Multidisciplinary applications of neutron activation analysis (NAA) at NPI

J. Kučera, J. Kameník, J. Mizera, Z. Řanda

Nuclear Physics Institute, CAS, Husinec-Řež, Czech Republic

EUROPEAN UNION
European Structural and Investment Funds
Operational Programme Research,
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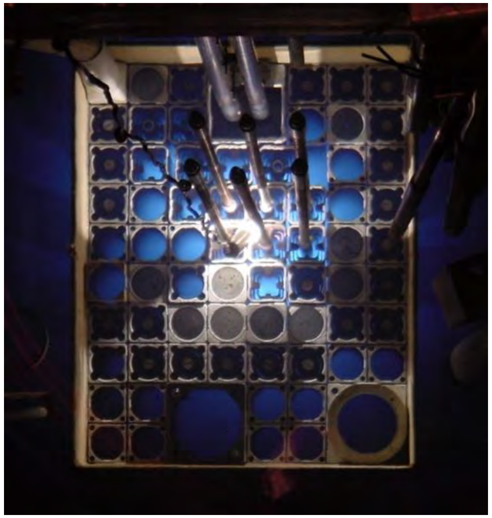
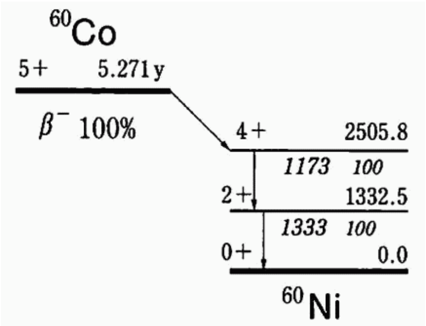
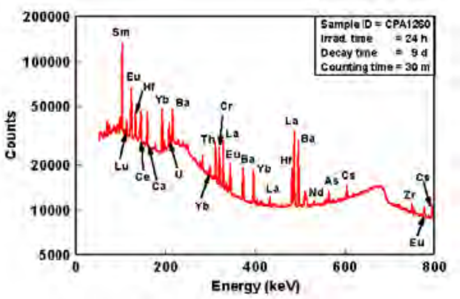
Facilities for NAA at NPI



- Irradition in the LVR-15 reactor at a thermal neutron fluence rate up to 5x10^{13} cm^{-2} s^{-1}
- Facilities for both short-time (10 s – 180 s) and long-time (several hours – several days) irradiation
- 4 coaxial high efficiency (20-78 %), high resolution (FWHM 1.75-1.85 keV @ 1332.5 keV) HPGe detectors
- 2 planar HPGe detectors (thickness 15 mm, area 500 mm^2, FWHM 550 eV @ 122 keV)
- 1 well-type HPGe detector (eff. volume 150 cm^3, FWHM 2.02 keV @ 1332.5 keV)
- Radiochemical laboratories

Advantages and major applications of NAA





- highly penetrating nature of neutrons as activation particles and emitted γ-rays of activation products leading to **virtual matrix independence**
- **non-destructive and multi-elemental character** of analysis as a result of the above properties
- **very low detection limits** and high specificity for a large number (30-40) of elements
- independence of chemical state and virtual **absence of analytical blank**
- **high potential for accuracy and low uncertainty of results**
- **Applications:** Environmental control and monitoring, biomedicine, **geo- and cosmochemistry, material science, cultural heritage**, mycogeochemistry, chemometry (control analyses, certification of reference materials)

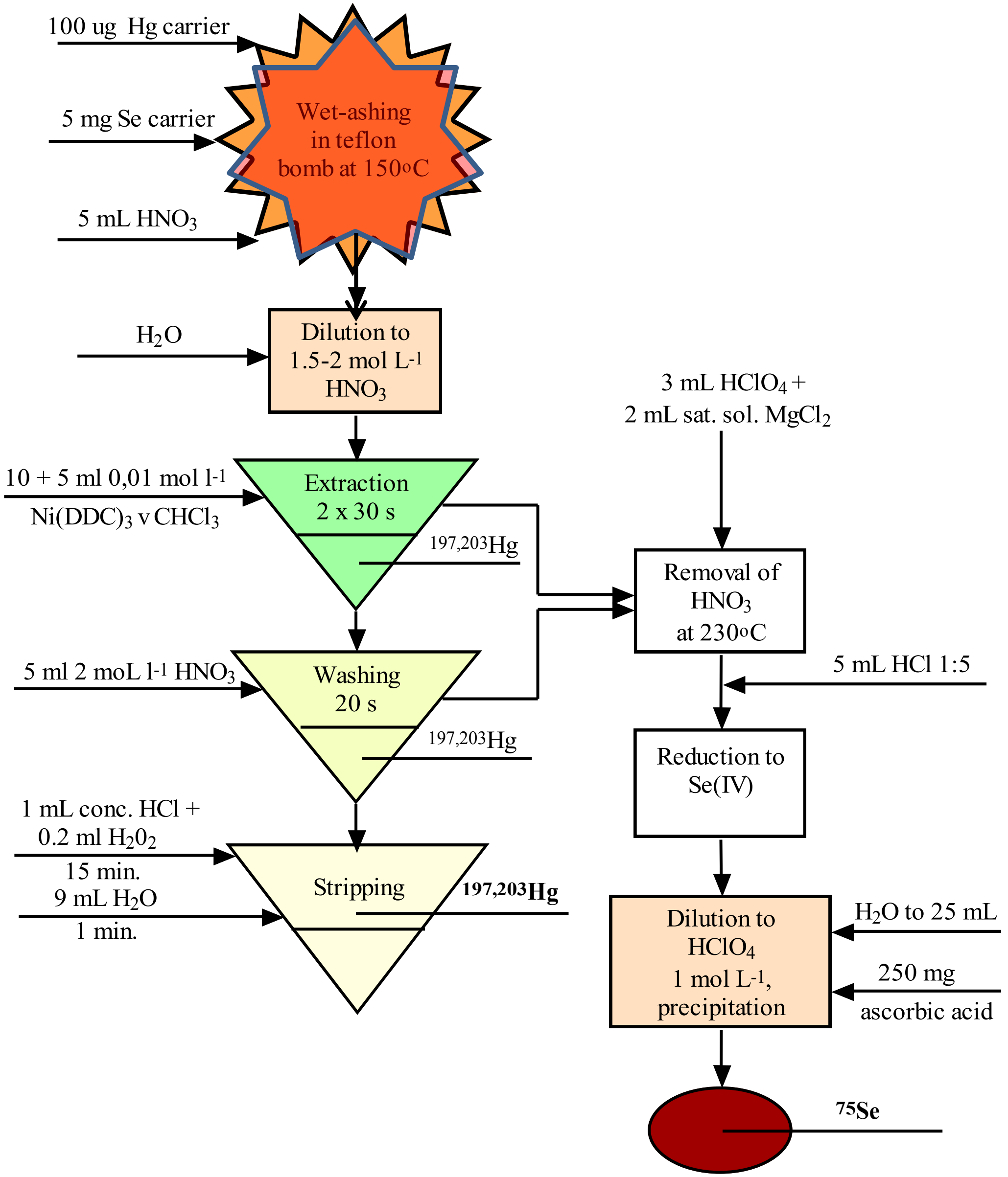
Culture heritage studies

Cultural heritage studies (historical forensics)

Determination of Hg in Tycho Brahe’s remains (hair, bones) by RNAA

Was Tycho Brahe poisoned by Hg?

Hg determination by RNAA



100 ug Hg carrier

5 mg Se carrier

5 mL HNO_3

Wet-ashing in teflon bomb at 150°C

H_2O

Dilution to 1.5-2 mol L^{-1} HNO_3

10 + 5 ml 0,01 mol l^{-1} $Ni(DDC)_3$ v $CHCl_3$

Extraction 2 x 30 s

$^{197,203}Hg$

5 ml 2 moL l^{-1} HNO_3

Washing 20 s

$^{197,203}Hg$

1 mL conc. HCl + 0.2 ml H_2O_2

15 min.

9 mL H_2O

1 min.

Stripping

$^{197,203}Hg$

3 mL $HClO_4$ + 2 mL sat. sol. $MgCl_2$

Removal of HNO_3 at 230°C

5 mL HCl 1:5

Reduction to Se(IV)

Dilution to $HClO_4$ 1 mol L^{-1}, precipitation

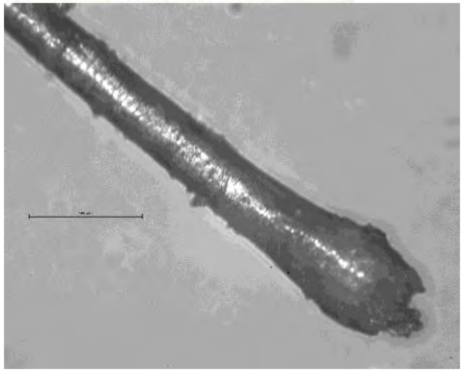
H_2O to 25 mL

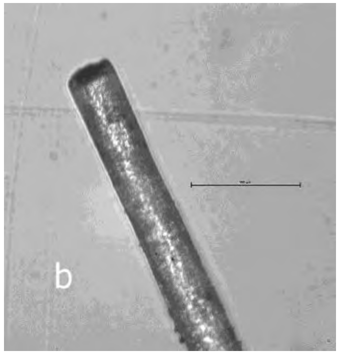
250 mg ascorbic acid

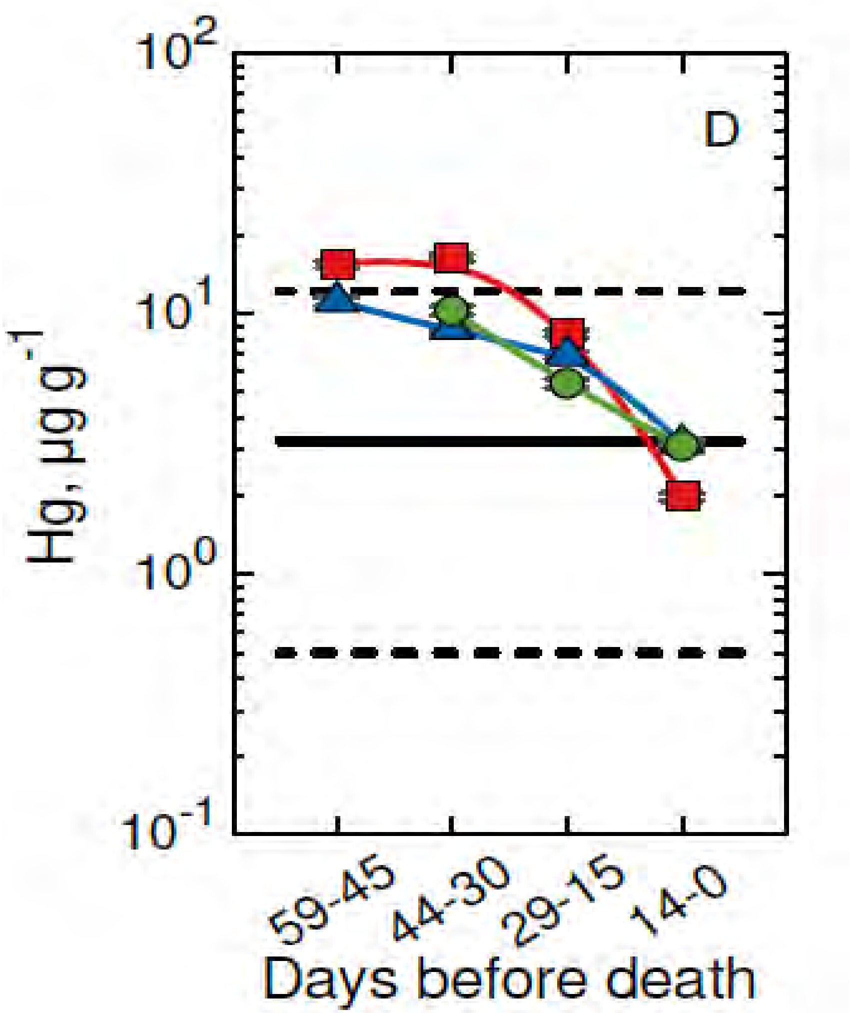
^{75}Se

Mercury (^{203}Hg, $t_{1/2}$=46.6 d)

RNAA of remains of Tycho Brahe (hair, beard hair, bones)





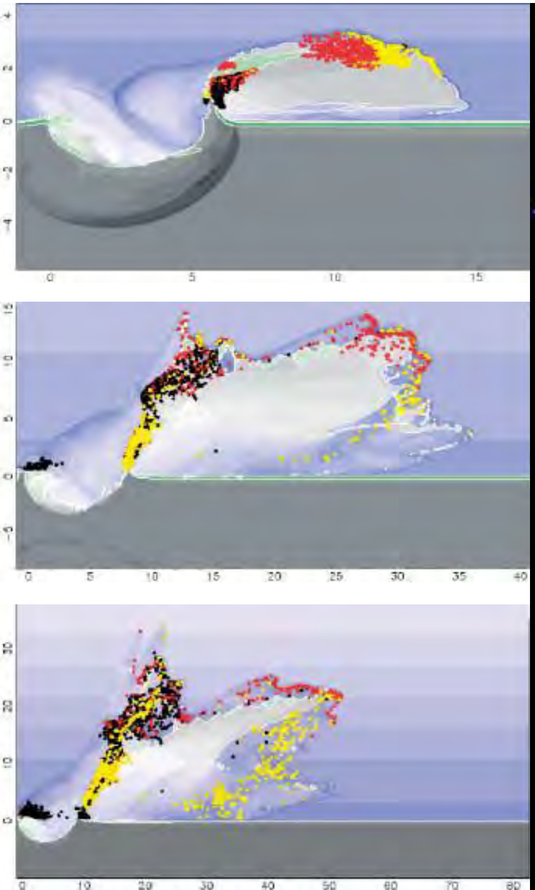


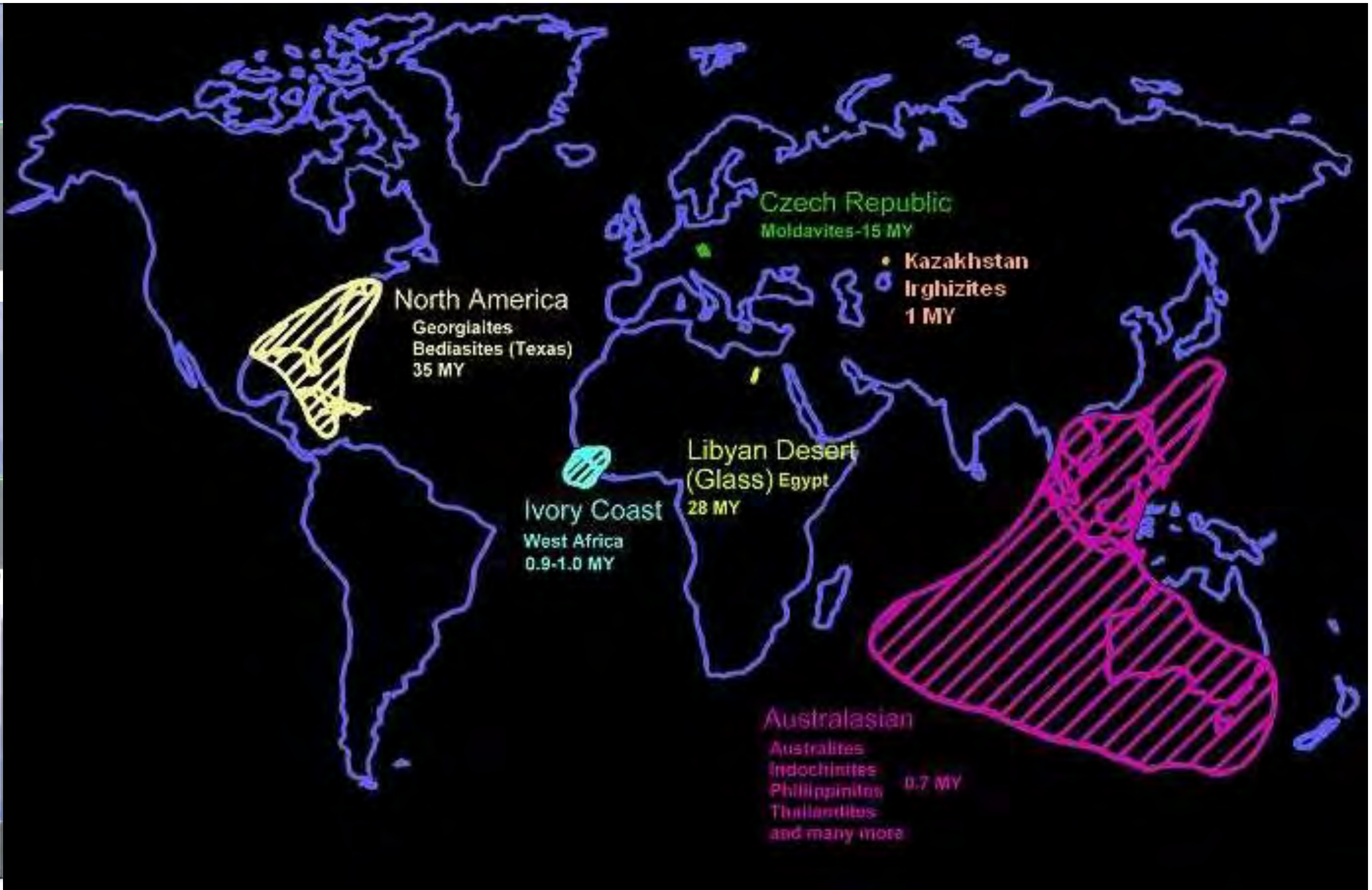
K.L. Rasmussen, J. Kučera, L. Skytte, J. Kameník et al., Archaeometry *55 (2013) 1187–1195*

Geo- and cosmochemical studies

INAA/IPAA of tektites & impact glasses

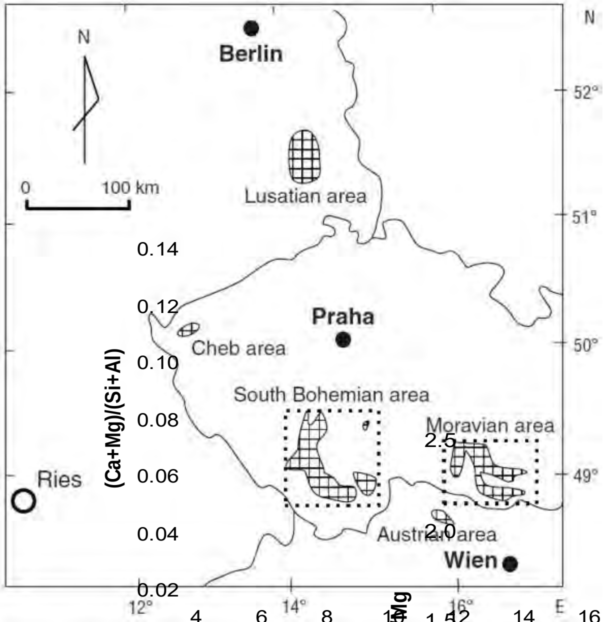
- Natural glasses produced during large meteoritic impacts
- ***Tektites*** – ejected from parent crater to distant strewn fields
- Moldavites, irghizites, Australasian tektites and Libyan Desert Glass systematically studied at NPI by INAA/IPAA since 2007



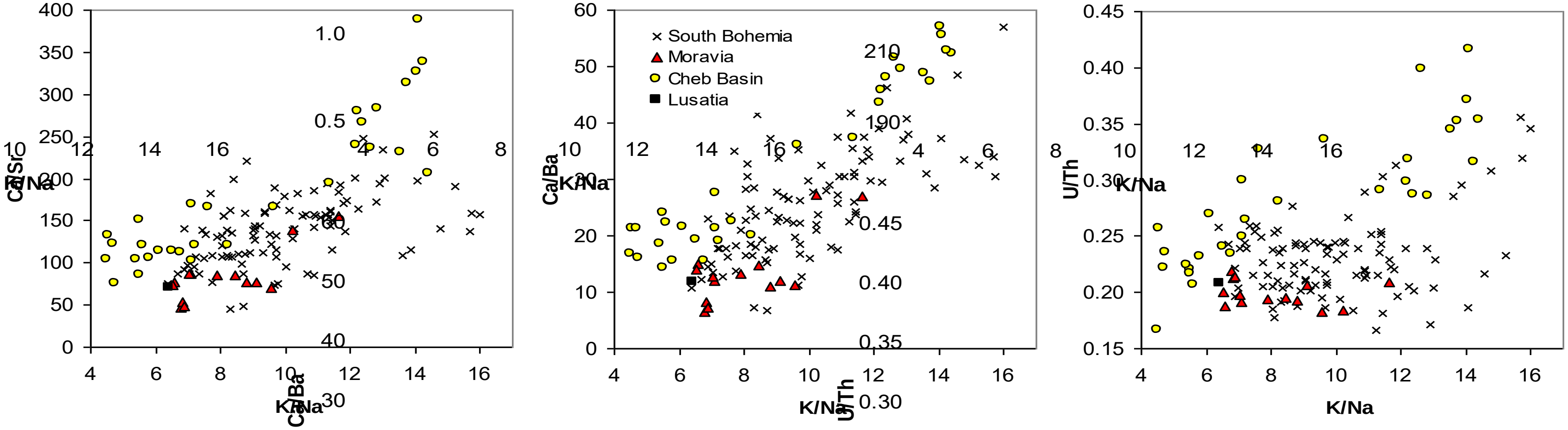


Moldavites: unconventional source components

- Parent impact structure – crater Ries (14.7 Ma, Ø 25 km)
- Precursors: Miocene sedimentary rocks / sediments composed of quartz sands, clay and soil, and **Ca-Mg component** - carbonates?
- New substrewn field – the Cheb Basin (CBM)
- New theory – **Ca-Mg component assigned to ash from biomass** burned at the early stage of the meteoritic impact
- Evidenced by, e.g., correlation between K/Na, Ca/Sr, Ca/Ba (essential/nonessential element differentiation in plants) and U/Th ratios, recently by C isotopic analysis

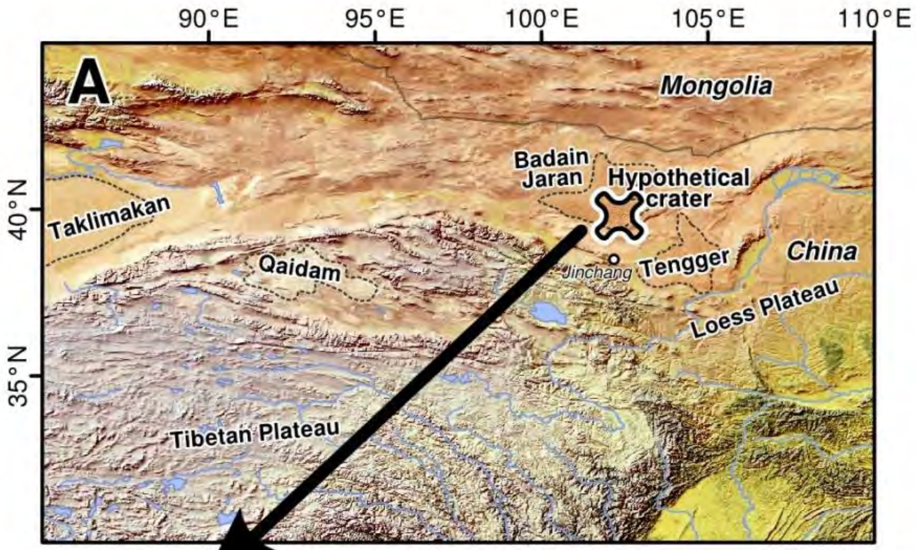


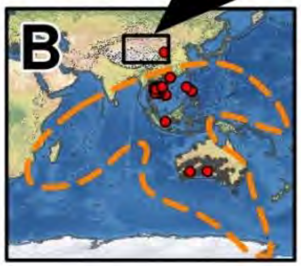
- Řanda Z et al. (2008) Meteor Planet Sci 43: 461–477
- Žák K et al. (2012) Meteor Planet Sci 47: 1010-1028
- Žák K et al. (2016) Geochim. Cosmochim. Acta 179, 287-311

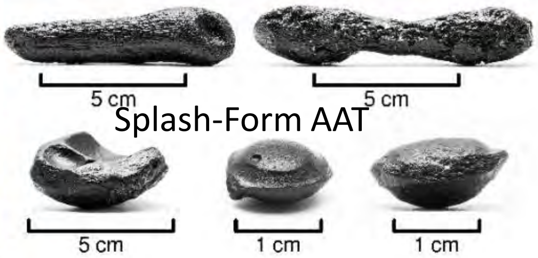


Source materials and parent crater for Australasian tektites (AAT)

- Largest tektite strewn field with **unknown location of parent crater**
- Set of AAT from various parts of the strew field analyzed at NPI ⇨ origin of AAT from Chinese loess and its precursors (desert sand) should be revisited based on geochemical and isotopic constraints
- **Criticism of** currently proposed **crater location in Indochina** with ambiguous definition of target materials ⇨ with the aid of global gravity data, **hypothetical crater location in the Badain Jaran Desert proposed**

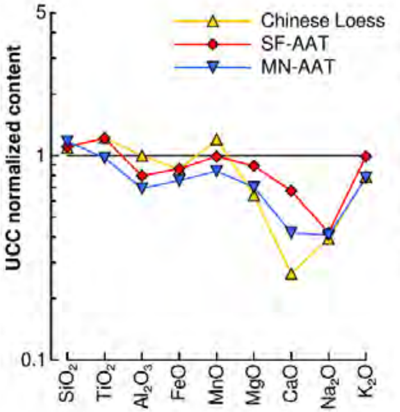


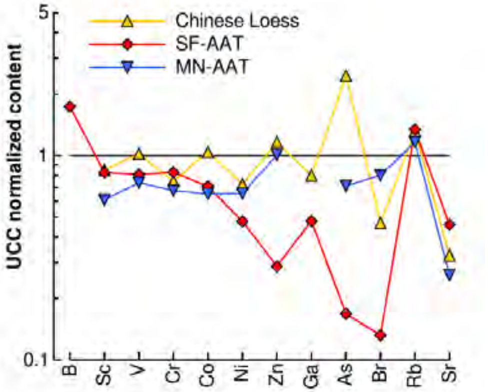


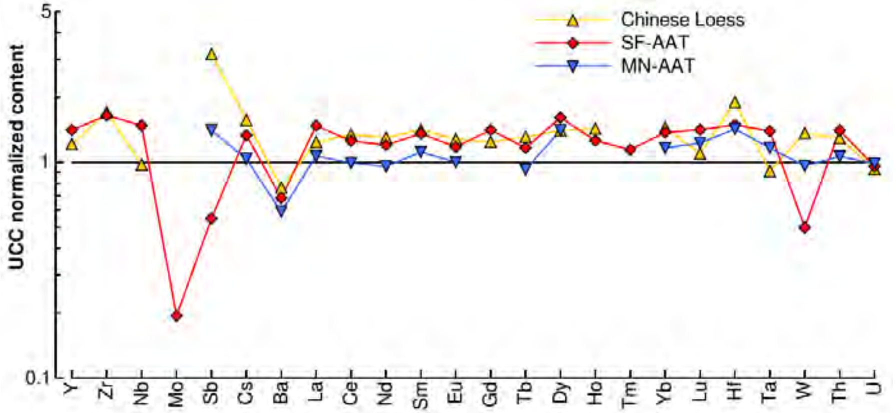


Splash-Form AAT

Muong Nong AAT







Multielemental analysis of meteorites by INAA, RNAA and IPAA

Morávka meteorite fall on on May 6, 2000

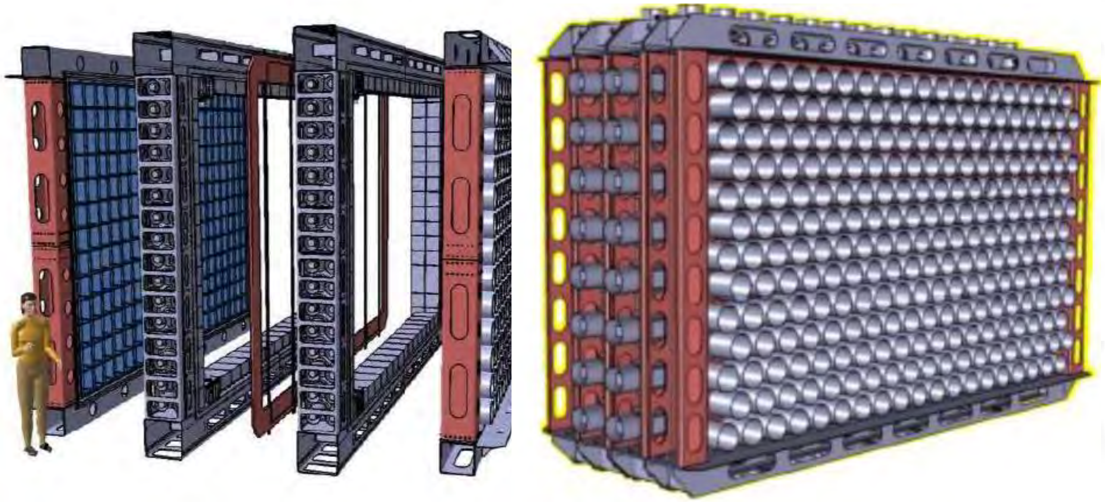
The set of up to 42 elements determined by INAA, RNAA and IPAA in meteorites helps in their classification

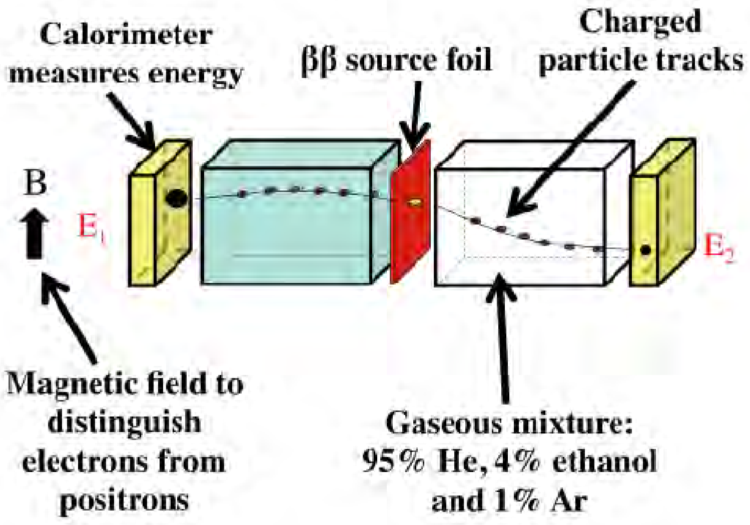
Meteorite/Fall or Found	Classification
Morávka/6 May 2000	H5 chondrite
Jesenice/9 April 2009	L6 chondrite
Rumanová/1994	H5 chondrite
Uhrovec/2012	L6 chondrite
Vel’ké Borové/1895	L5 to L4 chondrite
Košice/ 28 February 2010	H5 chondrite
Chelyabinsk/15 February 2013	LL5 chondrite

Material science/Basic physical problems

Search for neutrinoless double beta-decay

Determination of long-lived radionuclides ^{232}Th and ^{238}U in materials for **SuperNEMO experiment** related to background constraints for neutrinoless double beta-decay searches. Underground installations require detectors made of radioactively pure materials with **minimum contamination by natural radionuclides**.





Demonstrator: The 7-kg ^{82}Se source foil is located in the centre, tracking chambers and calorimeters are on both side of the foil.

Need for ultra low-level determination of ^{232}Th and ^{238}U in:

- **isotope source (^{82}Se)**
- **copper**

RNAA for long-lived natural radionuclides

^{232}Th ($t_{1/2}$= 1.40x10^{10} y), **^{238}U** ($t_{1/2}$= 4.47x10^{9} y)

$$A = \lambda N = \frac{\ln 2}{T_{1/2}} N$$

Decay counting

$$A_1 = N_1 \lambda_1 \gamma_1 \varepsilon_1$$

Atom counting by NAA ($^{x}A_1(n,\gamma)^{(x+1)}A_2$)

$$A_2 = N_1 \gamma_2 \varepsilon_2 SDC(G_{th}\Phi_0\sigma_0 + G_e\Phi_e I_0(\alpha))$$

Advantage factor AF= $\dfrac{\gamma_2\varepsilon_2}{\lambda_1\gamma_1\varepsilon_1}(\Phi_{th}\sigma_0 + \Phi_{epi} I_0)(1 - e^{-\lambda_2 t_i})e^{-\lambda_2 t_d}(\dfrac{1 - e^{-\lambda_2 t_c}}{\lambda_2 t_c})$

AFs for NAA of some long-lived radionuclides

Nuclide pair	$t_{1/2}(1)$	AF
$^{238}U/^{239}U$	4.468×10^{9} y	7.0×10^{6}
$^{238}U/^{239}Np$	4.468×10^{9} y	8.0×10^{5}
$^{232}Th/^{233}Pa$	1.40×10^{10} y	4.0×10^{5}
$^{230}Th/^{231}Th$	7.54×10^{4} y	27
$^{237}Np/^{238}Np$	2.144×10^{6} y	640
$^{231}Pa/^{232}Pa$	3.276×10^{4} y	106

$\Phi_{th}=10^{13}$ cm^{-2}, s^{-1}, $\Phi_{epi}=5\text{x}10^{11}$ cm^{-2} s^{-1}
$\varepsilon_1(\alpha)=25$ %, $\varepsilon_2(\gamma)=5$ mL fractions in well-type HPGe detector

A.R. Byrne, L. Benedik, Czech J. Phys. 49, S1 (1999) 263-270

Detection limits of ^{232}Th and ^{238}U

Nuclide	Half-life	Detection limit, mBq		
		Radiometric[a]	ICP-MS[b]	RNAA[c]
^{232}Th	1.40E+10 y	0.1	2×10^{-3}	8×10^{-5} (for Cu) 4×10^{-4} (for ^{82}Se)
^{238}U	4.468E+9 y	0.1	2×10^{-3}	2×10^{-4} (for Cu) 4×10^{-4} (for ^{82}Se)

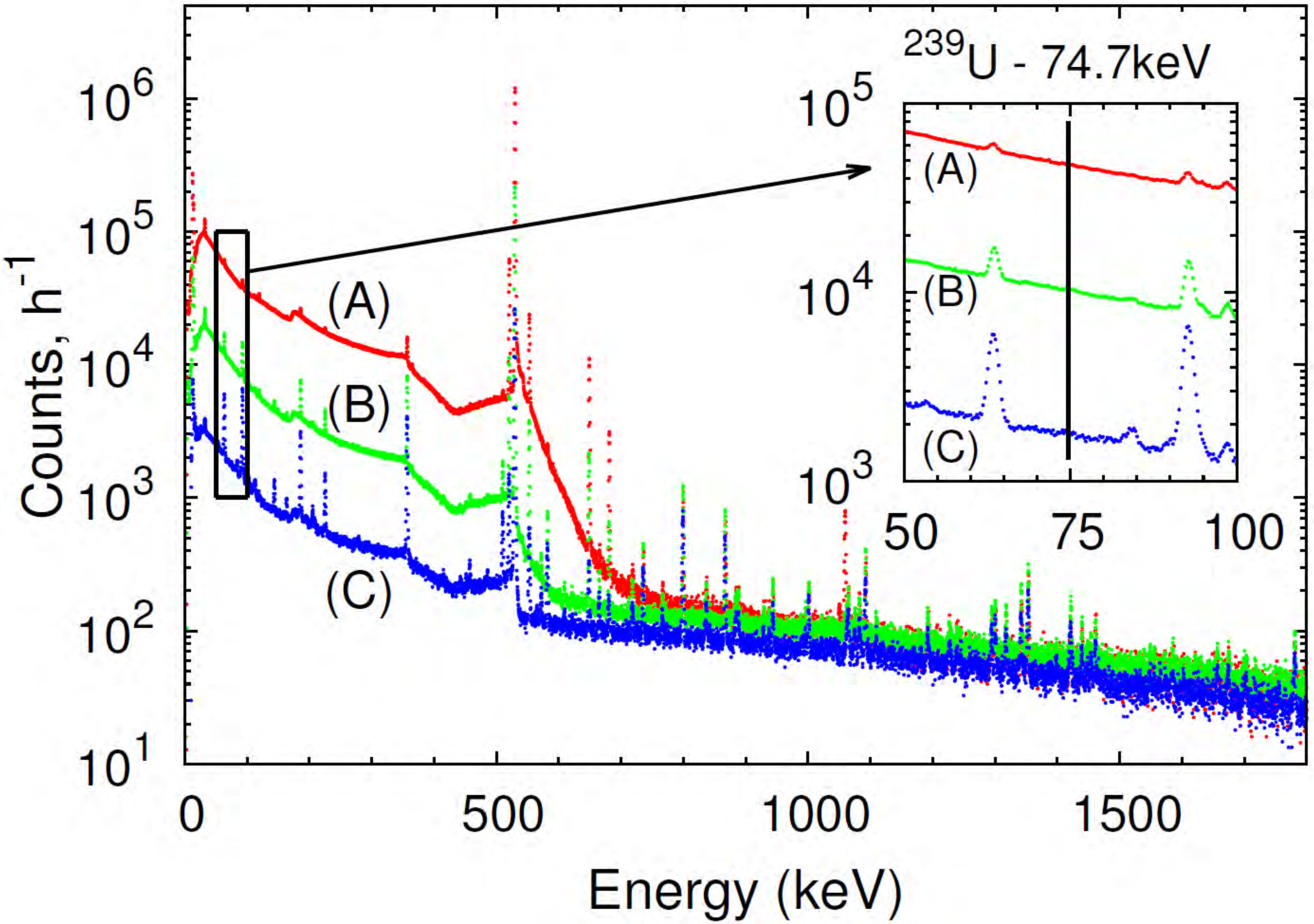
[a] – α-spectrometry

[b] – 3 mL of solution used for measurement

[c] - U: Φ_{th}=3×10^{13} cm^{-2} s^{-1}, t_i=1 min, t_d=30-35 min, t_c= 1 h, 150 cm^3 well-type HPGe detector
Th: Φ_{th}=4×10^{13} cm^{-2} s^{-1}, t_i=20 h, t_d=35 d, t_c=4 h, 150 cm^3 well-type HPGe detector

[a,b] – X. Hou et al., J. Radioanal. Nucl. Chem., 2016, DOI 10.1007/s/10967-016-4741-5

Determination of U via ^{239}U by RNAA



RNAA results for ^{232}Th and ^{238}U

Matrix	^{232}Th (μBq/g)	^{238}U (μBq/g)
Cu	1.6 ± 0.2	< 1.6
^{82}Se	7.7 ± 1.2	< 12

Radionuclidic impurities in RNAA fractions

Matrix	^{233}Pa	^{239}U
Cu	^{59}Fe, ^{124}Sb	^{64}Cu, ^{66}Cu
^{82}Se	^{75}Se ^{130}Te(n, γ)^{131}Te ($T_{1/2}$=1.26 d) → **^{131}I ($T_{1/2}$=8.04 d, E_γ=364.5 keV)**	^{82}Se(n,γ)^{83}Se ($T_{1/2}$=22.3 min)→ **^{83}Br($T_{1/2}$=2.4 h, E_γ=529.6 keV)**

J. Kučera, J. Kameník, P.P. Povinec, J. Radioanal. Nucl. Chem., 311 (2017) 1299–1307

Thank you for your attention!