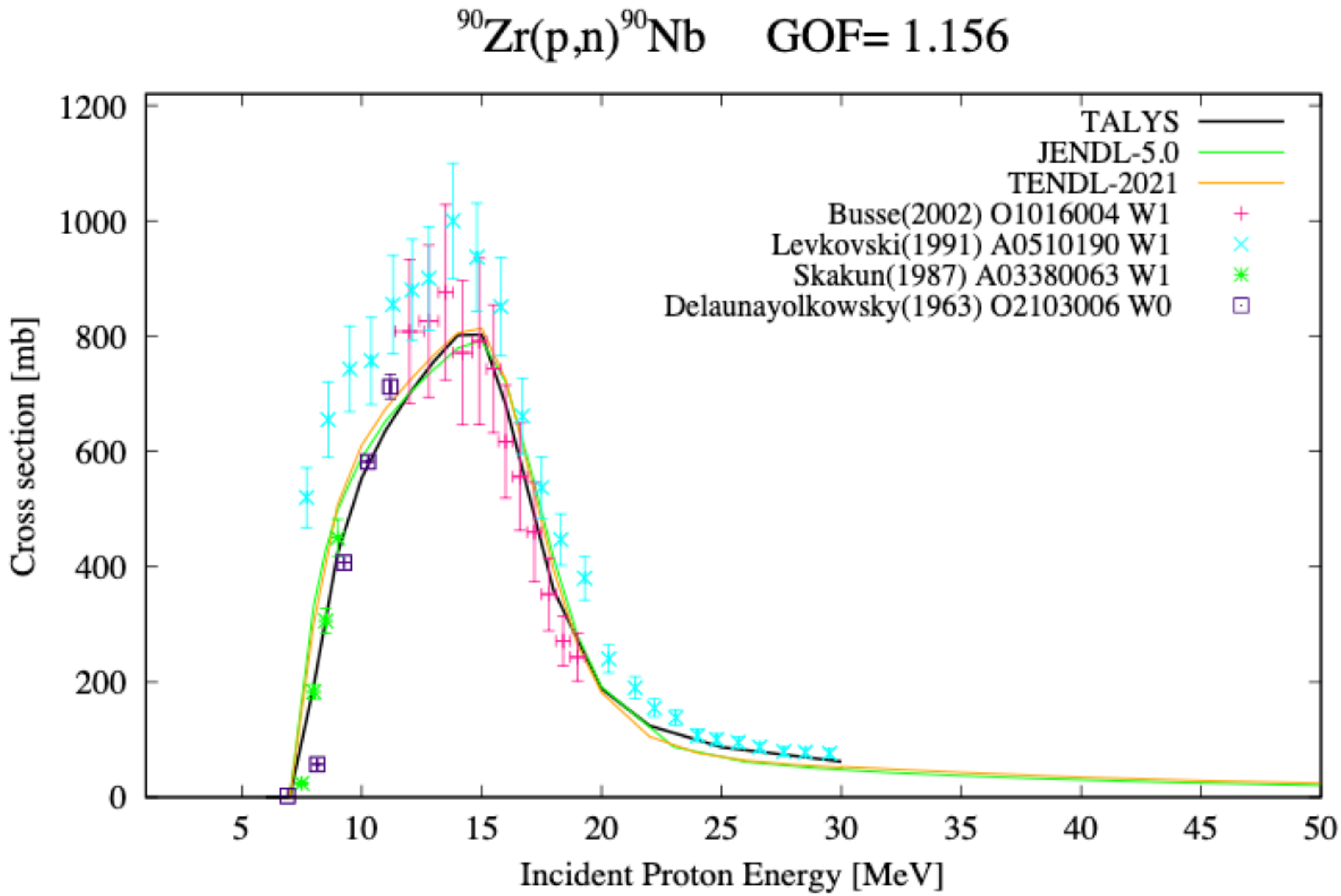
- 1. Compare every (e.g.) (p,n) for which exp. data exists (140 nuclides) with the available nuclear data libraries.
  - Always TENDL, but also JENDL-5 , ENDF/B-VIII and IAEA evaluation
  - Flag outliers, or discarded data sets, in EXFOR with a weight of 0
- 2. Write a script which uses TASMAN: statistical code for TALYS for covariance generation and automatic optimisation of nuclear model parameters to exp data
  - Loop over all 140 nuclides
  - Use the same 5 most sensitive parameters for (p,n) reactions
  - Use Nelder-Mead optimisation to all EXFOR data sets and pseudo data, this requires about 80 TALYS runs
  - Plot the results
  - If needed declare more, or less, outliers
- 3. Do this for every reaction channel

(p,n): several nuclides with JENDL-5 evaluation



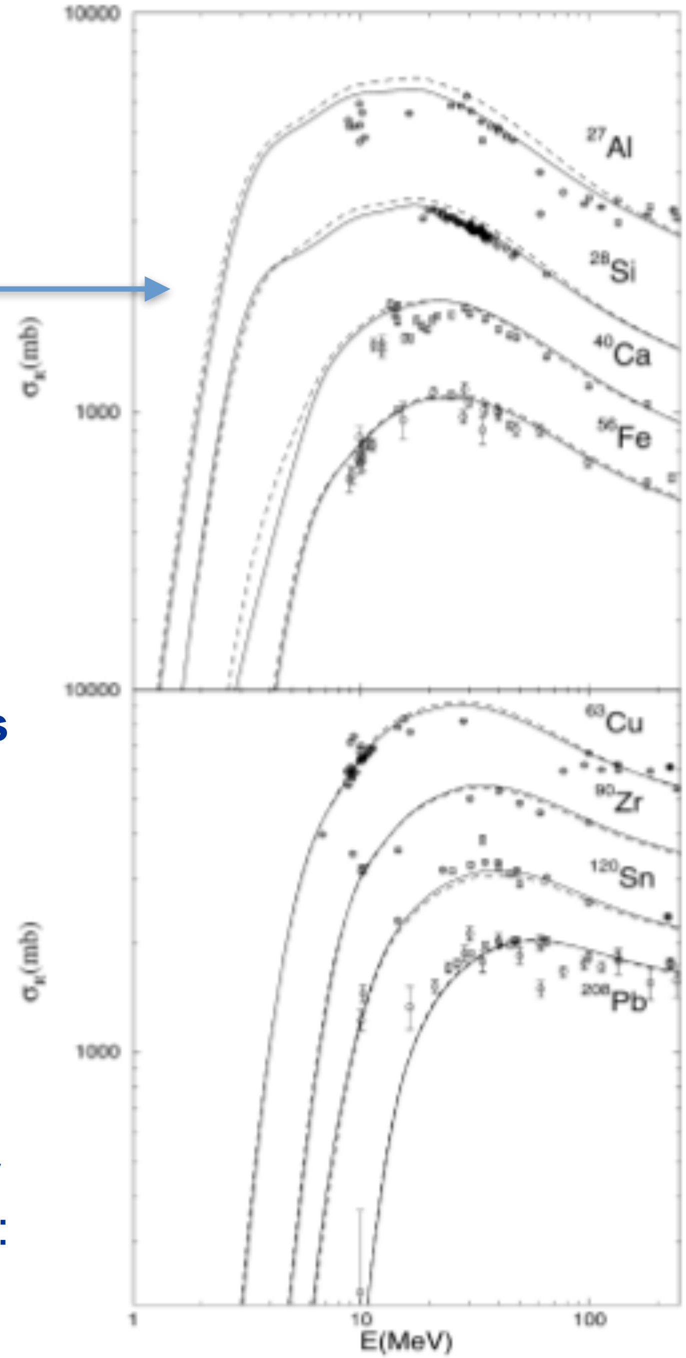
TALYS parameters for optimization

Reaction	Nuclides in EXFOR	Parameter	Parameter	Parameter	Parameter	Parameter	Parameter
(n, γ)	278	wtable					
(n,f)	34	vfiscor	betafiscor	ctable(1)	ptable(1)	ctable(2)	ptable(2)
(n,n'), (n,2n), (n,p)	210	rv(p)	$g_{ph}(0)$	$g_{ph}(n)$	ctable(n)	ctable(p)	
(N,α)	157	rv(α)	Cstrip(α)	$g_{ph}(0)$	ctable(α)		
(p,n)	142	rv(p)	rwd(p)	rv(n)	$g_{ph}(0)$	$g_{ph}(n)$	ctable(n)
(γ,n)	77	wtable	ftable	etable			
(α,n)	93	rv(α)	rwd(α)	rv(n)	$g_{ph}(0)$	ctable(α)	
(d,n)	40	rv(p)	rwd(p)	rv(n)	$g_{ph}(0)$	$g_{ph}(n)$	ctable(n)

TASMAN code (AK): Nelder-Mead optimisation.
Number of TALYS trials: N(parameters) x 20

Essential 1: Optical model potential (OMP) for proton reaction cross sections

Predictive power for KD03 global proton OMP up to 200 MeV ~ 8%



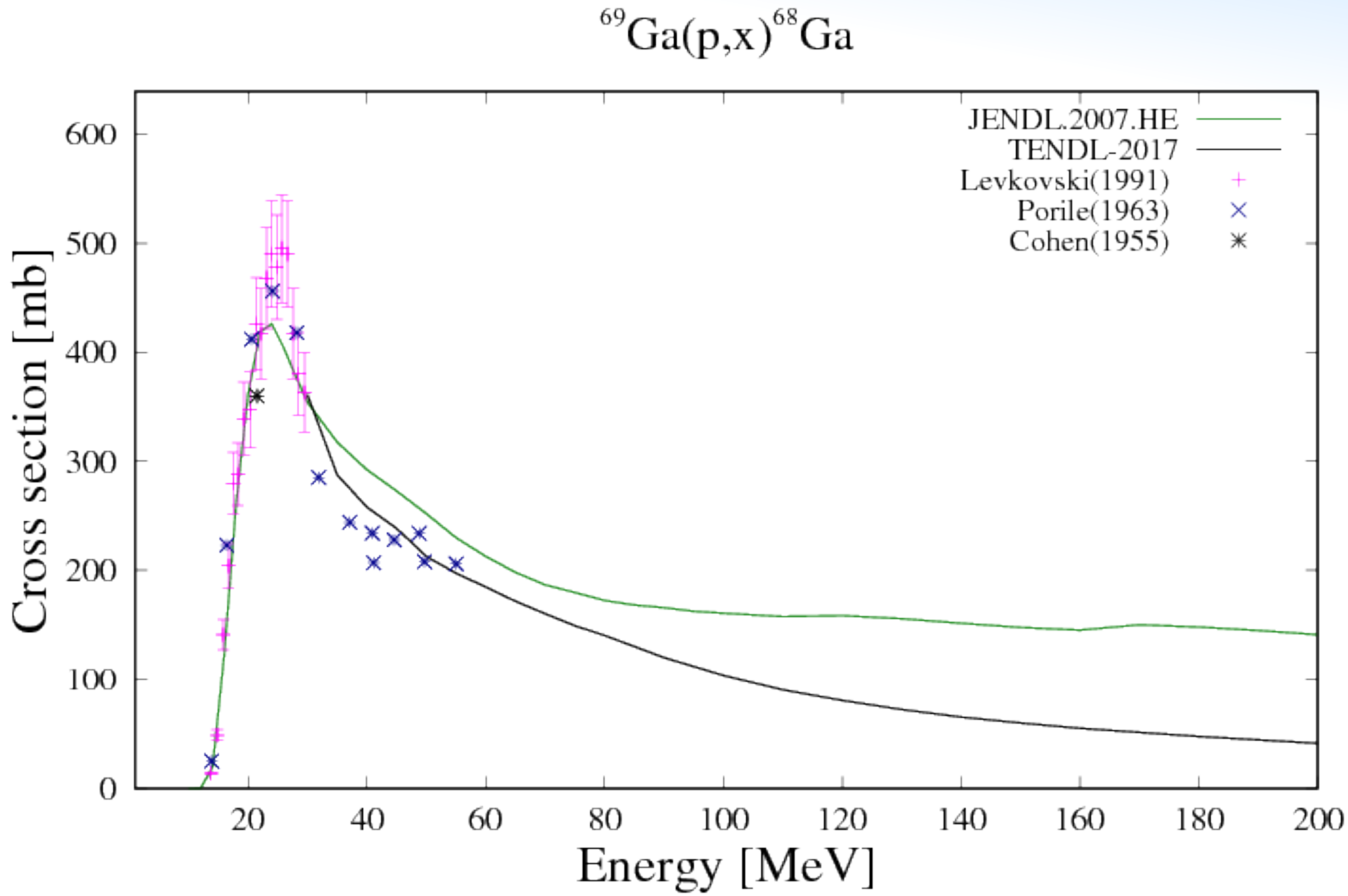
Essential 2: Nuclear level densities

Different level density models may give differences of 10-15% in peak values of excitation functions

Essential 3: Pre-equilibrium nuclear models (and multiple pre-equilibrium models above 30-40 MeV)

TALYS exciton model parameterisation has been established from single- and double-diff emission spectra, not from production cross sections
(See M.B. Fox et al, Investigating High-Energy Proton-Induced Reactions on Spherical Nuclei: Implications for the Pre-Equilibrium Exciton Model, Phys Rev C, tbp 2021)

Typical example #1: direct good fit!



Plots for all particles, all nuclides, all reactions:
nds.iaea.org/talys

Typical example #2: further TALYS adjustment needed

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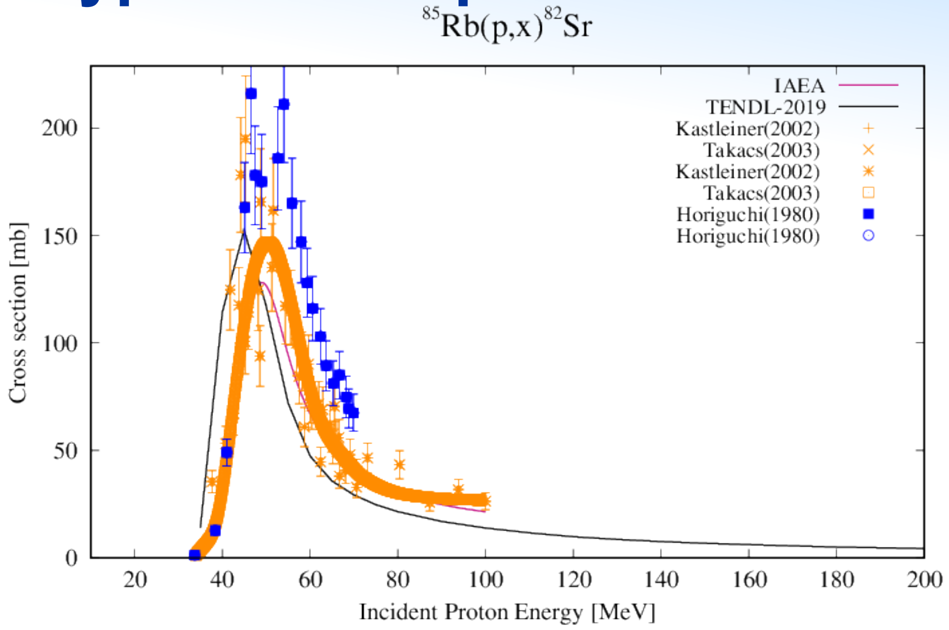
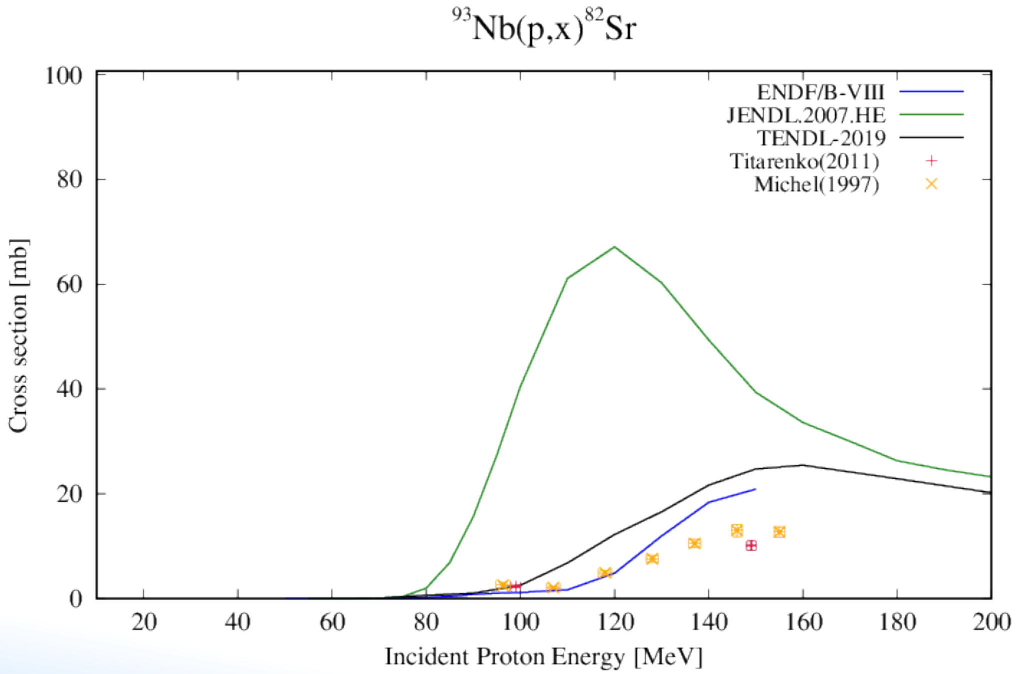
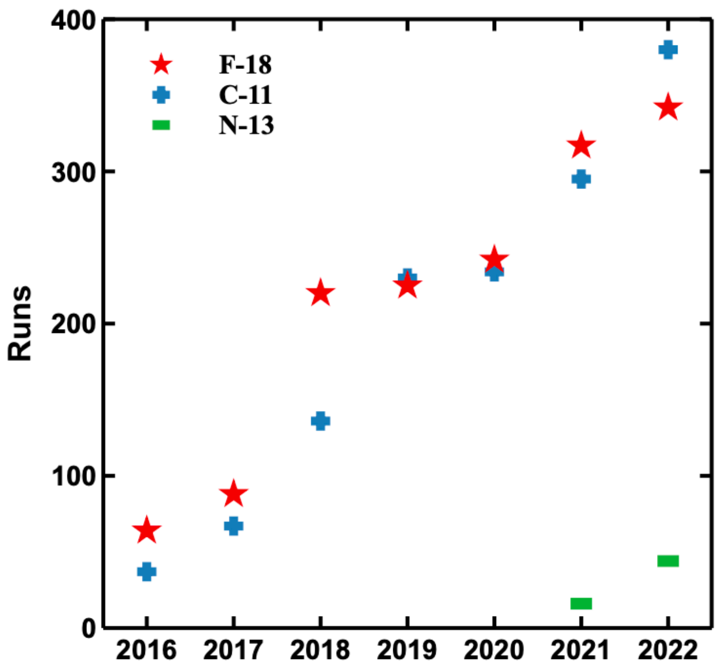


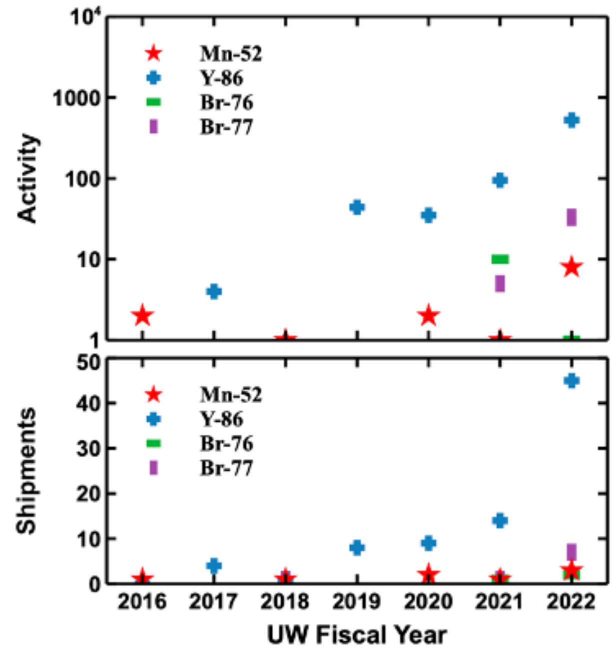


Reaction-specific adjustments
for level density and pre-equilibrium
parameters required

Radionuclide Demand



Internal runs of ^{18}F, ^{11}C, and ^{13}N by radionuclide as an aggregated annual total vs UW fiscal year. A "Run" is defined as a single production, or uninterrupted period of irradiation, whose product is delivered to a single user.



Breakdown of individual radionuclides ^{52g}Mn, ^{86}Y, and $^{76/77}Br$. The top plot shows activity produced in millicuries and the bottom shows individual shipments made against UW fiscal years from 2016 – 2022.

Nuclear Data Needs for Medical Radionuclide Producers

IAEA Technical Meeting on Nuclear Data for Medical Applications

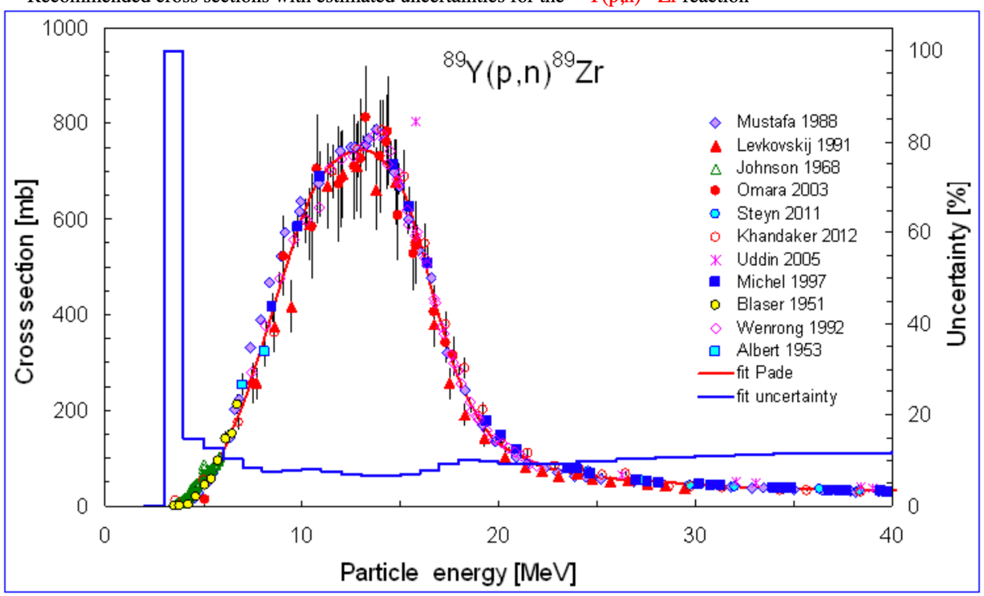
Jonathan Engle, Associate Professor

UW-Madison

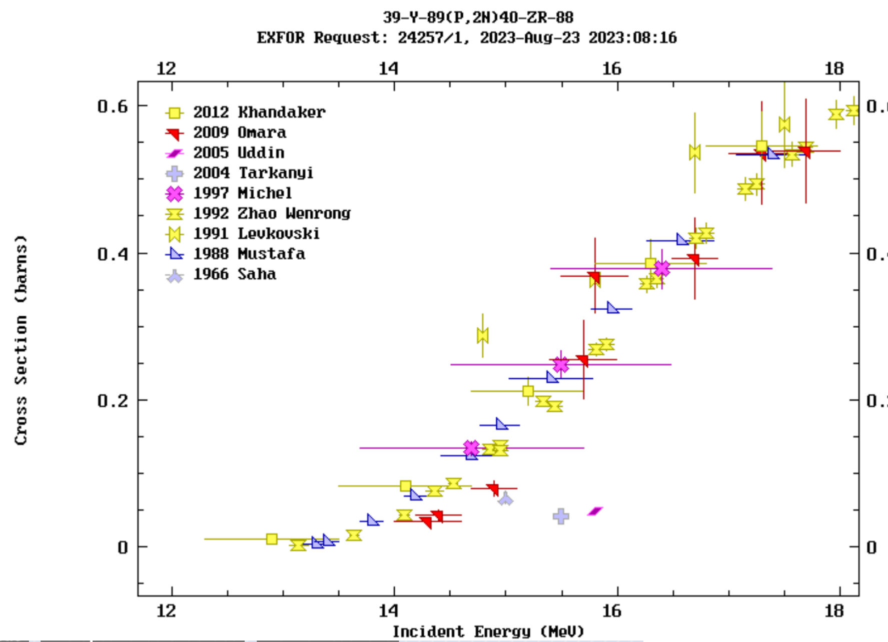
Need 1/2: Evaluations of Near-Threshold XS Data and of Produced Impurities

Many available accelerators operate at energies close to thresholds of relevant production reactions, e.g., (p,α), (p,pn), (d,n), (p,2n), (d,2n), (d,α).

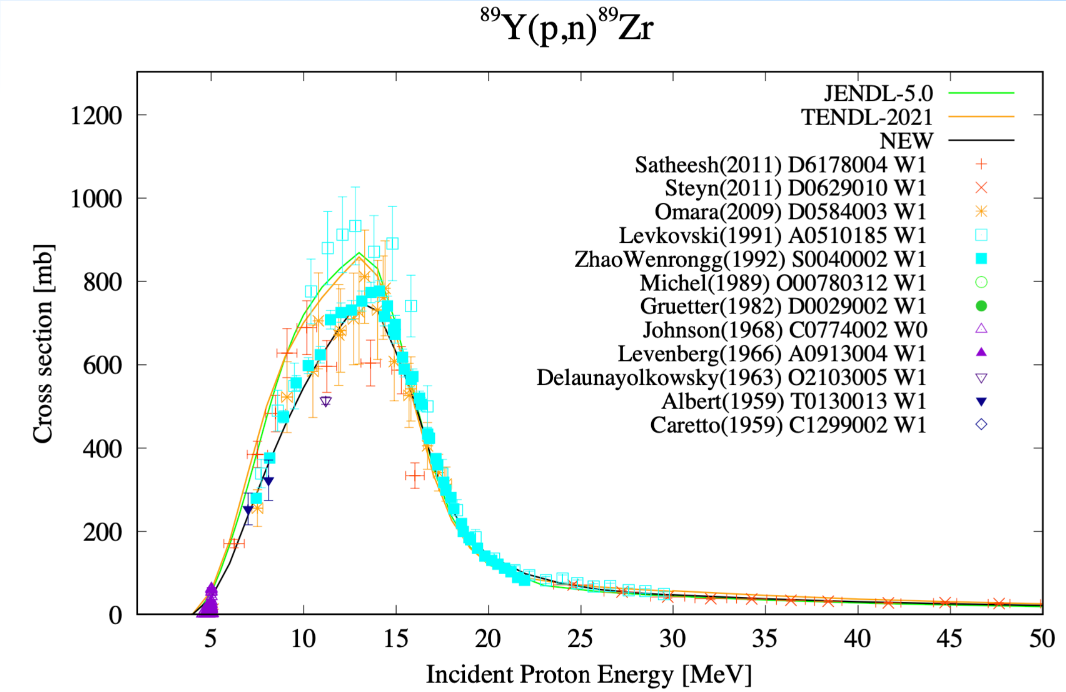
Recommended cross sections with estimated uncertainties for the $^{89}Y(p,n)^{89}Zr$ reaction



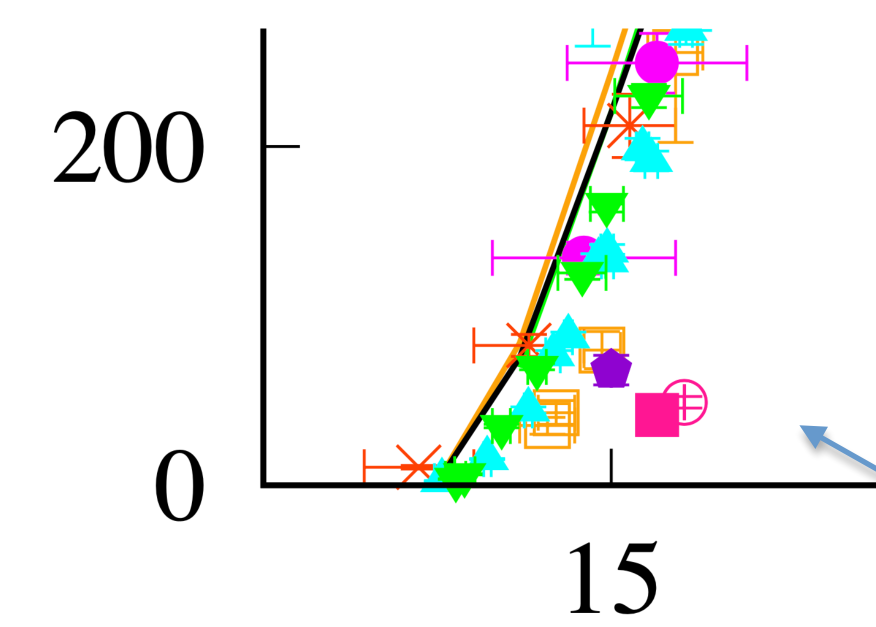
Updated: Aug. 2018.

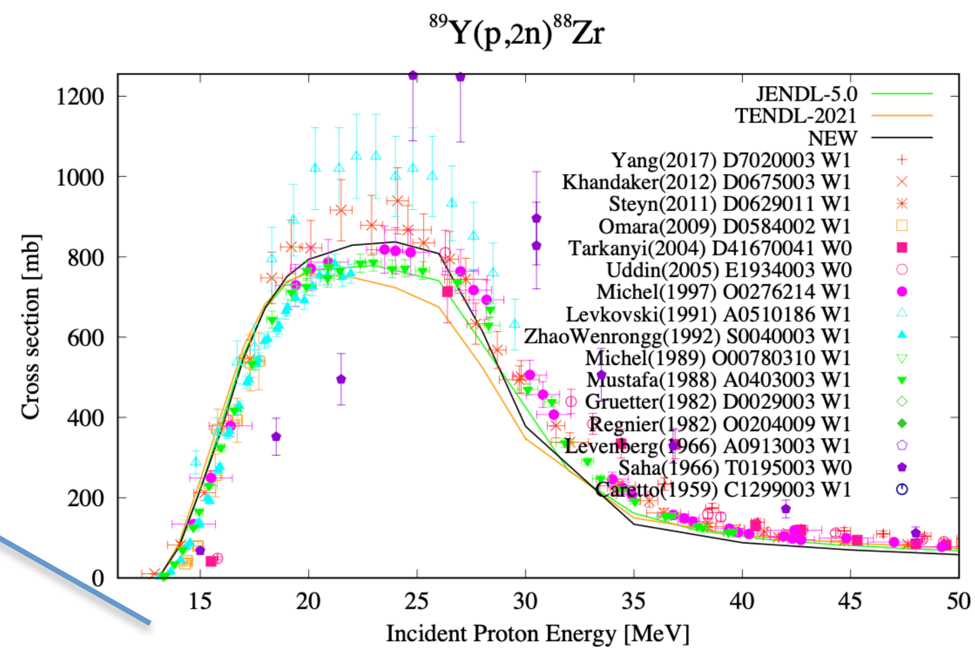


How large in the Zr-88 impurity?




```
# Adjusted parameters from fitting
rvadjust     p    0.72464
rwdadjust    p    0.99070
rvadjust     n    1.00842
gadjust       40   90     1.11250
gadjust       40   89     1.10842
ctableadjust  40   89     0.65147 0
```





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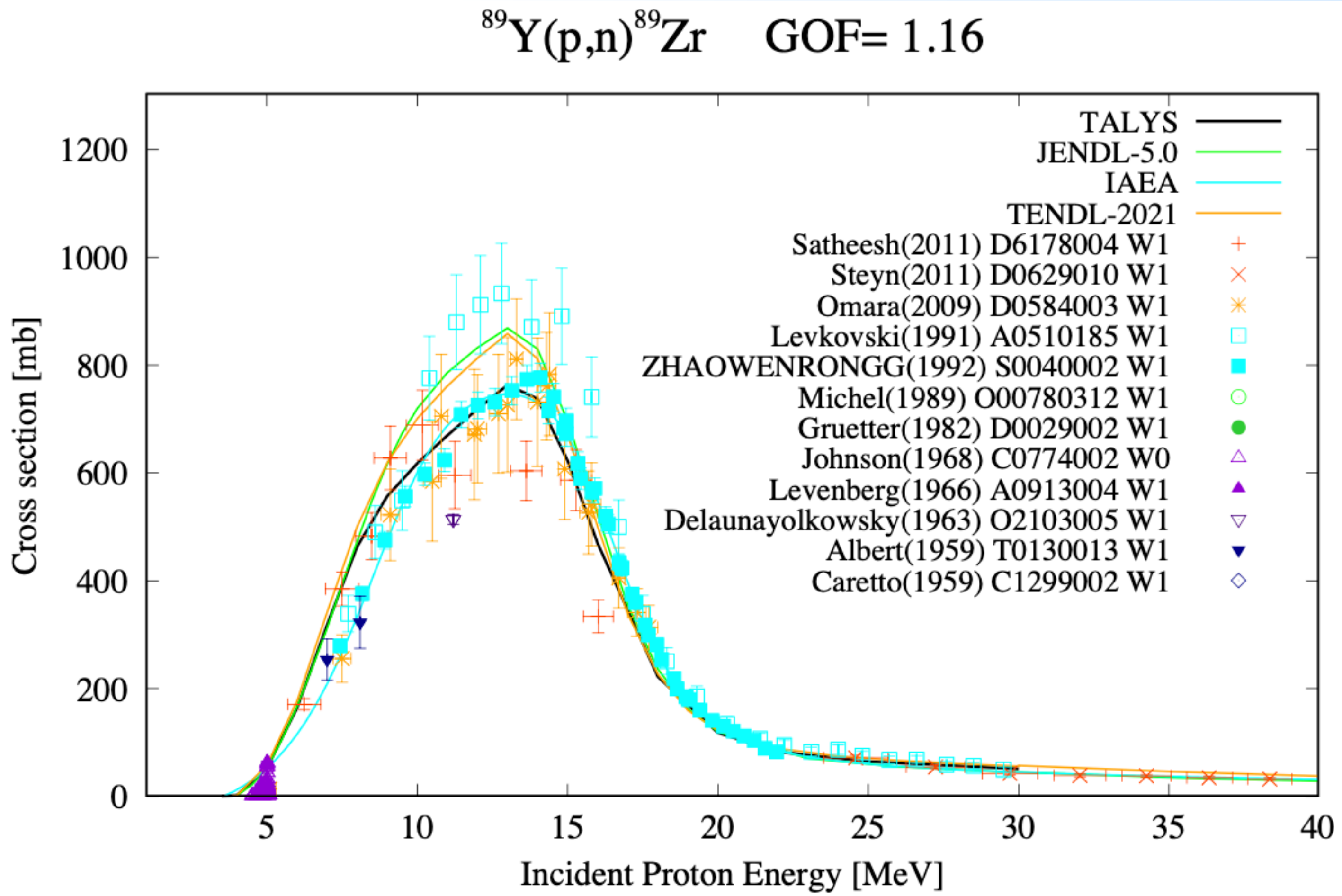
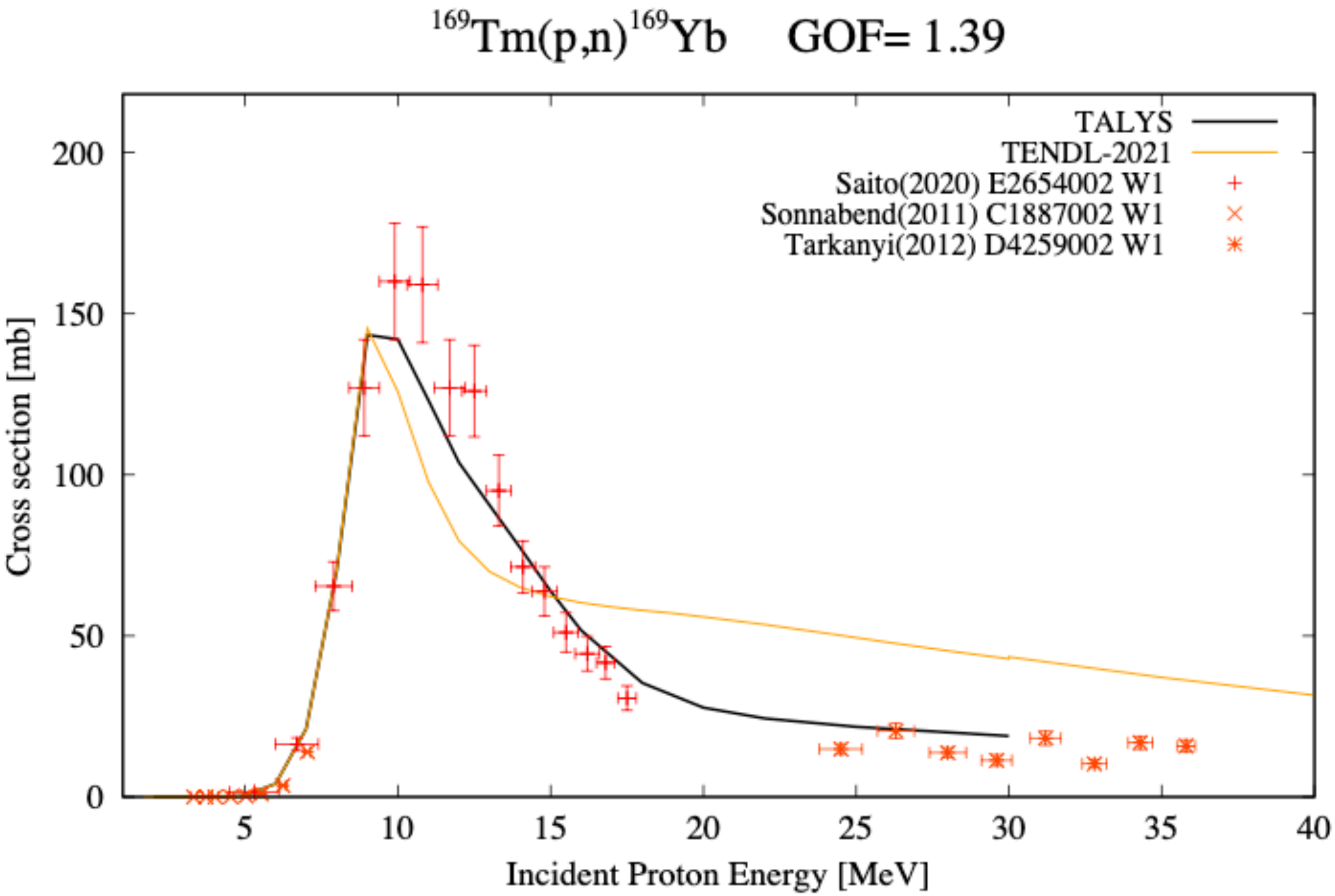
Automated fit (excluding outliers)




```
rvadjust       p    0.92464
rwdadjust      p    0.99070
rvadjust       n    1.00842
gadjust       40   90     1.11250
gadjust       40   89     1.10842
ctableadjust  40   89     0.65147 0
```

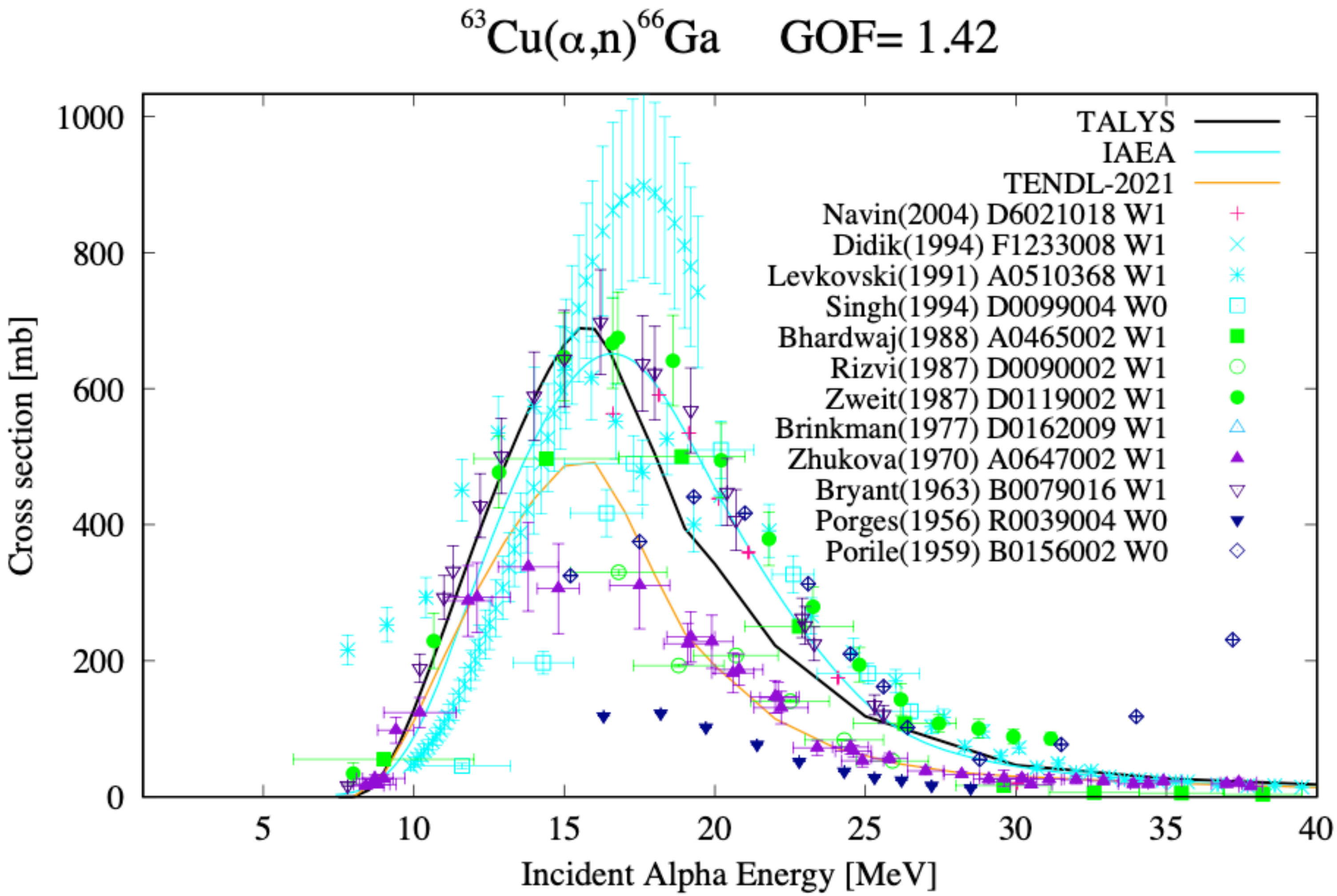
142 nuclides

Sometimes significant differences




```
rvadjust      p     0.97574
rwdadjust     p     0.87606
rvadjust      n     1.02987
gadjust        70 170     1.18983
gadjust        70 169     0.92669
ctableadjust   70 169     0.31802 0
```

TENDL-2023 will have optimised fits



93 nuclides

```
rvadjust      a    1.00307
rwdadjust     a    0.93064
rvadjust      n    1.01999
gadjust        31  67      0.98591
ctableadjust   31  66      0.88253 0
```

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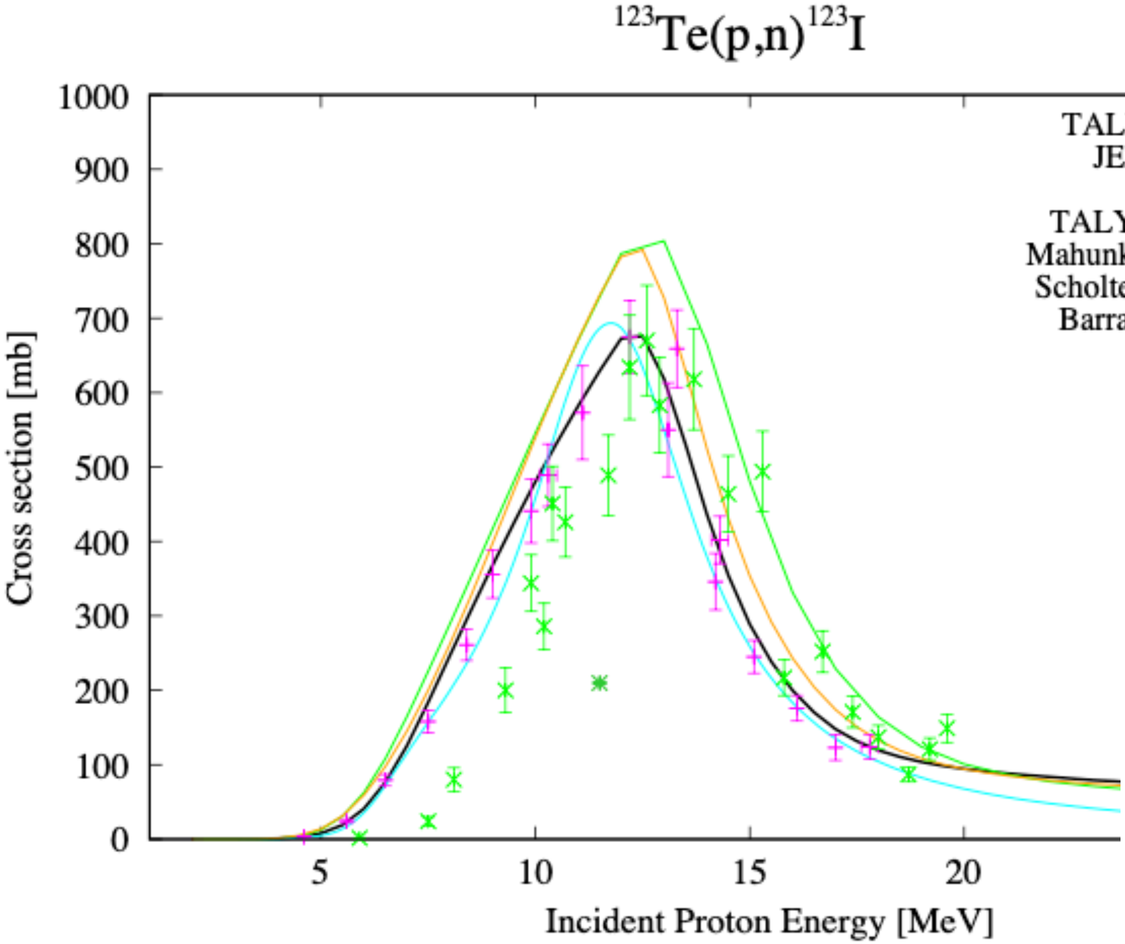
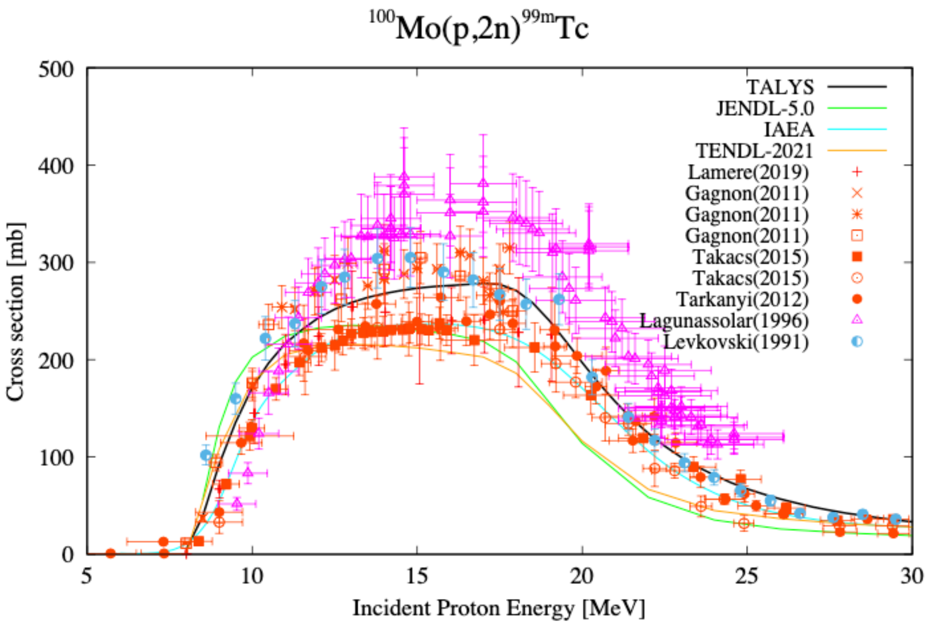
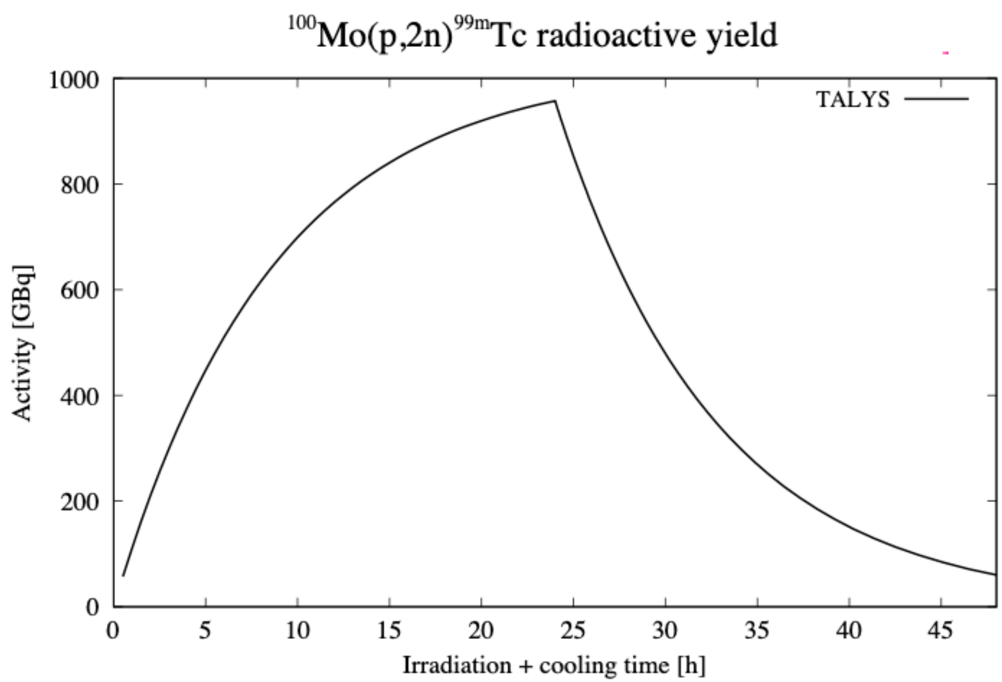


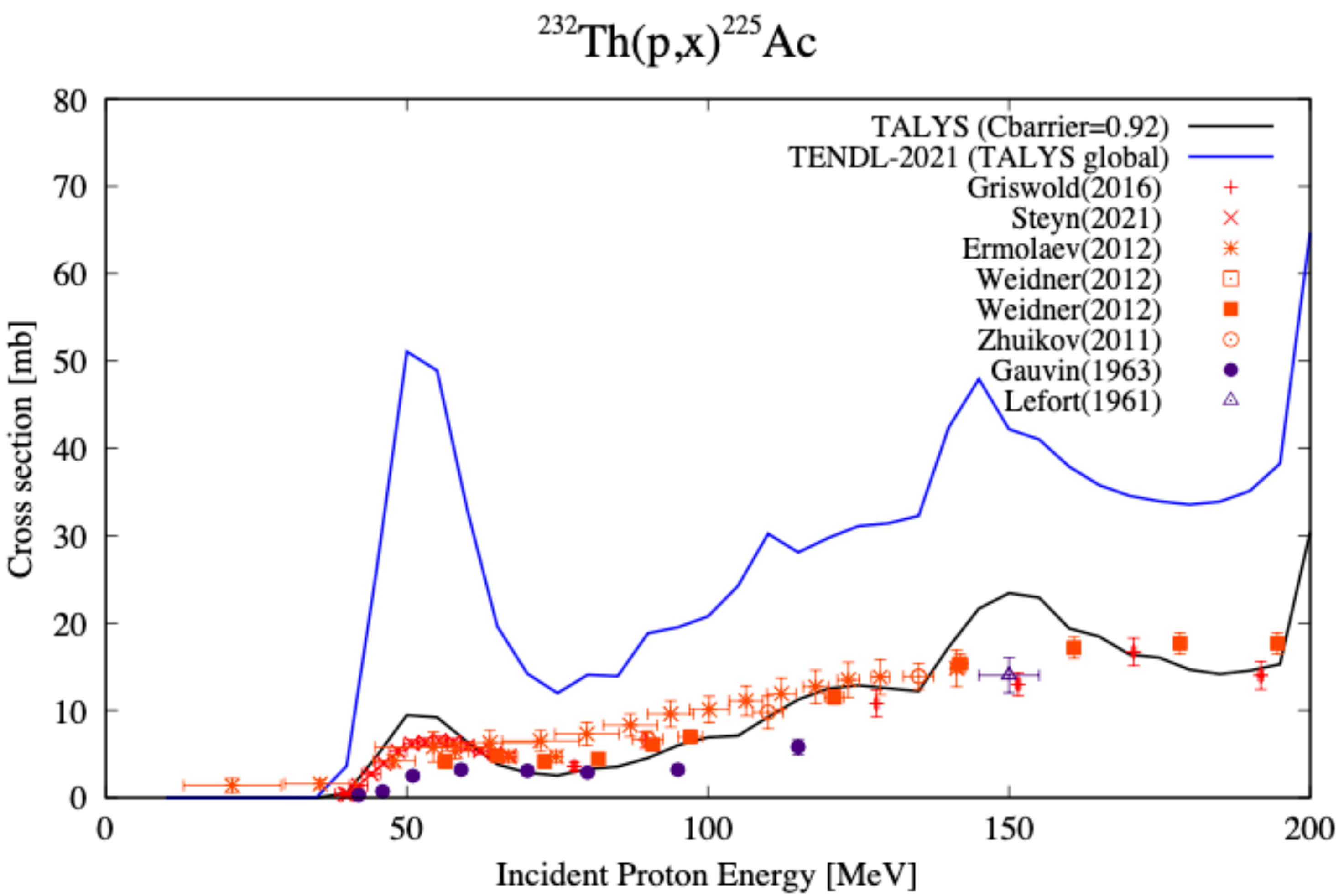

```
#
# p-Te123-pn-fit
#
# General
#
projectile p
element Te
mass 123
energy 1. 30. 0.5
#
# Adjusted parameters
#
rvadjust      p    0.95947
rwdadjust     p    0.93338
rvadjust      n    1.02377
gadjust        53 124    0.99089
gadjust        53 123    1.09408
ctableadjust   53 123 -2.54809E-01 0
```






```
#
# p-Mo100-medical
#
# General
#
projectile p
element    Mo
mass       100
energy     8. 30. 0.5
#
# Spherical OMP and adjusted parameters
#
spherical y
rvadjust      p    1.00676
rwdadjust     p    1.11091
rvadjust      n    1.04395
gadjust        43 101    1.22030
gadjust        43 100    1.04828
ctableadjust   43  99    1.34123 0
s2adjust       43  99    0.14784 0
#
# Medical isotope production
#
production y
Ibeam 0.15
Ebeam 24.
Eback 10.
```

Actinides: uncertainty due to fission

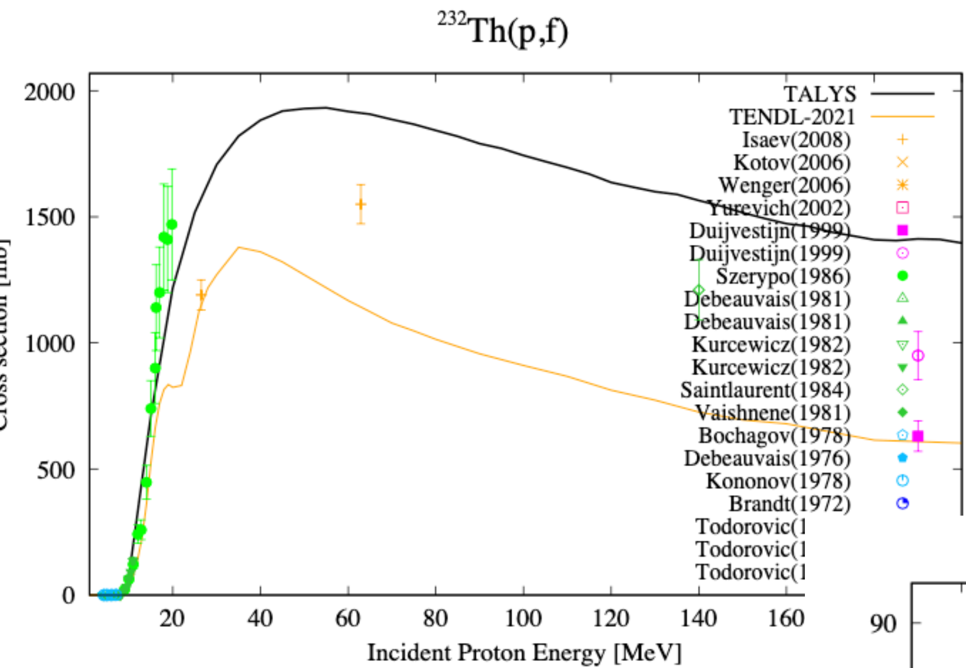


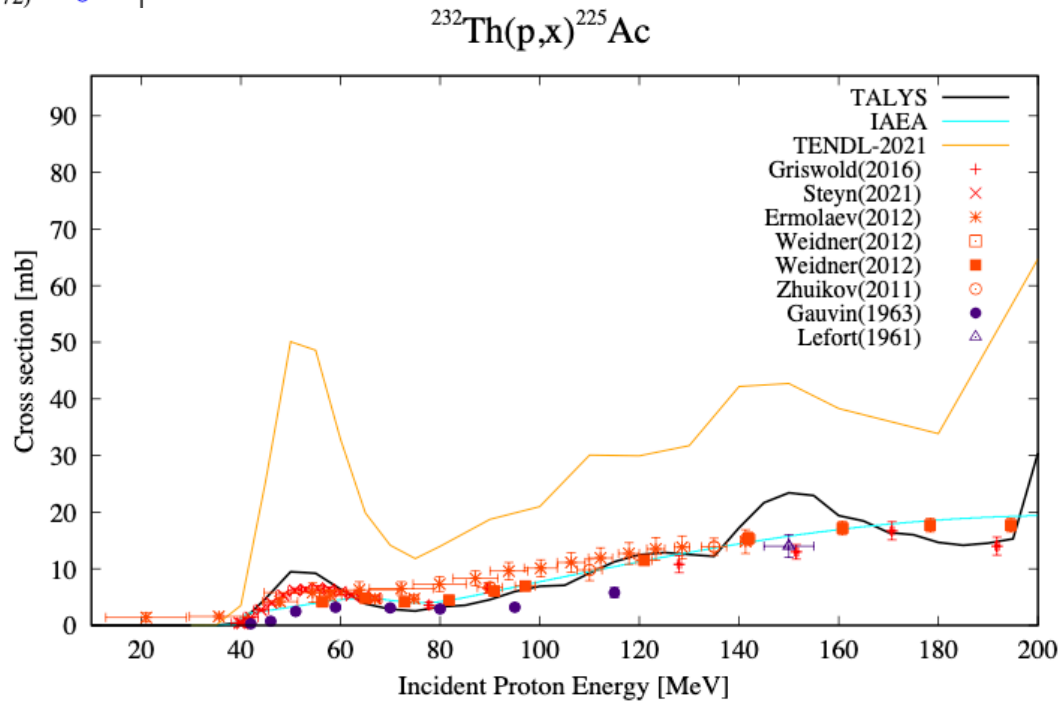
TALYS input file:

```
projectile p
element Th
mass 232
energy 10. 200. 5.
partable y
bins 100
Cbarrier 0.92
```

TALYS-2: one parameter ‘Cbarrier’ to reduce/increase all fission barriers

Th232(p,f) could be higher compared to TENDL-2021





Consistency with neighbouring channels

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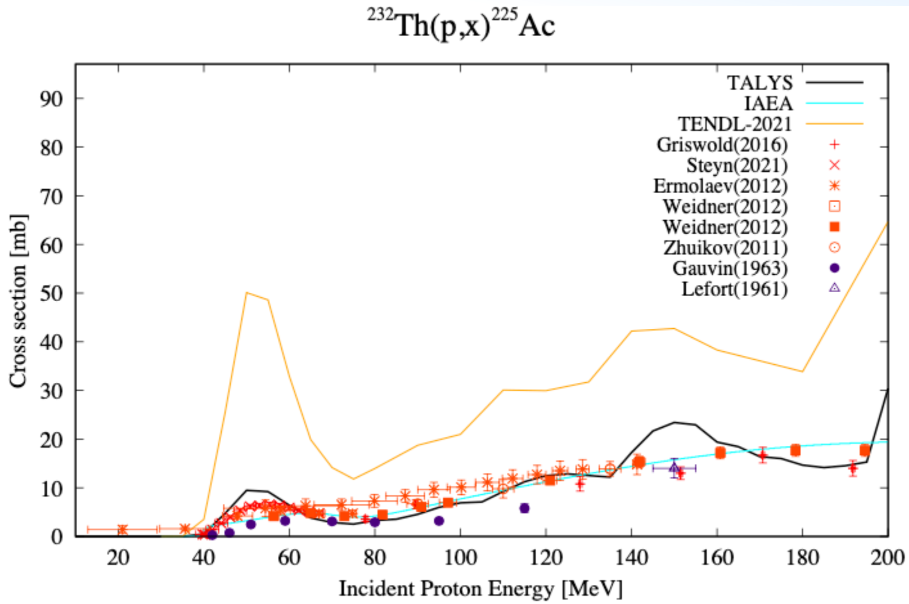
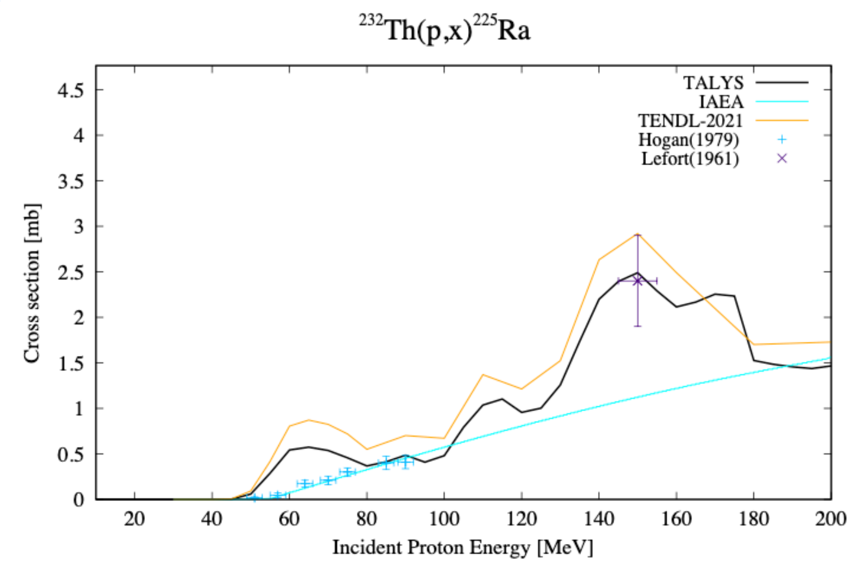
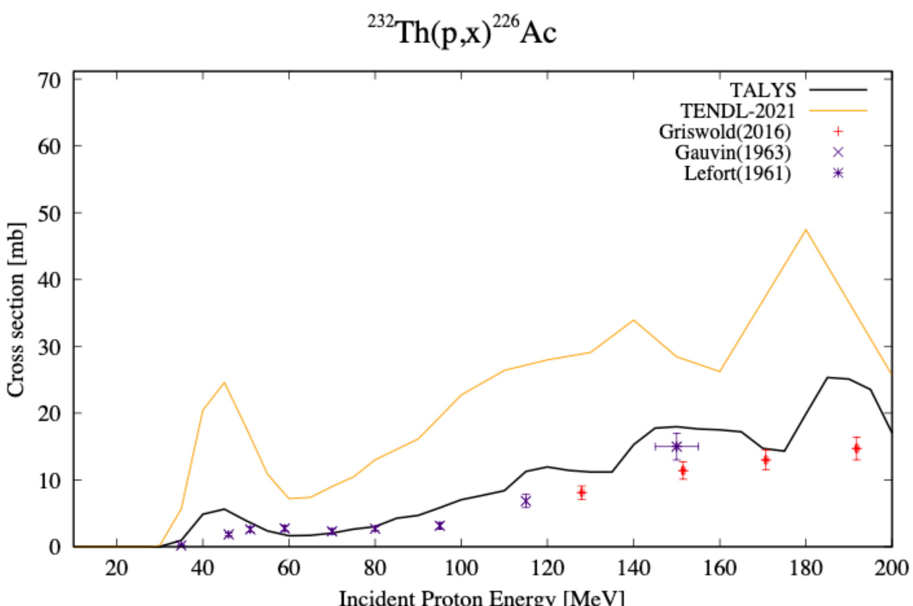
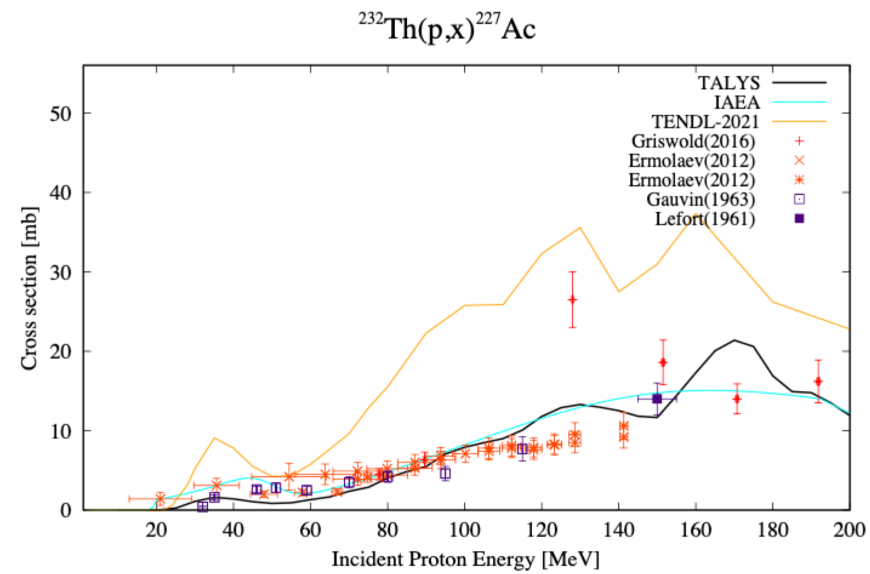
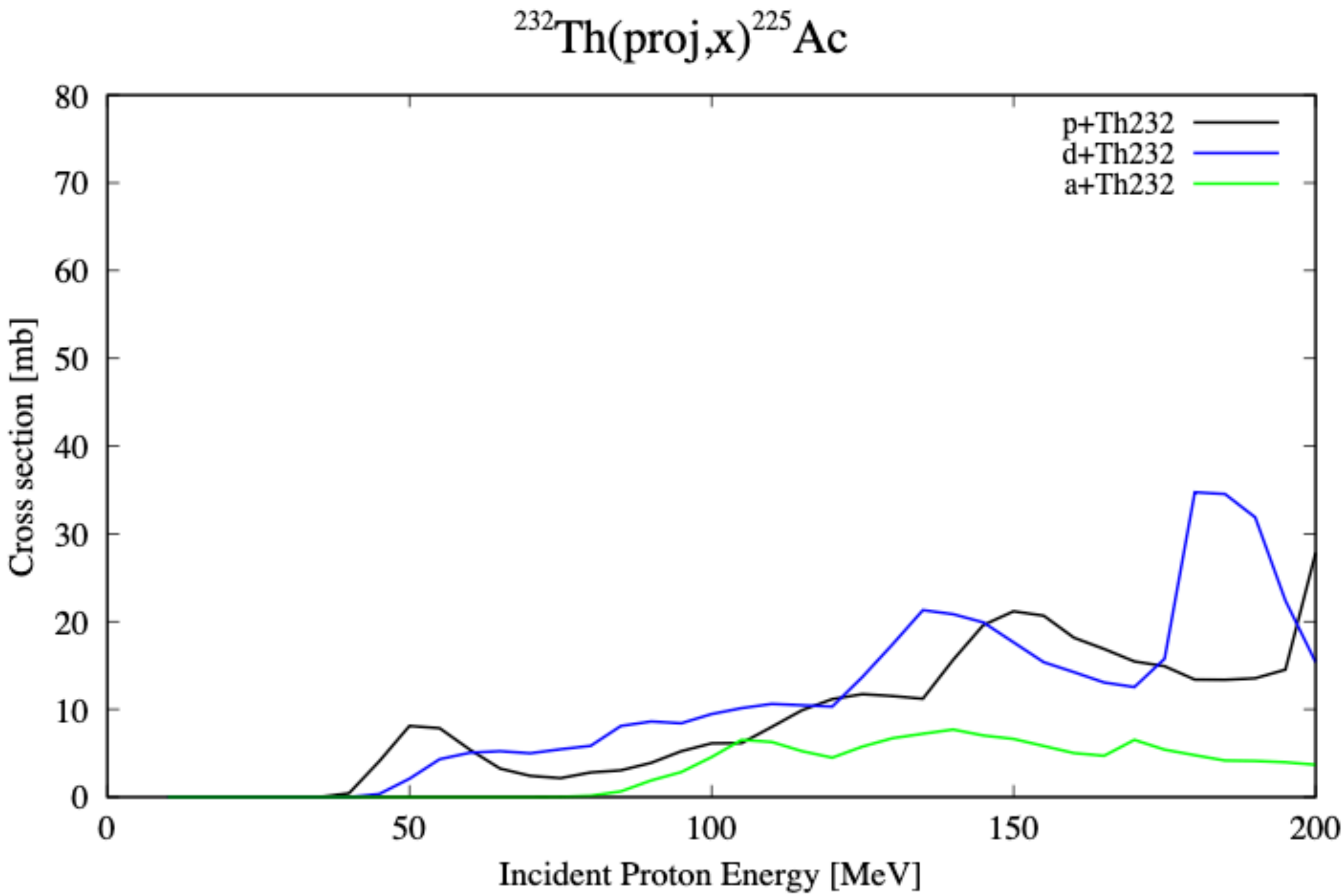








Other projectiles?



Conclusions

- Automated optimisation to many reaction channels with a relatively small number of TALYS parameters
  - Requires computational access to entire EXFOR database at once
  - Requires extensive outlier database
- TENDL-2023 will contain optimised excitation functions

Thank you!